

Diurnal and Seasonal Variation of BTX in Ambient air of One Urban Site in Carmen City, Campeche, Mexico

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ABSTRACT

BTX (benzene, ethylbenzene, toluene and p-xylene) and meteorological parameters were measured in ambient air of an urban site located in Carmen City, Campeche, Mexico. A total of 412 samples were collected for BTX and analyzed by Gas Chromatography with Flame Ionization Detection (GC-FID). Meteorological parameters were measured by a portable station. A marked diurnal variation was found for all measured BTX. The highest concentrations occurred during midday (13:00-14:30 h). A clear seasonal pattern was observed too for all compounds registering their highest levels during summer sampling period. Mean concentrations for benzene, ethylbenzene, toluene and p-xylene were: 5.42, 3.97, 11.24 and 8.32 ppbv, respectively. BTX abundance showed the following order: toluene > p-xylene > benzene > ethylbenzene. BTX maximum concentrations were found when winds blowed from E and NE. Important oil industry sources and avenues are located at these directions. These sources could contribute to the levels of BTX in this site.

Keywords: BTX; Carbonyls; Volatile Organic Compounds; Air Pollution; Carmen City; Mexico

1. Introduction

Reductions in air pollutants emissions have been made from many sources over the world. In spite of volatile organic compounds (VOCs) and their effects on public health is still a concern and an unsolved problem in many urban cities. Spatial and temporal concentrations patterns outdoors of VOCs are complex; they are originated from different sources, and they are reactive compounds. All of these characteristics make them a difficult challenge for environmental policy makers [1]. In urban areas, these compounds are important atmospheric pollutants because of such detrimental effects as their contribution to stratospheric ozone depletion, tropospheric photochemical ozone formation, toxic and carcinogenic human health effects, and enhancement of the global greenhouse effect [2,3].

Benzene and its alkyl derivatives belong to the VOCs group commonly named as BTX and they are especially harmful for human health, because of their toxic, mutagenic carcinogenic properties [4].

In order to develop efficient air pollution abatement strategies for BTX, it is important to determine their concentrations and to identify their sources and behaviors in ambient air of urban areas [5-7].

Measurements of BTX in some urban areas of Mexico have been carried out, but most of them have been focused to big cities like the Metropolitan Areas of Mexico City and Monterrey [8-11].

At the southeast of the country is located Carmen City, which has experimented an accelerated development since 1996 due to it is an urban area located near from the most important oil offshore platforms in Mexico. In addition, the most important source gas recompression station is located at 25 km from this city. Demographic growth and floating population working for oil industry have been increased in this island last 15 years. For all these reasons its inhabitants are starting to face problems of air quality every day. The objective of this research was to determine the atmospheric concentrations of some BTX (benzene, toluene, ethylbenzene and p-xylene) at one site located in Carmen City, Campeche, Mexico during three seasons of 2012 (spring, summer and autumn) and to try to infer their probable sources from a

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meteorological analysis.

2. Methodology

2.1. Site Description

The study was conducted in a site located within the Botanical Garden of the Autonomous University of Carmen (Lat. 18°38'36"N, Long. 91°49'51"W, elev. 2 masl) on the southeast edge of Carmen Island in Campeche, Mexico (**Figure 1**). According to the Köeppen classification modified by Garcia [12], the climate is sub-humid warm with summer rains. The average annual rainfall is 1393.1 mm and the average annual temperature is 27.2°C. Winds prevail from NE and SE all year around and blow at an average speed of 1.4 ms⁻¹ [12]. This site is within the Natural Protected Area named "Términos Lagoon".

2.2. Sampling Method

412 samples were collected from June 1 to December 21, 2012, considering three climatic seasons: spring, summer and autumn 2012. Benzene, ethyl benzene, toluene and p-xylene were measured in ambient air.

Samples of air were collected within glass tubes containing 226-01 Anasorb CSC (SKC): length 70 mm; inner diameter 4.0 mm; outer diameter 6 mm packed in the first section with 100 mg of activated carbon and 500 mg in the second one, separated by a glass wool section (Method INSHT MTA/MA-030/A92) [13,24]. 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 (day, midday and afternoon). Sampling was carried out using a Universal XR pump model PCXR4 (SKC), at three sampling periods: B1 (from 08:00 to 09:30 h), B2 (from 13:00 to 14:30 h) and B3 (from 16:00

to 17:30 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 intended for use in the main study. This procedure included determining appropriate sampling times. Samplers tubes were protected from bad 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 along with samplers to the field and stored in the laboratory during the sampling period. Samples were analyzed within three weeks after sample collection at the Environmental Sciences Laboratory in the Autonomous University of Carmen City (UNACAR).

2.3. Analytical Method

Samples were extracted with 1 ml of CS₂ for each section of the samplers tubes, shaking during 30 s to assure a maximum desorption. Extracted samples were analyzed using a TRACE GC Ultra gas chromatograph (Thermo-scientific) and one flame ionization detector (FID; Thermo-scientific Technologies, Inc) (Method INSHT MTA/MA-030/A92) [13,24]. 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, which was then increased at a rate of 5°C/min up to 100°C, and was 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 [14]. Four BTX that included benzene, p-xylene, ethyl benzene, and toluene, were investigated. Five-point calibration was performed using 99.98% Sigma-Aldrich analytical reagents at a concentration of 2 ppm for each BTX.

The established calibration curves for the four investigated BTX were found to have R-square values of 0.999. Method detection limit (MDL) for each compound was 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; 0.025; μg·m⁻³, respectively. The amount of BTX in blank samples was below the limit of detection (LOD) for all studied compounds.

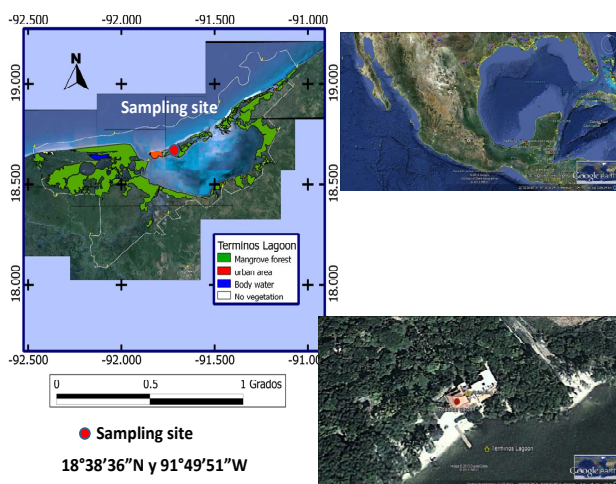


Figure 1. Location of the sampling site.

2.4. Monitoring of Meteorological Parameters

Wind conditions (direction and speed), relative humidity, temperature, and barometric pressure were monitored from June 1 to December 21, 2012 (spring, summer and autumn). A portable meteorological station Davis Vantage Pro II model was used in order to measure the meteorological parameters. Wind roses were constructed for each day using the software WRPLOT (from Lakes Environmental) [15]. 24 hr back air masses trajectories were calculated for the studied period using HYSPLIT model from the NOAA (National Oceanic Administration Agency, USA) in order to identify the probable origin of the air masses.

3. Results

3.1. Diurnal and Seasonal Variation of BTX

Benzene (Bz) and Toluene (T) showed the same diurnal pattern registering the highest concentrations during the midday (B2) decreasing during the afternoon (B3) and showing the lowest values during the mornings (B1). Ethylbenzene (EBz) and p-Xylene (p-X) had the same behavior along the day with the highest levels during the midday (B2) decreasing during the mornings (B1) and registering their lowest concentrations during the afternoons (B3) as it can be observed in **Figure 2**.

In **Table 1** is shown the comparison of the results of this study with ambient air concentrations found in other sites around the world. It can be observed that BTX levels found in this study are comparable to those reported for cities like Mexico [8,22] and Hong Kong [18] and lower than those measured in Rome [16].

All measured BTX showed the same seasonal variation, registering the highest concentrations during summer, decreasing during spring and showing their lowest

values during autumn, as it can be observed in **Figure 3**.

During summer 2012, all measured BTX showed their highest concentrations during the midday (B2) decreasing during the mornings and registering their lowest levels during the afternoon (**Figure 4**). During summer 2012, all the studied compounds registered their highest values of concentration during the afternoon (B3), decreasing during midday (B2) and showing their lowest values during the mornings (B1) as it can be observed in **Figure 5**. In autumn 2012, Bz, EBz and p-X showed their highest

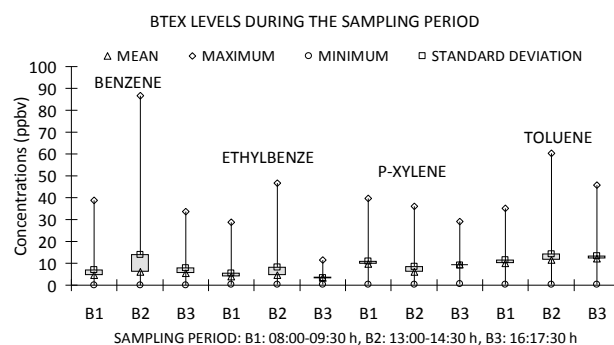


Figure 2. BTX diurnal variation in this study.

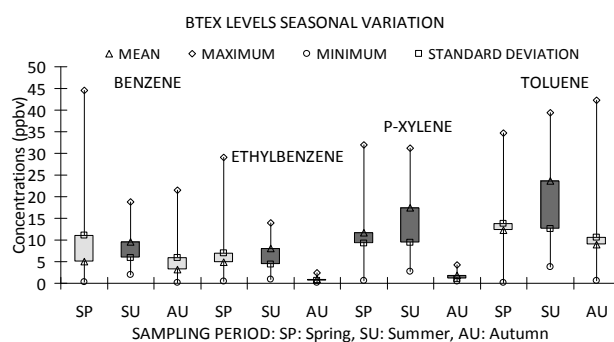


Figure 3. BTX Seasonal variation during this study.

Table 1. Comparison of atmospheric concentrations of BTX (ppbv) found in this study with data of other studies around the world.

Location	Bz	T	EBz	p-X
Kocaely, Turkey (urban) [1]	0.7077	9.4277	2.2395	8.4919
Rome [16]	11.1167	26.4697	4.0552	12.460
Belgium (busy road) [17]	0.9614	1.9832	0.341	0.8959
Hong Kong (industrial) [18]	4.7191	36.9966	5.6864	6.4213
Izmir (urban) [19]	1.0365	4.0859	0.8409	1.7274
Fuji (industrial) [20]	0.645	3.7170	0.3502	0.4215
Pamplona (urban) [21]	0.8893	3.5204	0.4954	0.7785
Mexico City (urban) [8,22]	6.5447 [8]	31.647 [22]	4.3086 [8]	15.6848 [8]
This study (urban)	5.4233	11.2383	3.9722	8.3284

Bz: Benzene; T: Toluene; EBz: Ethylbenzene; p-X: p-Xylene.

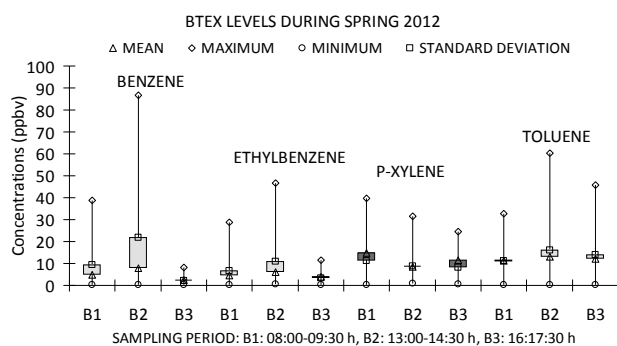


Figure 4. Diurnal variation for BTX during spring 2012.

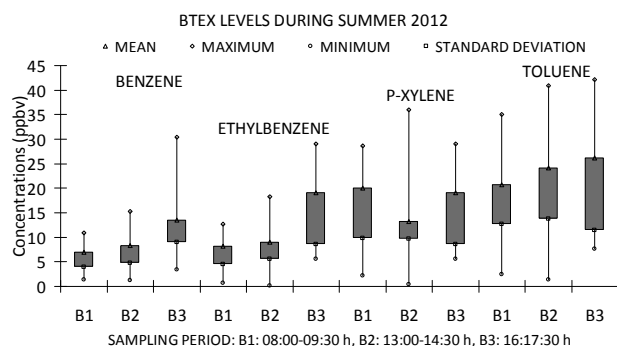


Figure 5. Diurnal variation for BTX during summer 2012.

values during the afternoons (B3), decreasing during the mornings (B1) and showing their lowest levels during the midday (B2) as it can be observed in Figure 6.

3.2. Toluene to Benzene Ratio (T/Bz Ratio)

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 exhausts. The toluene content of gasoline and motor vehicle exhaust is 3 - 4 times higher than Bz content [16]. A value of around 2 - 3 is characteristic of vehicular emissions in many urban areas worldwide [15,22,23]. The range for Carmen City in this study was between 2.45 and 2.8 being higher during autumn and lower for summer. Mean T/Bz ratios found were: 2.4526, 2.4762 and 2.8036 for spring, summer and autumn, respectively. These values are agree with typical values of vehicular emissions (2 - 3) reported for other urban areas, suggesting that this site was under the influence of mobile sources.

3.3. Pearson's Correlation

Table 2 shows the Pearson's correlation coefficient matrix for the mean concentrations of BTX. A significant correlation between Bz and T indicates that they were possibly originated from vehicular emissions. EBz and p-X had a good correlation, indicating that both com-

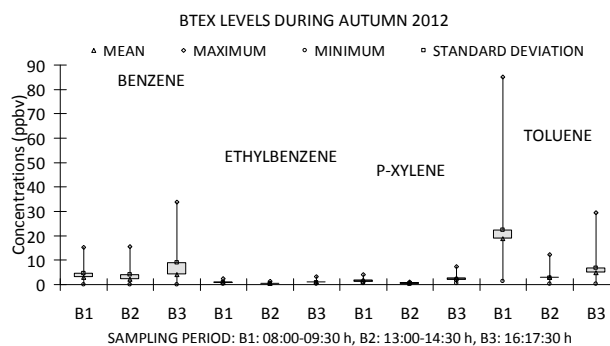


Figure 6. Diurnal variation for BTX during autumn 2012.

Table 2. Pearson's correlation coefficient matrix for studied BTX.

	Bz	Ebz	p-X	T
Bz	1	0.2193	0.6646	0.6648
Ebz	0.2193	1	0.7983	0.01682
p-X	0.6646	0.7983	1	0.1085
T	0.6648	0.01682	0.1085	1

Bz: Benzene; EBz: Ethylbenzene; p-X: p-Xylene; T: Toluene.

pounds probably had their origin from gasoline vehicles, petroleum plants and gasoline stations. T had the lowest coefficients in this work with EBz and p-X, suggesting that T had other additional sources. According to the USEPA SPECIATE [22] data base, T, EBz and p-X are consistent with sources profiles given for "gasoline vehicle emissions".

3.4. Meteorological Analysis

BTX concentrations were correlated with meteorological parameter at surface level and 24 h backward air masses trajectories were calculated for maximum concentrations events using the HYSPLIT model from NOAA at 500, 700 and 1000 masl. The backward trajectories analysis helps to determine the origin of air masses and probable location of sources for the measured pollutants.

During spring season, the highest concentrations for all BTX were registered when wind blew from E, where an avenue with high vehicular traffic is located being highest the BTX levels during the midday sampling (B2) as it can be observed in Figures 7 and 10. In summer (Figures 8 and 11), Bz and EBz showed the highest values when wind blew from E and NE. The most important offshore platforms facilities of Mexico are located at NE and N of the sampling site. p-X showed their highest concentrations during summer when wind prevailed blowing from NE (B1), SW (B2) and SE (B3). A sour gas recompression station is located at the SW of the sampling site. Local sources related to one building

construction are located at SE of the sampling site.

During autumn (**Figures 9 and 12**) mornings the highest levels for all BTX were registered when wind blew from NE. During the midday, the highest concentrations for EBz and p-X were observed when wind blew from NE, from SE and from N for Bz and T, respectively. In the autumn afternoons, Bz and T showed their highest levels when wind blew from N, and from SW and W for p-X and EBz, respectively. The principal activities of this urban area are located at W from the sampling period.

3.5. Principal Component Analysis (PCA)

A principal component analysis was carried out in order to study patterns in a multivariate data set for the three climatic seasons (spring, summer and autumn) and for the three sampling periods (08:00-09:30 h; 13:00-14:30 h; and 16:00-17:30 h). The PCA analysis was applied for the BTEX concentrations (benzene, toluene, ethylbenzene and p-xylene), meteorological parameters (temperature: T, barometric pressure: P, relative humidity: RH, wind direction: WD, and wind speed: WS). **Figures 11-19** show the PCA loadings obtained for the morning,

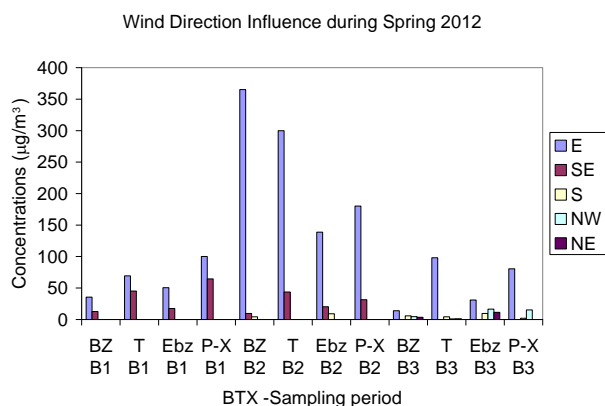


Figure 7. Wind direction influence on BTX concentrations during spring 2012.

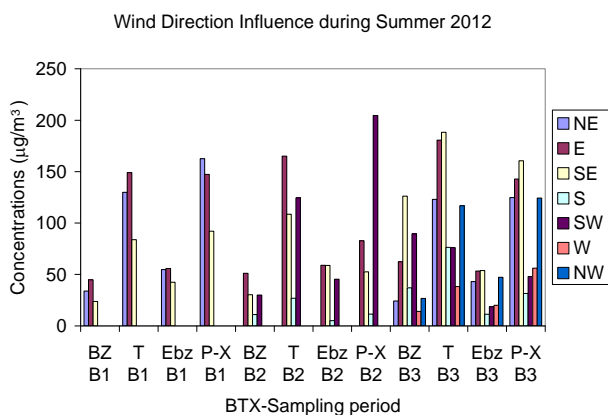


Figure 8. Wind direction influence on BTX concentrations during summer 2012.

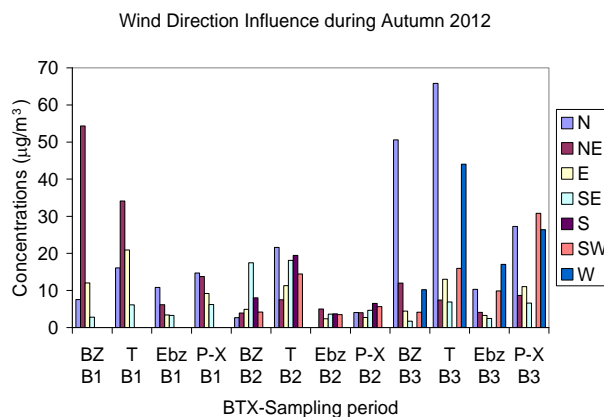


Figure 9. Wind direction influence on BTX concentrations during autumn 2012.

midday and afternoon sampling periods (B1, B2 and B3, respectively) for the three climatic periods (spring, summer and autumn).

In **Figure 13** it can be observed that there were some associations among compounds: a positive correlation among p-Xylene (p-X), Benzene (B), Ethylbenzene (Ebz) and Toluene (T), suggesting that these compounds had probably sources in common during the mornings of spring season (B1). BTX concentrations showed a negative correlation with wind speed indicating that as wind speed increases, BTX concentrations decrease due to a dilution effect. BTX-temperature (T) correlation was not important, suggesting that these compounds did not have their origin in evaporative emissions from oil service stations.

In **Figure 14** it can be observed that there were some associations among compounds during the mornings of summer season: BTX concentrations had a negative correlation with wind speed; it suggests that as wind speed increases, BTX concentrations decrease, due to a dilution effect. Relative humidity (RH) showed a negative correlation with BTX suggesting that this condition contributed to remove the atmospheric pollutants by wet deposition or condensation [25,26]. Benzene (B) and p-Xylene (p-X) showed positive correlations with temperature (T) indicating that at least partially, these compounds had their origin in evaporative emissions. In general, all measured BTX showed high positive correlations between each other, suggesting that they could have the same emissions sources (**Figure 14**).

In **Figure 15** it can be observed that for the autumn season during the morning sampling period (B1), BTX showed good correlations between each other, indicating that they probably had sources in common. BTX concentrations had a negative correlation with wind speed; it suggests that as wind speed increases, BTX concentrations decrease, due to a dilution effect. Relative humidity (RH) showed a negative correlation with BTX



Figure 10. Air mass backward trajectory for a maximum BTX high concentrations event during spring (June 17, 2012).

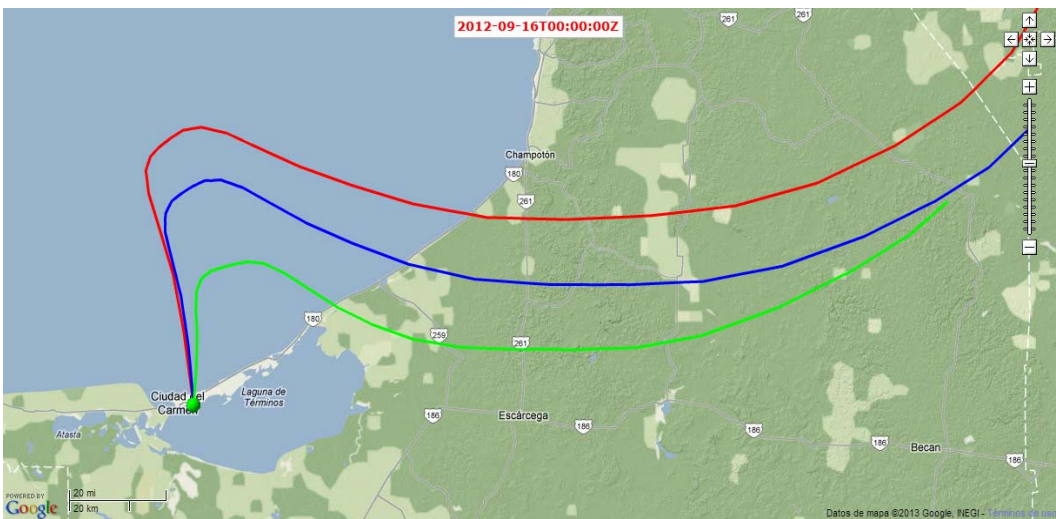


Figure 11. Air mass backward trajectory for a maximum BTX high concentrations event during summer (September 16, 2012).

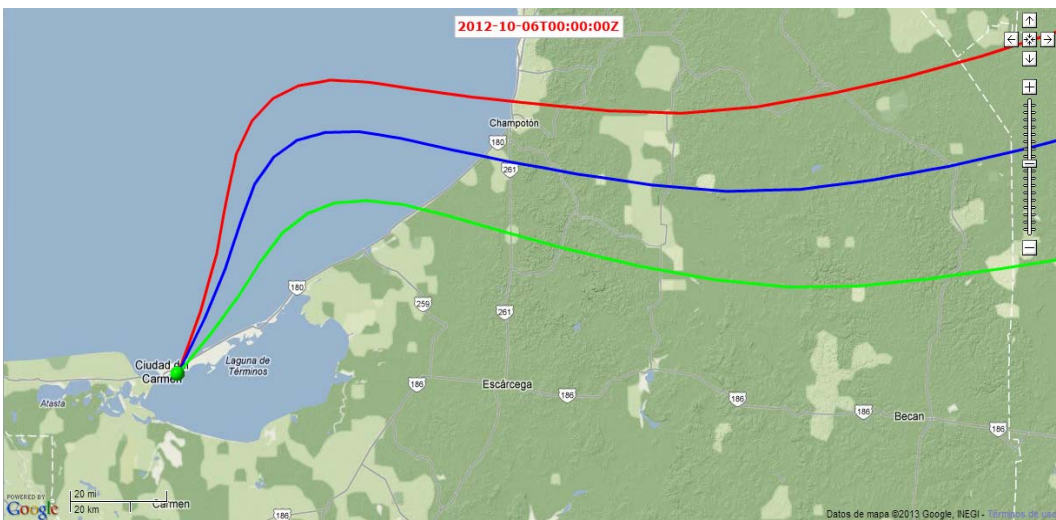


Figure 12. Air mass backward trajectory for a maximum BTX high concentrations event during autumn (October 6, 2012).

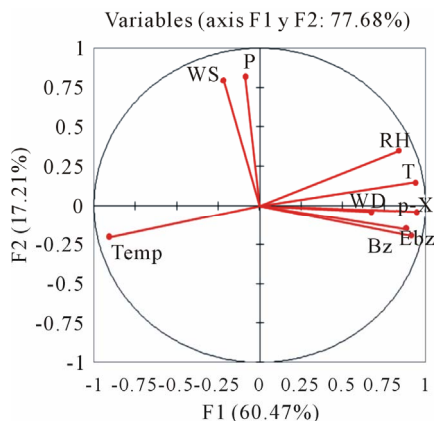


Figure 13. PCA for spring season during the morning sampling period B1: 08:00-09:30 h.

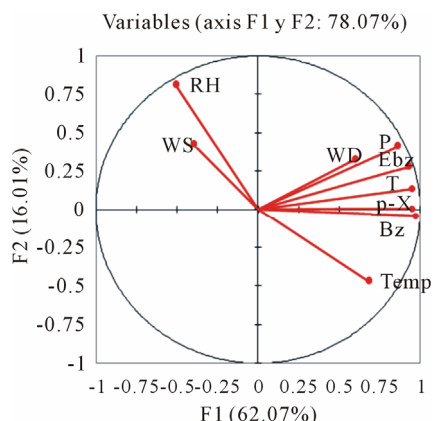


Figure 14. PCA for summer season during the morning sampling period B1: 08:00-09:30 h.

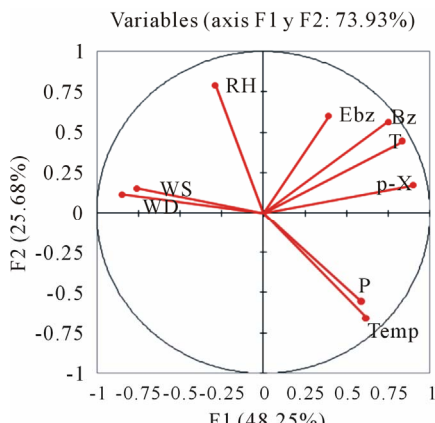


Figure 15. PCA for autumn season during the morning sampling period B1: 08:00-09:30 h.

suggesting that this condition contributed to remove the atmospheric pollutants by wet deposition or condensation [25,26].

In Figure 16, it can be observed that BTX had positive correlations between each other for spring during the

midday sampling period (B2: 13:00-14:30 h), suggesting that these compounds had probably sources in common. The dilution effect was evident due to a negative correlation among wind speed and all BTX was observed. A negative correlation among temperature and BTX was observed indicating that all measured BTX had different sources than those related to evaporative emissions.

In summer season during the midday sampling period (B2: 13:00-14:30 h) (Figure 17), BTX correlated positively with temperature indicating that probably these compounds had their origin in evaporative emissions. BTX had negative correlation with wind speed (dilution effect) and relative humidity, suggesting that high water vapor concentrations found in air masses removes partially the air pollutants by wet deposition or condensation.

In Figure 18 it can be observed that during autumn season for the midday sampling period (B2: 13:00-14:30 h), BTX had mixed sources. Bz-T and Ebz-p-X showed good correlations indicating that these pairs of compounds probably had their origin in common sources.

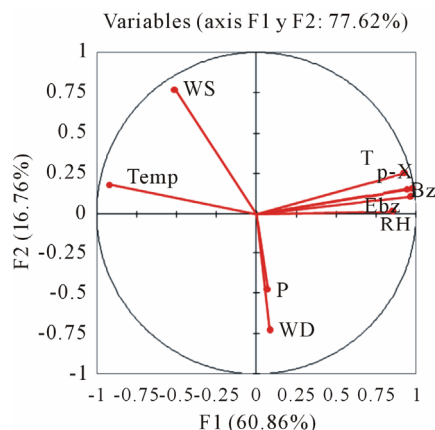


Figure 16. PCA for spring season during the midday sampling period B2: 13:00-14:30 h.

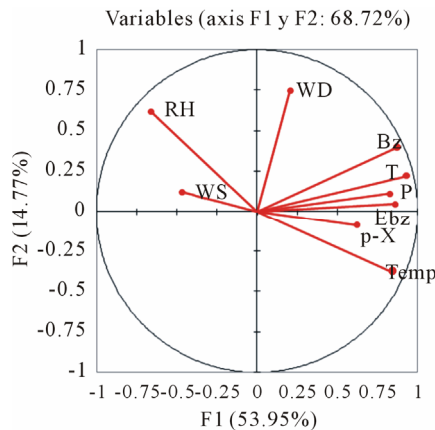


Figure 17. PCA for summer season during the midday sampling period B2: 13:00-14:30 h.

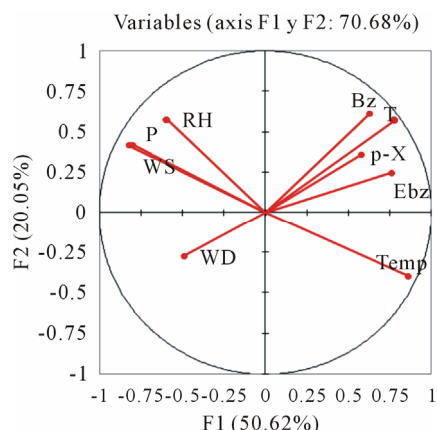


Figure 18. PCA for autumn season during the midday sampling period B2: 13:00-14:30 h.

Ebz and p-X had a moderate positive correlation with temperature, suggesting that at least partially these compounds could be originated from evaporative emissions. All measured parameters showed for this period a negative correlation with wind speed (WS).

During spring season for the afternoon sampling period (B3), Bz-Ebz-p-X, and T-p-X had good correlations (**Figure 19**) indicating that these pair of compounds had probably common sources. BTX did not have a good correlation with temperature indicating that the contribution of evaporative sources was negligible.

In summer during the afternoon sampling period: B3 (**Figure 20**), T had high correlations with Ebz and p-X, suggesting that they had probably common sources. T had a moderate positive correlation with temperature, suggesting that this compound probably had its origin in evaporative emissions. All measured parameters had a negative correlation with wind speed (WS) and relative humidity (RH) indicating the influence of dilution and wet deposition/condensation processes.

In **Figure 21** it can be observed that during the autumn season for the afternoon sampling period (B3: 16:00-17:30 h) BTX correlated positively, it indicates that these compounds probably had their origin in the same sources. All measures BTX showed a negative correlation with wind speed and temperature for this period.

4. Conclusions

BTX concentrations in this work showed values comparable to those reported for cities like Mexico and Hong Kong. There was a clear diurnal pattern in the BTX concentrations, showing the highest levels during midday (B2). BTX seasonal variation levels had a clear behavior too, showing the highest concentrations during summer, decreasing during spring and registering the lowest values during autumn. It is more probable than during summer that BTX could be originated from photochemical

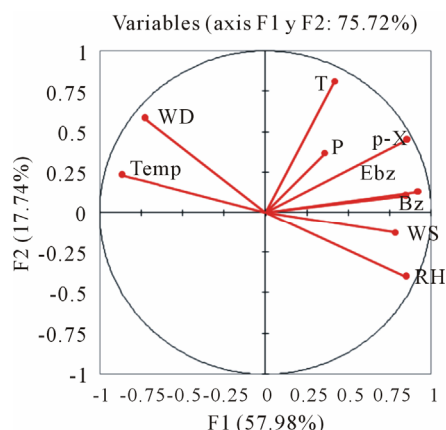


Figure 19. PCA for spring season during the afternoon sampling period B3: 16:00-17:30 h.

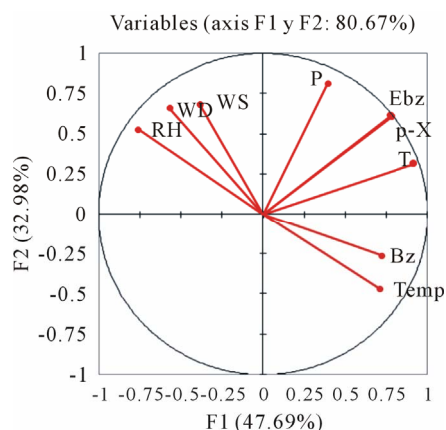


Figure 20. PCA for summer season during the afternoon sampling period B3: 16:00-17:30 h.

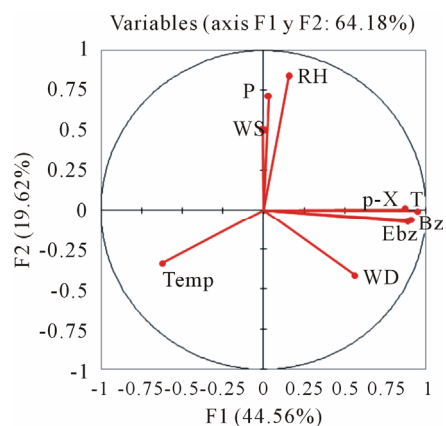


Figure 21. PCA for autumn season during the afternoon sampling period B3: 16:00-17:30 h.

reactions and evaporative emissions from gas and gasoline storage stations. BTX in this city had mixed sources during the sampling period. According to T/Bz ratios and the meteorological analysis, it was clearly under the influence of vehicular sources circulating in avenues located

at E of the sampling site. On the other hand, the wind direction study showed that this site was under the influence of sources related to oil industry located at N, NE and SW of the sampling site. We can therefore conclude that air quality in this city is already a problem to concern and it is necessary to establish air pollutants control strategies. However, it is necessary to carry out intensive monitoring campaigns in this site including criteria pollutants (Ozone, NO_x, NO₂, CO, SO₂) monitoring in order to get more information about the origin of BTX.

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