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Atmospheric levels of BTEX and criteria pollutants (CO, NO₂, O₃ and PM₂.₅) in an urban site located in North Baja California, Mexico: source implications and health risk

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Abstract: - Atmospheric levels of BTEX (benzene, toluene, ethylbenzene, p-xylene) and criteria air pollutants (O_3, NO_2, CO, $PM_{2.5}$) were measured in urban air of one site located in North Baja California, Mexico during winter 2017. Diurnal variation was studied collecting samples at 1.5 h time intervals in different sampling periods: 07:30-09:00 h, 12:00-13:30 h, 15:00-16:30 h, and 18:00-19:30 h and then analyzed by gas chromatography with flame ionization detection. The highest concentrations of BTEX occurred during the morning (07:30-09:00h) and afternoon (18:00-19:30h) sampling periods. Mean concentrations of BTEX were 32.40 μg/m³ for benzene; 13.28 μg/m³ for toluene; 17.16 μg/m³ for p-xylene; and 7.02 μg/m³ for ethylbenzene. BTEX concentration ratios and meteorological analysis indicated that these compounds were influenced by local and fresh emissions. Pearson correlation and Principal Component Analysis were performed in order to correlate BTEX concentrations with criteria air pollutants and meteorological parameters to infer their possible sources. A health risk assessment showed that local exposure to benzene exceeded 1 x 10⁻⁶ for integrated lifetime cancer risk. It suggests that people living in Tijuana, Baja California, Mexico may have a probable risk of suffering cancer in their lifetime. These results indicate that changes in the existing environmental policies should therefore be applied to improve air quality in this region.

Key-Words: - BTEX, Air Pollution, Tijuana, Health Risk Assessment, VOCs, Baja California, Mexico

1 Introduction

Volatile organic compounds (VOCs) play an important role in the production of tropospheric ozone and contribute to secondary organic aerosol (SOA) formation by their reaction with NOx in the presence of solar radiation [1]. Therefore, it is important to infer their sources and distribution in the urban atmospheres. VOCs are widespread atmospheric pollutants, having both, natural and anthropogenic sources including vehicular emissions, combustion processes and other industrial activities. VOCs in urban areas include a variety of pollutants including BTEX compounds (benzene and its alkyl derivatives: toluene, ethylbenzene, and xylenes) [2]. These pollutants have adverse effects on human health and may reduce the quality of life due to some of them are recognized as carcinogenic compounds [3].

BTEX have been classified as air toxics, carcinogenic and mutagenic at concentration levels typically found in urban environment. Benzene has been recognized as carcinogenic and have adverse effects on human health as anemia, acute and chronic non-lymphocytic and myeloid leukemia [4]. In spite of BTEX represent a danger to the health, currently, in Mexico there is not a standard that regulates their maximum concentrations levels in order to protect the health of population and existing air quality monitoring networks do not measure these pollutants (excepting Mexico City air monitoring network). In addition, there are not enough available data of BTEX atmospheric levels in Mexico (most of the studies have been focused on cities located at the center of the country). In the North of Mexico, numerous facilities of multinational companies and multiple maquila companies are located. In this region, one of

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the most developed cities is Tijuana, Baja California. This city is the most populated in the state of Baja California, and is the fifth most populous city in the country with 1,773,558 inhabitants, being part of the sixth metropolitan area of Mexico together with the cities of Rosarito and Tecate. In addition, it is the most visited border city in the world (more than fifty million people cross this border into the United States each year).

Consequently, Tijuana's air quality has been degraded during the last years. Emissions related to traffic and industrial activities are a major problem for quality ambient air in this city causing an increase in air pollutants concentrations.

2 Problem Formulation

According to the industrial and economic importance of Tijuana city, it is crucial to know the current state of air quality in this city. Therefore, this study is focused on 1) to evaluate diurnal variation of BTEX and criteria pollutants (CO, NO_2, O_3 and $PM_{2.5}$) atmospheric concentrations in an urban site of Tijuana, Baja California, Mexico during winter season 2017; 2) to infer the probable sources and origin for these air pollutants from meteorological and chemometric analysis (air masses trajectory models and principal component analysis); and 3) to assess the health risk that represents the inhalation of air with the found benzene concentrations in this study.

2.1 Study site
The study site is located within the facilities of the air quality monitoring station of the Baja California Norte State known as La Mesa near the Otay gate, the gateway to the United States (Figure 1) at an altitude of 72 masl.

2.2 Sampling and Analytical Method
Samples were collected during January 2017 in order to determine BTEX (benzene, toluene, ethylbenzene and p-xylene) concentrations in ambient air. A controlled flow of air (200 mL min-1) was passed through glass tubes containing active carbon (226-01 Anasorb CSC) using a vaccum pump model PCXR4 SKC [5]. Samples of 1.5 h were collected considering four different sampling periods: during the morning (B1: 07:30 to 09:00 h), midday (B2: 12:00 to 13:30 h), afternoon (B3: 15:00 to 16:30 h) and night (B4: 18:00 to 19:30 h). A total of 24 samples were collected during the study period and then they were extracted with 1 mL of CS_2 and

analyzed using a gas chromatograph (TRACE GC Ultra Thermo Fisher Scientific Technologies, Inc., Waltham, MA, USA) and a flame ionization detector (Thermo Fisher Scientific Technologies, Inc., Waltham, MA, USA) according to the MTA/MA030/A92 Method [6].
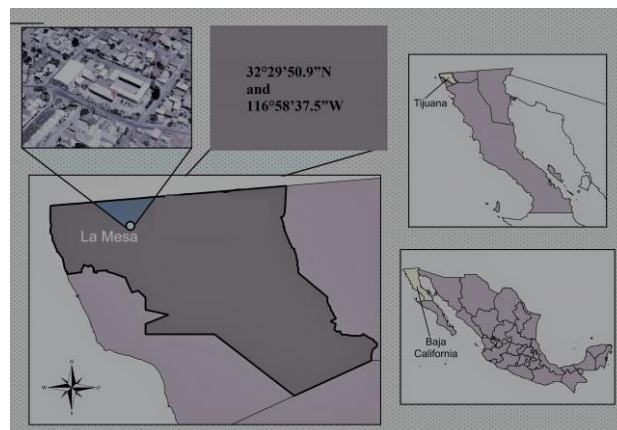

Figure 1. Sampling site location

2.3 Criteria Air Pollutants and Meteorological Parameters Measurements
Meteorological parameters (wind speed and direction, relative humidity, temperature and solar radiation) were measured during the sampling period using a Davis Vantage Pro II portable meteorological station. Criteria Air Pollutants were monitored (O_3, NO_2, CO, and PM2.5) using Thermo Scientific automatic analyzers. These data were obtained from an air quality station of the Integrated Air Quality Monitoring System of Tijuana located in the sampling site within the facilities of the Urban Development and Environment Secretary of the State of Baja California Norte, Mexico.

2.4 Statistical Analysis
Data were analyzed using XLSTAT software version 2017.1 [7]. The non-parametric Friedman's test ($\alpha = 0.05$) was used to evaluate differences among sampling periods of the measured air pollutant concentrations. Pearson's correlation was used to identify relations among BTEX and criteria air pollutants and meteorological variables. A Principal Component Analysis (PCA) was used for explaining the variance and for uncovering the structure of the data set [8].

2.5 Health Risk Assessment
The US Environmental Protection Agency have established quantitative risk assessments for the cáncer and non-cancer risks of VOCs [9]. To estimate these health risks we used the methodology described

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by Zhang and collaborators [10], considering only inhalation exposure. Daily exposure (E), the hazard quotient for non-hazard risk (HQ), and integrated lifetime cancer risks (ILTCR) were calculated using the following equations:

$$E = (C \times IRa \times DA) / BW \qquad (1)$$

where E is the daily exposure expressed in mg/kg per day of an individual by inhalation, C (mg m^{-3}) is the concentration of the air pollutant, IRa is the inhalation rate for adults (0.83 m3 h^{-1}) [11], DA is the exposure duration of an adult (24 h/day), and BW is the body weight of an adult (65 kg) [12]. The integrated lifetime risk (ILTCR) is then calculated as follows:

$$ILTCR = E \times SF \qquad (2)$$

where SF is the slope factor (kg day/mg) of inhalation unit risk for toxics when the exposure-carcinogenic effect is considered as linear. We used SF values in Table 1, provided by the US EPA [13]. The non-cancer risk (HQ quotient) is calculated by dividing the yearly average daily-received concentration (CY) by the inhalation reference concentrations of the specific air pollutant (RfC values shown in Table 1):

$$HQ = CY / RfC \qquad (3)$$

Table 1. Toxicity Profile for benzene

Pollutant	CAS No	RfC (mg m^{-3})	Inhalation Cancer Slope Factor (SF) (kg day/mg)
Benzene Carcinogenicity: Group A	71,432	3×10^{-2}	2.89×10^{-2}

3 Problem Solution

3.1 Diurnal variation and parametric statistics for BTEX

Diurnal variation and parametric statistics for measured BTEX is shown in Figure 2. Benzene (32.398 µg m^{-3}) was the dominant BTEX followed by p-xylene (17.155 µg m^{-3}), toluene (13.277 µg m^{-3}) and ethylbenzene (7.023 µg m^{-3}). Benzene, toluene and p-xylene showed a clear diurnal trend with their highest mean concentrations during the morning period (B1). Ethylbenzene showed its highest mean values during the nighttime (B4). Higher concentrations in the morning period can be attributed to vehicular emissions during the rush hours. Lower concentrations were found during midday (B2) and afternoon (B3) probably due to a faster removal of volatile organic compounds by

reactions with hydroxyl (•OH) and nitrate (NO$_3$) radicals due to enhanced solar radiation and temperature during these sampling periods.
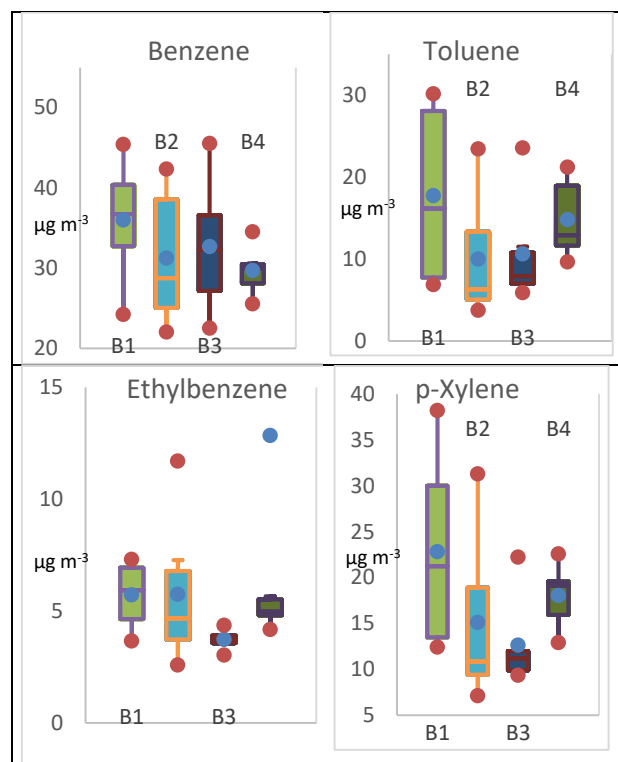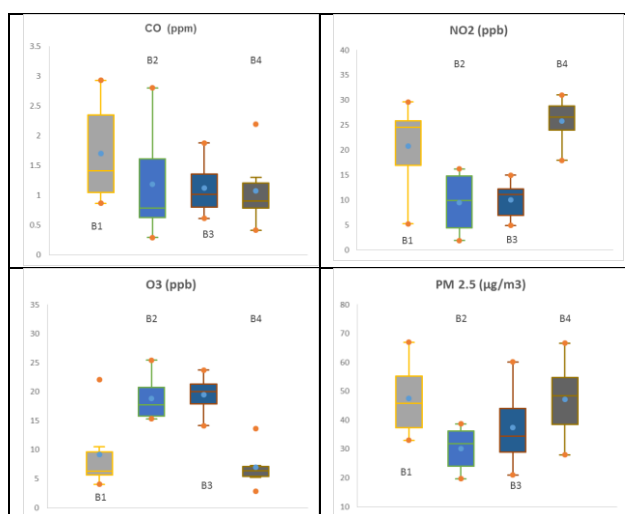

Figure 2. Diurnal variation and parametric statistics for BTEX in the study site. NOTE: Where • represents mean value; • represents maximum and minimum values; the horizontal width of the box has not statistical significance, and is only for better visualization.

3.2 Diurnal variation and parametric statistics for Air Criteria Pollutants

Diurnal variation for measured air criteria pollutants is shown in Figure 3. CO and PM$_{2.5}$ showed the highest mean concentrations during the morning sampling period (B1), whereas, NO$_2$ and O$_3$ showed their highest mean concentrations during the night and the afternoon sampling periods, respectively. Variations in atmospheric stability control the extent to which ground-level pollutants are mixed and diluted. Mixing is greatest during the midday and afternoon causing a decrease in air pollutants concentrations for CO, NO$_2$ and PM2.5. Ozone behaves different showing its higher concentrations during the midday period (B2) when solar radiation and therefore the photochemical activity is high, conditions that drive its production. On the other hand, mixing is limited during the morning (B1) period, particularly during winter, when temperature inversions associated with nocturnal cooling commonly occur causing an increase in air pollutants concentrations. Mean concentrations for CO, NO$_2$,

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O_3 and $PM_{2.5}$ were of 1.27 ppm, 16.53 ppb, 13.60 ppb, and 40.56 µg m^{-3}, respectively.

Figure 3. Diurnal variation for air criteria pollutants monitoring during the study period. NOTE: Where • represents mean value; • represents maximum and minimum values; the horizontal width of the box has not statistical significance, and is only for better visualization.

The only air pollutant that presented exceedances to the air quality Mexican standard (NOM-025-SSA1-2014) [14] was $PM_{2.5}$ (12 exceedances were registered for winter months) (See Figure 4). Diurnal variation for meteorological parameters is shown in Figure 5. Solar radiation and temperature showed their highest mean values during the midday sampling period (B2), whereas, atmospheric pressure and relative humidity showed their highest mean values during the morning sampling period (B1). Wind conditions (direction and speed) were higher during the afternoon sampling period (B3). Mean values for solar radiation, atmospheric pressure, temperature, relative humidity, wind direction and wind speed were of 252.8 W m^{-2}, 1005.92 mmHg, 11.1°C, 53.84%, 240 ° and 13.7 km h^{-1}, respectively.

3.3 Wind Analysis

Dominant winds blowed from WNW and NW. Toluene had higher concentrations with winds blowing from NNE, while, p-xylene showed higher levels of concentration when winds came from NNW and NE. Benzene had higher concentration values when wind blew from WNW and WSW, whereas, higher values of ethylbenzene were found when air masses came from W (See Figure 6). Sources located in these directions could influence the BTEX levels in the study site, highlighting the International Airport "Abelardo Rodriguez" and an industrial zone (Industrial Park "Chilpancingo") located in NNE to be possible sources.
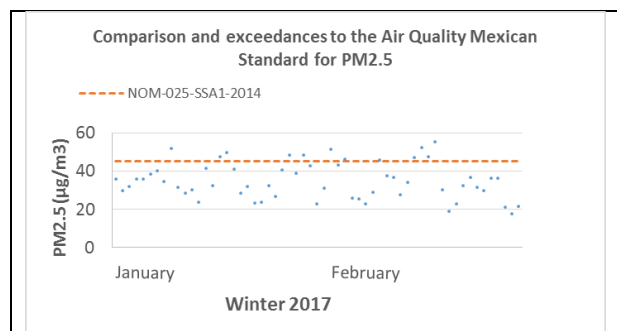
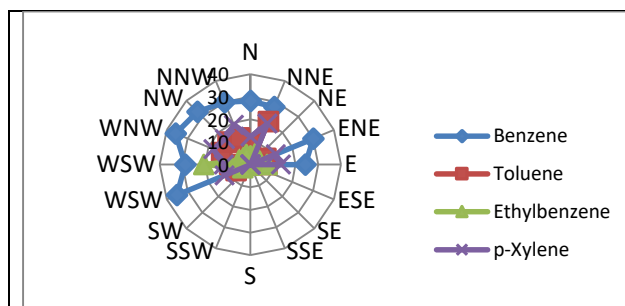**Figure 4.** Comparison and exceedances to the air quality Mexican standard for $PM_{2.5}$.
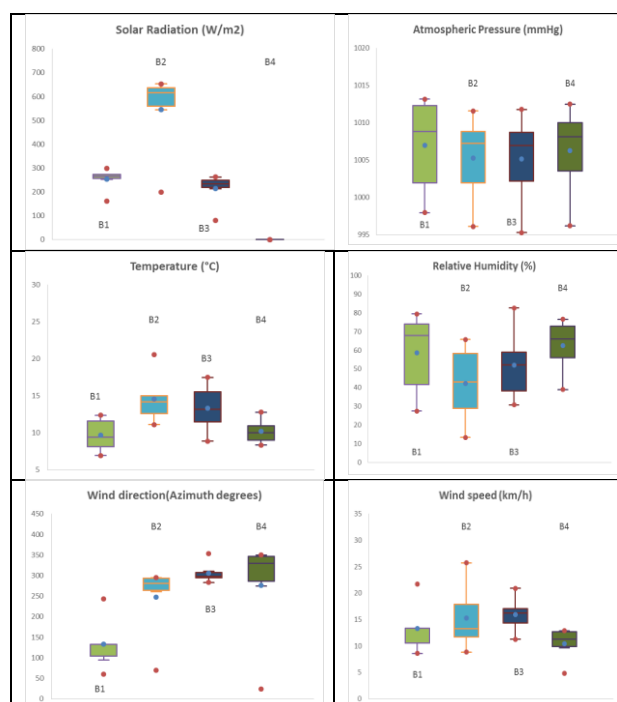
Figure 5. Diurnal variation for meteorological parameters. Where • represents mean value; • represents maximum and minimum values; the horizontal width of the box has not statistical significance, and is only for better visualization.

The Center of Tijuana city is located at WNW and NNW from the study site, whereas, several avenues with high vehicular traffic are located at the WSW from the sampling site (highway Alamar Oriente, Las Torres Boulevard, Otay Gate Boulevard and freeway Gato Bronco). All these sources could contributed to the observed BTEX levels.

3.4 BTEX correlations, ratios, and Principal Component Analysis Results

BTEX concentration ratios are commonly used to infer emission sources and to get information about the photochemical processing of air masses [15]. Toluene to benzene ratio (T/B) is used to determine the relative abundance between traffic and non-traffic sources of BTEX.

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Figure 6. Mean concentrations (µg m⁻³) for BTEX and wind direction diagrams for winter period.

Benzene and toluene are highly correlated with vehicular emissions; however, toluene has other sources such as solvent usage, whereas benzene is not commonly present in solvent content. A low T/B ratio (< 2) indicates a strong influence of vehicular emissions on BTEX concentrations at a given site [16-18], while values greater than two may indicate that BTEX levels are associated with sources beyond vehicular traffic such as industrial facilities and regional sources. Average toluene/benzene ratios in this study were: B1 (0.46), B2 (0.28), B3 (0.30) and B4 (0.49) indicating that the contribution of vehicle sources to BTEX levels was important. The p-xylene to ethylbenzene ratio (X/Ebz) is commonly used as an indicator of the photochemical age of air masses at a given site. Values higher than 3.8 indicate aged air masses and low values indicate that the air masses are fresh (recent emissions) [19]. X/Ebz ratios were the following: B1 (4.056), B2 (2.73), B3 (3.37) and B4 (3.18) indicating that air masses corresponded to fresh emissions, emitted in situ, excepting during the morning sampling period (B1: 4.056). A Pearson correlation analysis was carried out in order to determine bivariate relations among the measured variables (See Table 2). Benzene, toluene and p-xylene showed good correlations among each other indicating that they had sources in common. Ethylbenzene did not correlate with the rest of BTEX, indicating that this compound was originated from a different source. Toluene and p-xylene showed a good correlation with NO_2 and $PM_{2.5}$, indicating that these compounds were originated from high temperature combustion sources. Ozone showed negative correlations with toluene, p-xylene, NO_2 and $PM_{2.5}$, indicating that these compounds act as ozone precursors. A Principal Components Analysis was applied in order to investigate the relations among BTEX and the different measured variables. Figure 4 shows the results of the Principal Component Analysis (PCA). Two principal factors were enough to explain 62.79% of the total variability of the data. Three groups (PCs) were

identified: PC1, PC2 and PC3. PC1 included benzene, toluene, p-xylene and CO (air pollutants emitted from vehicular sources). PC2 included $PM_{2.5}$ and NO_2 (air pollutants emitted from high temperature combustion sources) and that showed a negative significant relation with O_3, acting as ozone precursors through photochemical reactions. PC3 included only ethylbenzene, which probably had a source beyond vehicular traffic (regional or industrial sources).

Table 2. Pearson correlation coefficients for measured air pollutants

Variables	Benzene	Toluene	Ethylbenzene	p-Xylene	CO	NO₂	O₃	PM 2.5
Benzene	1	**0.765**	-0.134	**0.714**	0.006	0.223	-0.226	0.351
Toluene	**0.765**	1	0.036	**0.854**	0.077	**0.545**	**-0.540**	**0.432**
Ethylbenzene	-0.134	0.036	1	0.056	-0.185	0.048	-0.007	0.153
p-Xylene	**0.714**	**0.854**	0.056	1	0.241	**0.532**	**-0.510**	0.358
CO	0.006	0.077	-0.185	0.241	1	0.154	-0.100	0.121
NO₂	0.223	**0.545**	0.048	**0.532**	0.154	1	**-0.952**	**0.548**
O₃	-0.226	**-0.540**	-0.007	**-0.510**	-0.100	**-0.952**	1	**-0.417**
PM 2.5	0.351	**0.432**	0.153	0.358	0.121	**0.548**	**-0.417**	1
Values in bold are different at a significant level of α=0.05								

3.5 Health Risk Assessment
Daily average exposures to benzene (mg/kg per day) were calculated for children and adults being higher for children. The individual non-cancer risk quotient (HQ) for benzene was >1, indicating that long-term exposure to benzene could represent a health risk in the study area. The estimated cancer risk (ILTCR values) for adults and children were of 2.87 x 10⁻⁴ and 5.43 x 10⁻⁴, respectively, which are higher than those values established in the guidelines of US EPA (1 x 10⁻⁶) [9]. Cancer risks can be classified into three categories according to Sexton and collaborators [20]: as definitive risk if ILTCR > 1 x 10⁻⁴, probable risk (if ILTC ranks from 1x10⁻⁵ to 1x10⁻⁴) and possible risk (if ILTCR ranks from 1x10⁻⁵ to 1x10⁻⁶). According to this classification, population living in Tijuana would be at definitive risk of suffering cancer.

4 Conclusion
Relative abundance of BTEX had the following order: benzene (32.398 µg m⁻³) > p-xylene (17.155 µg m⁻³) > toluene (13.277 µg m⁻³) > ethylbenzene (7.023 µg m⁻³). BTEX levels showed a diurnal pattern at study site, with higher concentrations during the morning sampling period. Low T/B and p-X/Ebz ratios were observed during the study period, indicating that this site was influenced by local and fresh emissions (vehicular traffic). From the Pearson correlation and principal component analysis, it was possible to confirm that vehicular traffic was the main source for benzene, toluene and p-xylene, and that toluene and p-xylene could contributed to tropospheric ozone. Ethylbenzene did not correlated

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with the rest of measured air pollutants, indicating that it probably had its origin in a different source. Hazard quotient value (HQ) for benzene exceed unity, so it is possible to conclude that the population of Tijuana is exposed to a definitive risk of suffering cancer in their lifetime due to inhalation exposure to benzene. Therefore, it is necessary to strengthen existing environmental policies, promote the establishment of a national standard that regulates BTEX levels in ambient air, as well as to improve the quality of fuels, subsequently promoting the usage of alternative energy sources and especially improved control of area source emissions.

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