



Influence of the seasonality and of urban variables in the BTEX and PM_{2.5} atmospheric levels and risks to human health in a tropical coastal city (Fortaleza, CE, Brazil)

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Abstract

The International Agency for Research on Cancer (IARC) classifies benzene in group 1 (carcinogenic to humans). Particulate matter (PM) has recently also been classified in this category. This was an advance toward prioritizing the monitoring of particles in urban areas. The aim of the present study was to assess levels of PM_{2.5} and BTEX (benzene, toluene, ethylbenzene, and xylene), the influence of meteorological variables, the planetary boundary layer (PBL), and urban variables as well as risks to human health in the city of Fortaleza, Brazil, in the wet and dry periods. BTEX compounds were sampled using the 1501 method of NIOSH and determined by GC-HS-PID/FID. PM_{2.5} was monitored using an air sampling pump with a filter holder and determined by the gravimetric method. Average concentrations of BTEX ranged from 1.6 to 45.5 $\mu\text{g m}^{-3}$, with higher values in the wet period, which may be explained by the fact that annual distribution is influenced by meteorological variables and the PBL. PM_{2.5} levels ranged from 4.12 to 33.0 $\mu\text{g m}^{-3}$ and 4.18 to 86.58 $\mu\text{g m}^{-3}$ in the dry and wet periods, respectively. No seasonal pattern was found for PM_{2.5}, probably due to the influence of meteorological variables, the PBL, and urban variables. Cancer risk ranged from 2.46E⁻⁰⁴ to 4.71E⁻⁰³ and 1.72E⁻⁰⁴ to 2.01E⁻⁰³ for benzene and from 3.07E⁻⁰⁶ to 7.04E⁻⁰⁵ and 3.08E⁻⁰⁶ to 2.85E⁻⁰⁵ for PM_{2.5} in the wet and dry periods, respectively. Cancer risk values for benzene were above the acceptable limit established by the international regulatory agency in both the dry and wet periods. The results obtained of the noncarcinogenic risks for the compounds toluene, ethylbenzene, and xylene were within the limits of acceptability. The findings also showed that the risk related to PM is always greater among smokers than nonsmokers.

Keywords BTEX · PM_{2.5} · Ambient air · Seasonal variation · Health risks · Coastal city

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Introduction

Air quality in urban areas is the result of the interaction of the climate, meteorology, topography, social aspects, and pollutants emitted by human activities (Bagieński 2015). Urban development in recent decades has had a considerable impact on air quality in cities around the world. The United Nations estimates that the world population will reach 11 billion people by the end of the current century and approximately 7.2 billion will be urban residents (United Nations 2019), which will result in the increased use of resources as well as greater waste production and the emission of pollutants.

Among the substances found in the air of urban centers, particulate matter (PM) and BTEX (benzene, toluene, ethylbenzene, and xylene) are important classes of atmospheric pollutants. The dominant sources of these pollutants are

industrial and urban activities, especially vehicular emissions (Karagulian et al. 2015; Liu et al. 2018; Rivas et al. 2020). PM is a complex mixture of suspended solid and liquid particles of a natural and/or anthropogenic origin (Hamra et al. 2014) and is composed of gross (2.5 μm to 10 μm), fine (0.1 μm to 2.5 μm), and ultrafine ($\leq 0.1 \mu\text{m}$) particles, which are, respectively, denominated PM_{10} , $\text{PM}_{2.5}$, and $\text{PM}_{0.1}$ (US-EPA 2016). BTEX is a widely studied group of pollutants in the class of volatile organic compounds (VOCs) (US-EPA 2016).

As demonstrated in studies conducted in the past 50 years, the complex mixture of pollutants in outdoor air contains several carcinogenic agents associated with lung cancer (Hamra et al. 2014; US-EPA 2016). The International Agency for Research on Cancer (IARC) classifies benzene in group 1 (carcinogenic to humans) and has recently classified atmospheric PM in urban centers in this same group (IARC 2018). The other pollutants in the BTEX group are also classified according to their potential risk to human health. Toluene and xylene are part of group 3 (unclassifiable regarding carcinogenicity in humans), and ethylbenzene is part of group 2B (possibly carcinogenic to humans) (IARC 2018).

Air pollution has become a global problem that especially affects developing countries, such as Brazil (Toledo et al. 2018; Almeida et al. 2019). According to the World Health Organization (WHO 2020), air pollution is one of the main causes of premature deaths (4.2 million deaths annually worldwide). Frequent exposure to this pollution has been associated with most cases of respiratory and cardiovascular morbidity and mortality in urban areas and is related to lung cancer, hypertension, nervous system imbalance, endothelial dysfunction, diabetes, heart failure, and genetic mutations (Lu et al. 2015; Perez et al. 2015; Garcia et al. 2016; Renzic et al. 2018).

Studies report levels of $\text{PM}_{2.5}$ and BTEX in urban areas of countries around the world, such as India, France, Iran, China, the United States, and Brazil (Amini et al. 2017; Jiang et al. 2017; Hsu et al. 2017; Rocha et al. 2017; Garg and Gupta 2019; Yang et al. 2019; Cruz et al. 2020). The level of atmospheric pollution varies throughout the year due to specific climatic conditions in each region, the predominant polluting activities, and the composition of the pollutants. Moreover, the concentration of pollutants, especially PM, and the degree of exposure of the population are affected by the atmospheric conditions of a given period, directly influencing air quality (Hu et al. 2017; Chen et al. 2018; Han et al. 2020).

Due to improvements in the Brazilian economy in recent years, the city of Fortaleza has experienced a real estate boom along with a more than 100% increase in the fleet of vehicles. However, this has been occurring in a disorderly manner and without adequate urban planning. The construction of high rises and increased vehicular traffic has led to the formation of deep canyons with reduced environmental quality and poorer air pollution indicators (Sousa et al. 2015; Silva et al.

2016; Aguiar et al. 2017; Cavalcante et al. 2017; Rocha et al., 2017).

The aims of the present study were to (1) determine atmospheric levels of $\text{PM}_{2.5}$ and BTEX; (2) assess the influence of meteorological variables (wind velocity, solar irradiation, temperature, precipitation, and relative humidity), the planetary boundary layer, and urban variables (level of urbanization and vehicular fleet); and (3) determine risks to human health in the city of Fortaleza, CE, Brazil, during wet and dry periods. These data will be useful, as studies on air quality and associated variables in equatorial areas are scarce. The findings can also assist in sensitizing governments regarding the need for public policies that prioritize air quality in urban areas of emerging countries.

Experimental section

Sampling sites

The city of Fortaleza is located on the Atlantic coast in the northeast region of Brazil and has an area of approximately 314.93 km^2 , with a population of more than 2.5 million residents (IBGE 2016) (Fig. 1). The region has two well-defined seasons: a dry season from August to December and a wet season from January to July. The average temperature ranges from 25 to 28 $^{\circ}\text{C}$; the average annual wind velocity is 3.5 m s^{-1} , and the average annual rainfall is 1600 mm, with rains concentrated between February and May (Sousa et al. 2015; Gusev et al. 2014; Rocha et al. 2017).

In 2019, the city reached the mark of 1,133,901 vehicles, approximately 37.4% of which use gasoline, 2.9% are fueled by hydrated ethanol, and 8.2% are powered by diesel. Flexible-fuel (gasoline or ethanol) vehicles represent approximately 45.8% of the total fleet, and their number is rapidly increasing (DETRAN-CE 2019). Besides vehicular emissions, port, petrochemical, and food industries are found in the eastern portion of the city, an industrial center is found in the southern portion, and a port/industrial area is found in the western portion. Offshore oil extraction activities also occur in the area (Fig. 1).

Sampling was performed in 2015 between May and June (wet season) as well as in September and October (dry season) on Abolição Avenue (P1), University Avenue (P2), Marechal Deodoro Street (P3), Bezerra de Menezes Avenue (P4), and Aguanambi Avenue (P5). Aguanambi Avenue had a daily flow of 63,455 vehicles, with 34 bus lines, and was the busiest avenue in the city in 2015. Bezerra de Menezes Avenue had a daily flow of 48,837 vehicles, with 26 bus lines, and was the 4th busiest avenue in the city. Abolição Avenue had a daily flow of 47,434 vehicles, with 26 bus lines, and was the 6th busiest avenue. University Avenue had a daily flow of 25,656 vehicles, with 42 bus lines, and was the 30th busiest avenue in

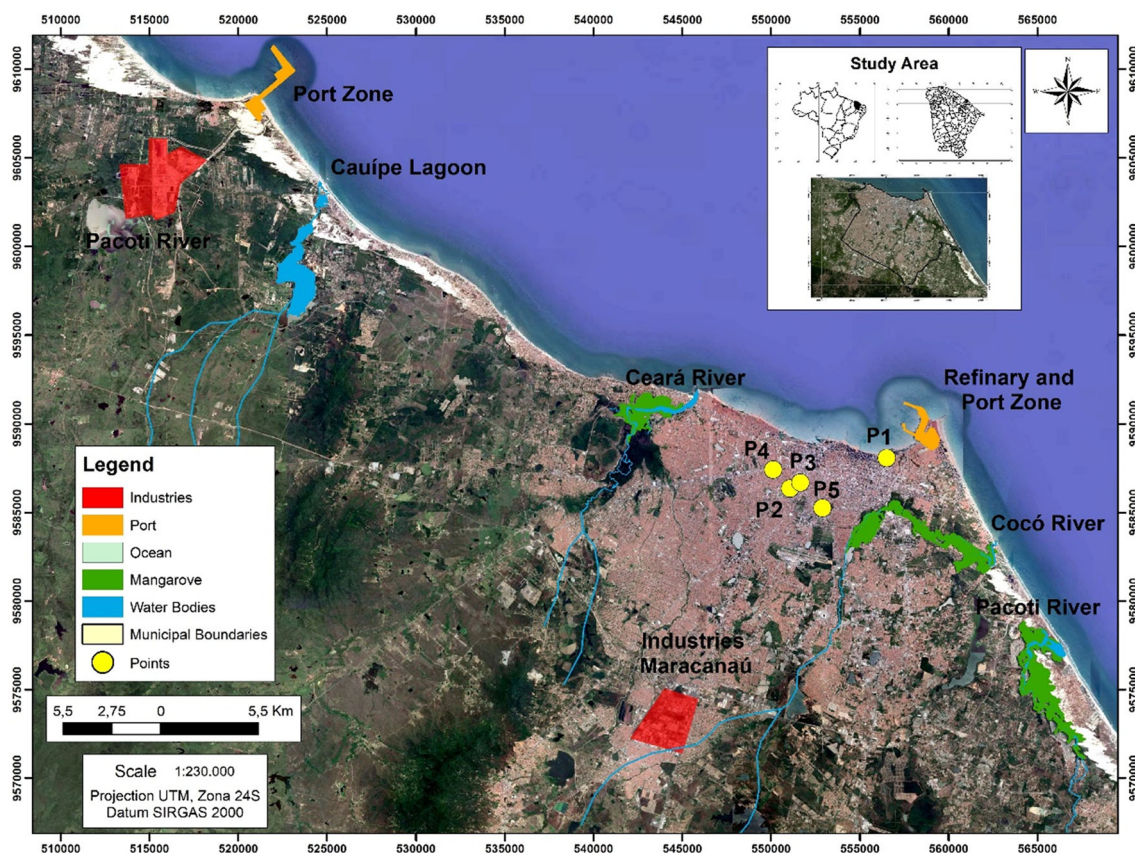


Fig. 1 Location of sampling points

the city. Marechal Deodoro Street is a secondary street and was chosen for having a low flow of vehicles (approximately six per minute), serving as a control point. Sampling was performed in duplicate (on different days) and twice daily between 08:00 and 19:00 h. Therefore, all data are the mean of four collections per point in each season.

Sampling and analysis

BTEX

BTEX sampling was conducted using the 1501 method of the US National Institute for Occupational Safety and Health (NIOSH 2003). Air was collected with the aid of a pump (4 h of active sampling at a flow rate of 0.5 to 1.0 L min⁻¹). The air was drawn through Orbo™-32 tubes (7 cm in length × 6 mm o.d., provided by Supelco), which have activated charcoal. The sampling pump was calibrated prior to each collection, and the error in the calculated variation of the flow was 2 to 7%. The system was mounted 1.50 m from the ground (approximate height of the breathing zone). After collection, the ORBO adsorbents were immediately placed in screw-top glass vials (22 mL with PTFE/silicone septa), which were sealed with the aid of grip pliers and stored at a low temperature until the chromatographic determinations. BTEX

quantifications were performed with a TriPlus HS headspace autosampler (Thermo Electronic Corporation, Milan, Italy) and a Trace GC Ultra gas chromatograph (Thermo Electronic Corporation, Milan, Italy) equipped with a photoionization detector and flame ionization detector connected in series (GC-HS-PID/FID). The 54-space autosampler included a robotic arm and a headspace generation unit, with an oven (100 °C) to heat and shake the samples and a 3-mL syringe (100 °C) to transfer the gas into the GC equipment. Samples were injected in a splitless mode for 0.8 min. Chromatographic separation was performed in an OV-624 (Ohio Valley, Ohio, USA) capillary column (60 m, 0.32 mm i.d., film thickness: 1.8 μm). The initial oven temperature was 40 °C for 2 min, which was increased to 140 °C at 40 °C min⁻¹ and 140 to 200 °C at 7 °C min⁻¹. The quantification and identification of BTEX compounds were performed using a mixture of standards from Supelco. Identification was based on retention time. Calibration curves were prepared using six concentrations of standards (5 to 500 μg L⁻¹), with a correlation coefficient (R²) greater than 0.99. The standards were injected at least three times, and the error between injections was <3%.

Quality control/analytical quality assurance was performed as reported in Cavalcante et al. (2010), followed by the standard operating procedure to minimize background

contamination. All glass devices were cleaned by rinsing with ultrapure water and kept overnight in a muffle furnace at 450 °C to remove organic contaminants. The septa were cleaned for 20 min in an ultrasonic bath filled with deionized water, rinsed with deionized ultrapure water, and dried in an oven at 105 °C. The septa and vials were not reused. System blanks free of BTEX (field blanks not attached) were analyzed before every standard of the calibration curve. Blank analytical samples of the gas chromatographic methods were also used to minimize background contamination and avoid carry-over between samples. Some samples were chosen for the duplication of the measurements to verify the validity and reproducibility of the data.

Particulate matter (PM_{2.5})

For the determination of PM_{2.5}, a portable air sampling pump with a filter cassette (SKC) was positioned approximately 1.5 m from the ground. Sampling was performed with a flow of 1.5 L min⁻¹ for 10 to 11 h. A 47-mm glass fiber filter with a porosity of 0.7 μm (Millipore) was used. Before and after collection, the filters remained in an oven at 60 °C for 48 h to remove the moisture. After sampling, the filters were taken to the laboratory, and the difference in weight (to five decimal places) between the clean and collected filters was determined on a scale, following the procedure suggested by Kim Oanh et al. (2009).

Health risk assessment

Carcinogenic and noncarcinogenic risks

Many studies have estimated the toxic risks of BTEX and MP_{2.5} to human health (Hamra et al. 2014; Masih et al. 2016; Miri et al. 2016; Rocha et al. 2017; Fontenele et al. 2018; Dehghani et al. 2018; Garg and Gupta 2019). Benzene poses a cancer risk, while toluene, ethylbenzene, and xylene have toxic but noncancerous effects. The method proposed by the US Environmental Protection Agency (US-EPA) was adopted to assess the health risks of cancer and other diseases through inhalation pathways. This method is the most widely used in health risk assessment studies, as cancer guidelines incorporate scientific principles and policies based on currently available information. The EPA constantly reviews these cancer guidelines to make substantial changes when needed (US-EPA 2005).

The risk to human health was estimated using the chronic daily intake (CDI) (mg.kg⁻¹.dia⁻¹) per lifetime model (Eq. 1) considering the variables listed in Table 1. These data were collected from questionnaires administered to users of the sampling sites.

Table 1 Variables used in the calculation of carcinogenic and noncarcinogenic risks

Variable	Description	Value	Unit
CA	Contaminant concentration	–	mg.m ⁻³
IR	Inhalation rate (adult)	1.02	m ⁻³ .h ⁻¹
ED	Exposure duration (adult)	7 ^a	h ⁻¹ .week ⁻¹
EF	Exposure frequency	52	week ⁻¹ .year ⁻¹
L	Exposure time	35 ^b	Year
BW	Body weight man/woman	70/60	Kg
ATL	Average life expectancy man/woman	72/79 ^c	Year
NY	Number of days of year	365	day ⁻¹ .year ⁻¹

^a Considering only the displacement factor, whose person is exposed, on average, 1 h a day on the avenues

^b Considering a person visits the avenues for a maximum of 35 years

^c IBGE (2014)

$$CDI = \frac{(CA \cdot IR \cdot ED \cdot EF \cdot L)}{(BW \cdot ATL \cdot NY)} \tag{1}$$

According to the US-EPA (1997), a person during normal activities has an inhalation rate ranging from 0.75 to 1.02 m³ of air per hour, which is considered low inhalation. A person performing functions quickly has an inhalation rate of 3.06 m³ per hour, which is considered high inhalation. In the present study, we used a low inhalation rate (1.02 m³ h⁻¹).

Risk related to carcinogenic substances was estimated by multiplying the CDI by the slope factor (SF) (Eq. 2) (0.0273 mg.kg.day⁻¹) based on a unit risk value of 7.8E 10⁻⁶ μg m⁻³ and a standard inhalation rate of 20 m³ day⁻¹ for benzene, according to the online database of the Risk Assessment Information System (RAIS 2020). For PM_{2.5}, the risk of cancer was estimated by multiplying the CDI by the relative risk (RR) (Eq. 3) according to the US-EPA (1992, 1996) and a study conducted by Hamra et al. (2014). In the present investigation, we used RR values for all types of people (1.09 – general risk), for those who smoked for many years (1.44 – ex-smokers), and for those who never smoked (1.18 – non-smokers).

$$CR = CDI \cdot SF_{Benzene} \tag{2}$$

$$CR = CDI \cdot RR_{PM2.5} \tag{3}$$

The noncarcinogenic risk was estimated by dividing the CDI (mg.kg⁻¹.day⁻¹) per lifetime by the reference dose (RfD - mg.kg⁻¹.day⁻¹) (Eq. 4) expressed in the literature, generating the risk index or hazard quotient (HQ) (NRC 1990; DEP 2002).

$$HQ = \frac{CDI}{RfD} \tag{4}$$

If the HQ is less than 1, exposure to the chemical in question is considered unlikely to cause adverse health effects. If the HQ is greater than 1, the likelihood of adverse health effects is high, and remedial action or mitigation is likely required. HQ is not a risk measure but rather a starting point for estimating risk (Williams et al. 2000; Yu 2004). The HQ was calculated for toluene, ethylbenzene, and xylene.

Atmospheric data

The meteorological data (wind velocity, solar irradiation levels, air humidity, temperature, and precipitation) used in this study were obtained from the Brazilian National Meteorology Institute (INMET) considering three periods per day: 8:00, 12:00, and 16:00. The mean of the three periods was calculated to determine the mean daily value of each variable.

The height of the planetary boundary layer (PBL) was determined by radio sounding data over the city of Fortaleza obtained during the dry and wet season from the following website: weather.uwyo.edu/upperair/sounding.html. The following are the data from the Fortaleza station: station number, 82397; observation time, 12:00 Z; station latitude, -3.76 ; station longitude, -38.60 ; and station elevation, 19 m. The sounding index used for the subjective determination of PBL height was the potential temperature (θ) in Kelvin (profile method). When θ values begin to increase systematically (at least three $d\theta/dz$ points greater than zero), the top of the PLB is surpassed (free atmosphere). The first level where this occurs is used to identify the top of the PLB.

Urbanization levels

The level of urbanization at each sampling point was estimated based on vehicular flow. The classification of each location was based on the ratio between the height of the buildings and width of the streets (H/W ratio) (Nakamura and Oke 1988; Battista et al. 2015). Urban canyons were divided into three types: shallow canyons ($H/W < 0.5$), regular canyons ($H/W \approx 1$), and deep canyons ($H/W > 2$).

Results and discussion

Concentrations of BTEX and PM_{2.5} in the ambient air: influence of seasonality and urban variables

The seasonal levels of BTEX and PM_{2.5} are displayed in Table 2. Benzene was the most abundant gas, ranging from 0.48 to 22.99 $\mu\text{g m}^{-3}$. The other values were 0.07–10.18 $\mu\text{g m}^{-3}$ for m/p-xylene, 0.69–7.96 $\mu\text{g m}^{-3}$ for toluene,

Table 2 Average concentrations of BTEX and PM_{2.5} in Brazil and other localities compared to this study

Location	$\sum_{\text{BTEX}} (\mu\text{g m}^{-3})$	Setting
Delhi, India ¹	61.9–237.5	Urban/industrial
Khuzestan, Iran ²	4.37–14.58	Urban/industrial
Nuevo Leon, Mexico ³	29.36–88.59	Urban
Almaty, Kazakhstan ⁴	135	Urban
Yazd, Iran ⁵	8–560	Urban
Sakakah, Saudi Arabia ⁶	134.79–1387.93	Industrial
Taiyuan, China ⁷	38.9	Urban
Salvador, Brazil ⁸	5.9	Urban
Fortaleza, Brazil	7.95	
	1.6–45.5	Urban/industrial
Recife, Brazil ⁹	PM _{2.5} ($\mu\text{g m}^{-3}$)	Setting
	7.3±3.1	Urban
Delhi, India ¹⁰	60.54±11.6	Urban
Constantine, Algeria ¹¹	57.8±39.6	Urban
Bursa, Turkey ¹²	53	Urban/industrial
Sao Paulo, Brazil ¹³	31.83	Urban
	23.74	
Rio Grande do Sul, Brazil ¹⁴	25.89–64.71	Urban
Ulsan, South Korea ¹⁵	24.7±13.0	Urban/industrial
	26.7±9.9	
Taiwan, China ¹⁶	21.98±17.1	Urban/industrial
Shenyang, China ¹⁷	80	Urban/industrial
Fortaleza, Brazil	4.12–86.58	Urban/industrial

Sources: ¹ Singh et al. (2016); ² Rad et al. (2014); ³ Cerón-Bretón et al. (2015); ⁴ Baimatova et al. (2016); ⁵ Hajizadeh et al. (2018); ⁶ El-hashemy and Ali (2018); ⁷ Li et al. (2020); ⁸ Cruz et al. (2020); ⁹ Miranda et al. (2012); ¹⁰ Goyal and Kumar (2013); ¹¹ Terrouche et al. (2015); ¹² Kendall et al. (2011); ¹³ Boas et al. (2018); ¹⁴ Silva et al. (2015); ¹⁵ Oh et al. (2018); ¹⁶ Lee et al. (2019); ¹⁷ Liu et al. (2020)

and 3.10 $\mu\text{g m}^{-3}$ for ethylbenzene, and nd-1.52 $\mu\text{g m}^{-3}$ for o-xylene. PM_{2.5} levels ranged from 4.12 to 86.58 $\mu\text{g m}^{-3}$. Due to the scarcity of studies on PM_{2.5}, the results were compared to findings from the majority of studies conducted since 2010 investigating BTEX and PM_{2.5} separately in urban and industrial environments, focusing on investigations conducted in tropical locations or regions with similar weather. Both \sum_{BTEX} and PM_{2.5} levels were lower than those reported for highly industrialized/urbanized regions and similar to those reported for locations with medium degrees of urbanization and industrialization (Table 2).

Mean total BTEX concentrations (\sum_{BTEX}) ranged from 1.6 to 45.5 $\mu\text{g m}^{-3}$ throughout the year. Mean PM_{2.5} levels in the dry and wet periods ranged from 4.12 to 33.00 $\mu\text{g m}^{-3}$ and 4.18 to 86.58 $\mu\text{g m}^{-3}$, respectively (Table 2). BTEX values were higher in the wet period than in the dry period. In contrast, no seasonal distribution pattern was found for PM_{2.5} (Fig. 2).

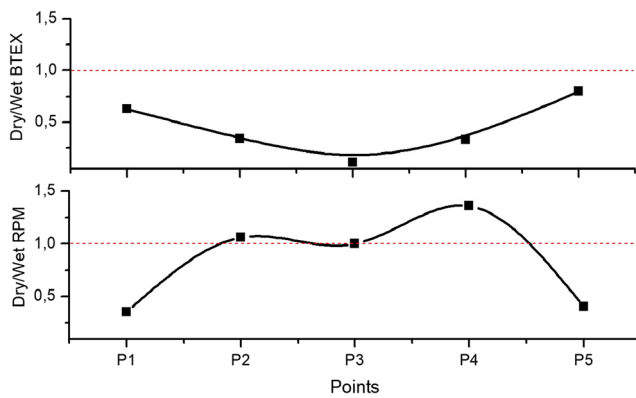


Fig. 2 Dry/wet ratio for annual distribution of PM_{2.5} and BTEX

Due to the scarcity of data from equatorial zones, studies conducted in regions with similar weather were considered for the purposes of comparison. Regarding seasonal variability, the highest mean Σ_{BTEX} values were found in the wet period at all sampling points. During this period, the concentration of pollutants tends to be higher due to greater atmospheric stability, with higher volumes of rain and small vertical and horizontal movements of the air layers due to the lower wind speed. In the dry season, atmospheric instability favors the dispersion and dilution of pollutants both vertically, due to the increase in the urban boundary layer, and horizontally, due especially to the increase in wind speed, leading to lower concentrations (Rad et al. 2014; Bauri et al. 2015; Singh et al. 2016).

According to Matysik et al. (2010), strong vertical temperature gradients in summer lead to greater vertical convection, with the efficient dispersal of BTEX in the air. In some regions, the stability of the atmosphere and low temperatures are more associated with higher concentrations than rain events (Singh et al. 2016). Rad et al. (2014) attributed the lower levels of BTEX in summer to the increase in solar radiation and photochemical reactions.

Some studies report higher BTEX concentrations in summer compared to autumn, presumably due to predominant winds from areas with sources of volatile organic compounds (industries, high traffic, gas stations, and airports), temperature (higher in summer), and relative humidity (higher in autumn) (Cerón-Bretón et al. 2015). Alghamdi et al. (2014) attributed higher BTEX levels in summer compared to winter to evaporative processes (e.g., solvent usage, gasoline evaporation from the headspace of fuel tanks, and evaporation from gasoline spillage during tank refueling).

PM can be primary or secondary. Primary PM is composed of materials emitted directly into the atmosphere, and its concentration depends on the emission/removal rate, transport, and dispersal (Beckett et al. 1998; Oke et al. 2017). Secondary PM is formed by condensed vapors due to a chemical reaction involving precursors of the gaseous phase or other processes (Oke et al. 2017). Thus, concentrations of

PM can change in accordance with the meteorological conditions and type of environment (rural, urban, or industrial) (Seinfeld and Pandis 2016; Xie et al. 2005; Buccolieri et al. 2009; Bagieski 2015).

As seen in Fig. 2, P1 and P5 had a dry/wet ratio < 1, P2 and P3 had a dry/wet ratio of around 1, and P4 had a dry/wet ratio < 1, with no seasonal distribution pattern. This is likely due to the effect of different variables, leading to variations in PM levels from 1 year to the next (Xie et al. 2005; Buccolieri et al. 2009). Thus, factors such as local atmospheric chemistry and meteorology as well as the characteristics of urban roads, architecture, and trees can affect the concentration and temporal distribution of PM (Uehara et al. 2000).

For example, rainfall is one of the main meteorological variables due to the more stable atmosphere, generally characterized by lower wind velocities and higher humidity, leading to less horizontal and vertical dispersion and, consequently, lower PM levels in the atmosphere. In contrast, the atmosphere is less stable in the dry period (especially summer), with higher wind speeds and lower humidity; therefore, vertical and horizontal winds exert considerable influence on the resuspension and dispersal of PM (Seinfeld and Pandis 2016; Gioia et al. 2010; Budhavant et al. 2015; Wang and Fang 2016). However, this is not a standard pattern. In subtropical cities at middle latitudes, such as São Paulo, Rio de Janeiro, Belo Horizonte, and Curitiba, winter is characterized by lower temperatures, lower relative humidity, and less precipitation, resulting in higher concentrations of PM_{2.5}, whereas lower concentrations are found in winter in the city of Recife, which is located in the equatorial zone (Miranda et al. 2012).

Studies conducted in coastal arid/semiarid regions report that rainfall is more efficient at lowering PM levels compared to wind velocity (Kendall et al. 2011; Flores-Rangel et al. 2015; Terrouche et al. 2015). Precipitation “cleans” the air, settling the sedimented particulate matter and reducing resuspension due to the moist soil in the period (Gioia et al. 2010; Rocha et al. 2017).

However, there are other aspects besides meteorological variables that must be considered. According to Flores-Rangel et al. (2015), high PM concentrations are due to heavy vehicular traffic and stability in the atmosphere. The high concentration of PM may be associated with the transport of pollutants from urbanized or industrialized areas, some of which travel long distances (Budhavant et al. 2015; Terrouche et al. 2015; Wang and Fang 2016). Kendall et al. (2011) found that PM₁₀ and PM_{2.5} fractions were strongly correlated in the cold season but not in the warm season, demonstrating a change in the annual distribution pattern.

Especially in urban areas, characteristics such as topography, vegetation, and focal sources are essential variables that must be considered in the spatiotemporal distribution of PM levels (Xie et al. 2005; Cavalcante et al. 2017; Rocha et al. 2017). Strong correlations are found between the building

index and high PM concentrations, especially in developing cities with urban canyons (Silva et al. 2016; Oke et al. 2017; Fontenele et al. 2018; Marques et al. 2019). More topographic features and the arrangement of buildings significantly reduce wind speed, consequently lowering the dispersal of pollutants through the streets (Xie et al. 2005). On the other hand, the green index has an inverse correlation, indicating that greater vegetation leads to a reduction in PM levels. This inverse correlation is plausible, as particulate pollutants could be deposited on plant surfaces through gravity (sedimentation) and impaction or could be simply deposited on the surface wax (Nowak 2006; Jim and Chen 2008). For example, ambient PM concentrations were found to be lower in neighborhoods with dense vegetation than those without dense vegetation (Freiman et al. 2006).

In urban areas, the influence of meteorology, sources, vegetation, air movements, and the shapes and sizes of urban constructions on air quality is not yet clear, primarily because these variables can act together or separately, making each urban environment unique (Silva et al. 2016; Oke et al. 2017; Fontenele et al. 2018; Marques et al. 2019; Cavalcante

et al. 2017; Rocha et al. 2017). Therefore, the effects of atmospheric variables on the distribution of BTEX and PMs were assessed in the present study using meteorological variables (e.g., wind velocity, solar irradiance, temperature, precipitation, and humidity) and the variability in the urban mixture layer (Table 3). The type of vehicular fleet (diesel or Otto combustion) and the classification of urban canyons were used for the assessment of the urbanization effect (Table 3).

Regarding the effect of the atmosphere on the annual distribution of BTEX levels, positive correlations were found with temperature, humidity, and precipitation, whereas a weak negative correlation was found with solar irradiance, and moderate inverse correlations were found with wind velocity and PBL height (Table A1, Supplementary matter).

Atmospheric conditions are important factors to the chemistry of the construction/destruction of BTEX (Tang et al. 2007; Alghamdi et al. 2014; Tiwari et al. 2017). Solar irradiance plays a fundamental role in equatorial regions, as it governs other variables that exert an influence on the chemistry of atmospheric pollutants. The increase in irradiance contributes to an increase in temperature and the instability of the

Table 3 Meteorological parameters, urban variables and concentrations ratios of T/B, *m,p*-X/B, *o*-X/B, and *m,p*-X/E during wet and dry periods in the city of Fortaleza, CE, Brazil

	P1		P2		P3		P4		P5	
	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
Meteorological										
Wind velocity (m/s)	2.67	4.0	3.17	4.0	3.0	4.17	2.67	4.0	3.0	4.0
Insolation levels (kJm ²)	1299.50	1406.33	1396.00	1467.0	1281.83	1616.83	1307.33	1460.33	1044.33	1607.17
PBL height (m)	799.0	1540.0	818.0	1435.0	801.0	1547.0	815.0	1543.0	811.0	1535.0
Air humidity (%)	69.67	61.33	64.33	63.33	70.0	55.0	67.83	58.0	63.83	60.17
Temperature (°C)	28.6	28.3	28.8	28.1	28.6	28.5	28.3	28.3	27.5	29.0
Precipitation (mm/h)	0.00	0.00	0.00	0.25	0.25	0.00	0.50	0.00	0.50	0.00
Urban variables										
Vehicular flow (v/h)										
Otto	1185	1345	2910	2800	350	390	1967	1683	2173	2338
Diesel	280	360	520	520	25	50	335	370	248	458
Ratio H/W	0.1		0.1		1.2		0.4		0.8	
BTEX ratios										
T/B	0.53	1.07	0.43	0.52	5.21	0.83	0.35	0.44	0.33	0.50
<i>m,p</i> -X/B	0.84	0.72	0.93	0.91	3.75	0.08	0.44	0.09	1.16	0.23
<i>o</i> -X/B	0.14	0.00	0.22	0.23	1.08	0.00	0.06	0.02	0.04	0.04
<i>m,p</i> -X/EB	3.72	5.67	6.43	4.76	3.08	0.00	3.29	4.30	8.62	0.00

T/B, toluene/benzene (close to 1, indicates traffic-originated emission sources (0.5–4.3), and the value increases with the closeness of the pollution source), and T/B > 10 indicates presence of industrial sources (Rad et al. 2014)

m,p-X/B, *m,p*-xylene/benzene (low reason, aging of the air mass and effects of photochemical reactions, while high reason, new air mass (recent emissions)), and *m,p*-X/B < 1.8 indicates traffic-originated emission sources (Rad et al., 2014)

o-X/B, *o*-xylene/benzene (low reason, aging of the air mass and effects of photochemical reactions, while high reason, new air mass (recent emissions)), and *o*-X/B < 0.9 indicates traffic-originated emission sources (Rad et al., 2014)

m,p-X/EB, *m,p*-xylene/ethylbenzene (low reason, aged air mass, while high reason new air mass (recent emissions))

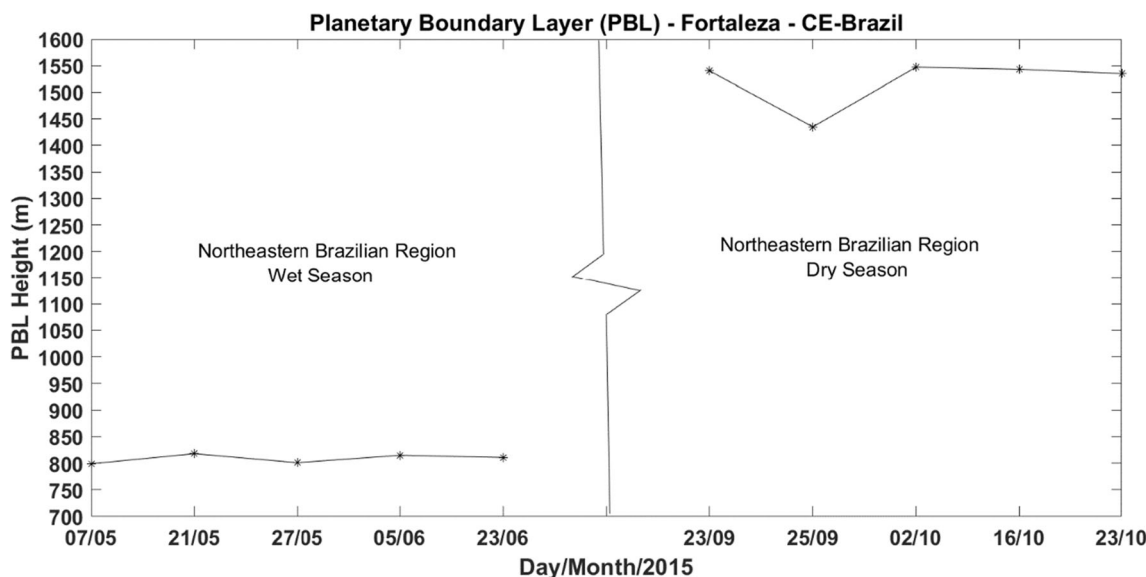


Fig. 3 PBL height over Fortaleza on different days and in different seasons in 2015: May 7, May 21, May 27, June 5, and June 23 (wet season); September 23, September 25, October 2, October 16, and October 23 (dry season)

atmosphere, which, in turn, increases wind velocity and both the height and variation of the PBL. Alghamdi et al. (2014) attribute the decrease in BTEX levels to enhanced atmospheric mixing, high temperatures, and the intensity of solar radiation, leading to an increase in photochemical reactions and, consequently, the increased chemical loss of such pollutants.

Regarding PM levels, the contributions of atmospheric variables were inversely proportional, with the exception of humidity and precipitation, which had a weak positive correlation. This is plausible, as solar irradiance exerts a proportional influence on temperature and wind velocity as well as PBL height, which tends to expand, causing a dilution effect on PM levels. According to Tiwari et al. (2017), the PBL remains at high altitudes for a long period of the day in warmer seasons (e.g., spring and summer), contributing to dilution processes in the atmosphere.

The PBL height ranged from 799 to 818 m in the wet season and 1435 to 1547 m in the dry season (Fig. 3). Thus, height was about twofold greater in the dry season. According to Chandra et al. (2014), specific humidity plays an important role in the variation of the height of the mixed layer. In the present investigation, specific humidity was approximately 1.2-fold higher in the wet season, helping to explain, together with the θ profiles, the observed PBL height in the different seasons.

A weak inverse correlation was found regarding the effect of urbanization on the distribution of BTEX and PM levels, indicating that other variables exert a more significant influence. Moreover, a weak positive correlation was found between the distribution of BTEX/PM and both Otto- and diesel-powered vehicles. Although this reflects the influence of burning fuels, there must also be contributions from other urban activities not considered in this study. Besides fuel

stations, the use of biomass as an energy source for barbecue restaurants and bakeries is a common activity in the city of Fortaleza that contributes greatly to the occurrence of volatile compounds and PM in urban areas (Sousa et al. 2015; Cavalcante et al. 2017; Rocha et al. 2017).

Ratios between concentrations of BTEX species are used to identify sources of emission and the time these species remain in the atmosphere (Rad et al. 2014). The data in Table 3 confirm the results of the correlation analyses: although BTEX levels reflect the influence of the burning of fuels, transformations in the atmosphere (old sources) and other sources also make contributions. For example, the T/B ratio > 10 is indicative of sources of industrial installations, while T/B between 0.5 and 4.3, $m,p\text{-X/B} < 1.8$, and $o\text{-X/B} < 0.9$ indicate traffic-originated emission sources or other sources (evaporative emissions, painting, cooking, etc.) (Rad et al., 2014). According to the T/B, $m,p\text{-X/B}$, and $o\text{-X/B}$ values, most places exhibited traffic-originated emission sources, except P3. Moreover, low X/B and X/EB ratios indicated aging of the air mass and effects of photochemical reactions (Tables 3) (Rad et al. 2014).

Risk to human health

Risk was estimated for carcinogens (e.g., benzene and PM_{2.5}) and noncarcinogenic compounds (e.g., toluene, ethylbenzene, m/p -xylene, and o -xylene) based on 1 hour per day of occupational exposure and 35 years separately for men during the wet and dry periods (Fig. 4). Unfortunately, no studies were found on cancer risk considering seasonality. Therefore, the present data were compared to findings from the few cancer risk studies conducted in outdoor environments. Based on the online RAIS database (slope factor of 0.0273 mg.kg.day⁻¹ and

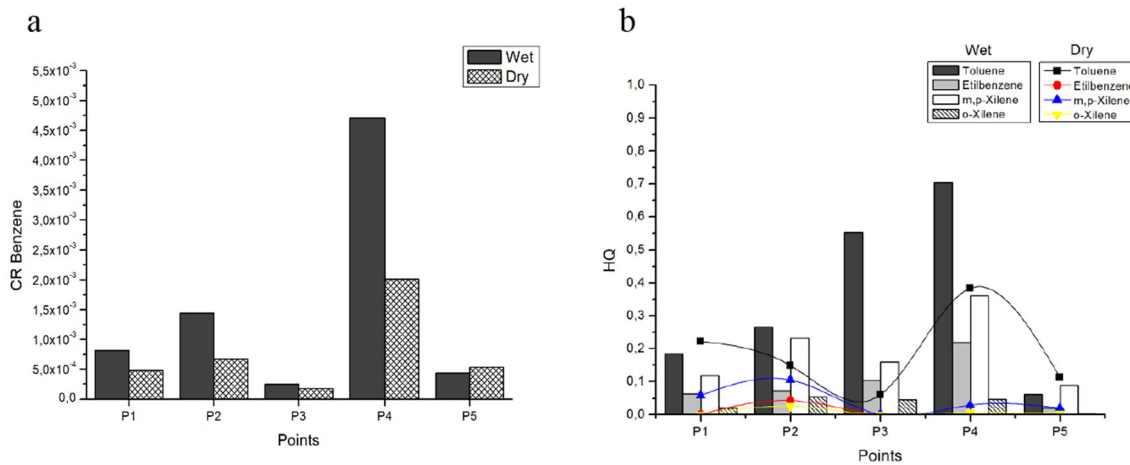


Fig. 4 Estimate of risk: **a** benzene and **b** toluene, ethylbenzene, *m/p*-xylene, *o*-xylene

unit risk value of $7.8E 10^{-6} \mu\text{g m}^{-3}$, the risk of cancer at all sampling points poses a threat to human health, as the values are well above the permitted limit. For benzene, which is the only carcinogen in the BTEX group, the risk was greater in the rainy season.

Nevertheless, one should bear in mind that this compound is a known carcinogen and greater exposure time increases the risk of cancer. This risk increases due to occupational exposure, specifically among individuals who work on the street. According to Pilidis et al. (2009), policemen who perform traffic-control duties and those who patrol on foot or in vehicles are exposed at a significantly higher (three to five times) benzene concentrations than the general population. Policemen who work with traffic are exposed to six times the amount of benzene compared to office policemen (Arayasiri et al. 2010).

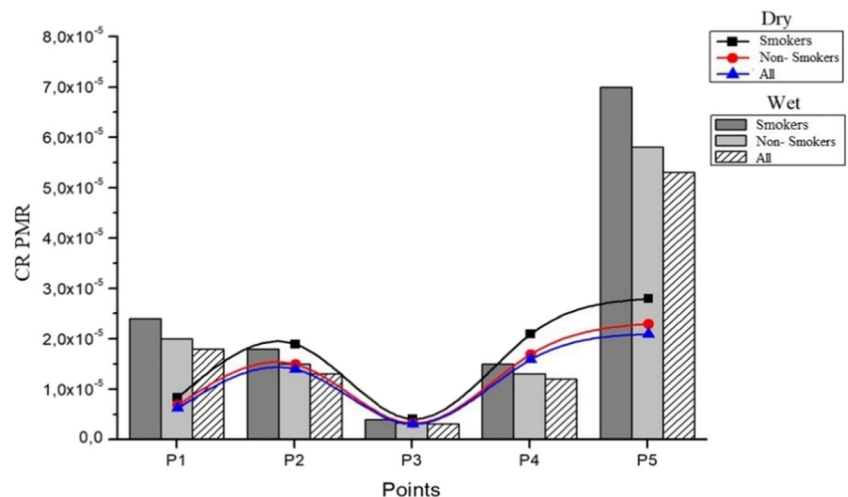
One should bear in mind that only 1 hour of exposure per day was considered in the present study and the calculation of the risk of cancer was specifically for individuals who use the avenues studied as travel routes. In calculating risk, not all variables directly linked to the probability of a person

developing cancer in life are considered specific morphological factors. Only a few external factors and general characteristics of a population were taken into account (Table 1).

The other compounds of the BTEX group (e.g., toluene, ethylbenzene, *m/p*-xylene, and *o*-xylene), which are considered toxic, were at safe levels ($HQ < 1$) in terms of the effects on human health. Among these compounds, toluene had the closest HQ to 1 in both seasons. However, even without these compounds surpassing the margin of safety ($HQ = 1$), long-term exposure can still affect one's health.

The cancer risk of $PM_{2.5}$ was estimated by multiplying the CDI by the relative risk (RR). Studies conducted in more than 30 countries have recently revealed that each $10 \mu\text{g m}^{-3}$ increase in $PM_{2.5}$ levels in the air has corresponded to a 9 to 36% increase in lung cancer rates in recent years (Raaschou-Nielsen et al. 2013; Hamra et al. 2014). The International Agency for Research on Cancer (IARC) recently classified external atmospheric particles (particles suspended in the air of outdoor/open environments) as group 1 carcinogens due to the fact that these particles cause permanent mutagenesis, heart attacks, diseases related to changes in blood pressure,

Fig. 5 Estimate of cancer risk due to $PM_{2.5}$



and even premature death (Hamra et al. 2014). This reclassification was an advance toward prioritizing the monitoring of PM, with the possibility of expansion through agencies linked to the occupational health of workers exposed to this class of contaminant (Hamra et al. 2014).

The highest risk of cancer found among the five sites was at P5 in both periods, and this can be due to the highest traffic. Moreover, the results show that the risk is always greater among smokers than nonsmokers (Fig. 5).

PM in urban areas is unfavorable to health and lowers environmental quality, especially in developing countries, where laws are insufficient and there is an absence of monitoring (Raaschou-Nielsen et al. 2013; Hamra et al. 2014; Cavalcante et al. 2017; Rocha et al. 2017; Fontenele et al. 2018). High levels of PM are also found in areas with considerable vehicular traffic and are directly related to hospitalizations due to respiratory diseases (e.g., asthma, rhinitis, etc.), especially in children under 5 years of age and cardiovascular diseases among middle-aged individuals, and can have an acute effect on the birth of premature infants (Lima et al. 2014; Nicolussi et al. 2014).

Conclusion

Particulate matter has recently been included in the class of carcinogens alongside benzene. This report is one of the first studies carried out in a tropical city on the risk of cancer involving both BTEX and PM_{2.5}. As no previous studies have analyzed these pollutants together, the findings of this work were compared to those from studies evaluating these pollutants separately. PM_{2.5} levels in the city of Fortaleza are in the same order of magnitude as those found in cities with medium to high urbanization and even industrialized areas. In contrast, BTEX levels were lower than those found in cities with medium to high levels of urbanization. Mean BTEX levels were higher in the wet season, whereas no seasonal pattern was found for PM_{2.5}. The cancer risk for benzene was considered high, posing a threat to human health at all sites studied based on the limit established by the Risk Assessment Information System. However, this risk can vary if the time of exposure to polluted air is shorter or longer than that considered in this study and if the person performs activities that require greater effort, such as physical exercise. In contrast, only one of the five locations evaluated in this study had a high risk of cancer related to PM. The comparison of these results to findings from other places was not possible due to the lack of studies.

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Author contribution Íthala S. Santiago, Tamiris F. A. Silva, Elissandra V. Marques, and Camille A. Rocha: conceptualization, investigation, and methodology; Francisco M. and S. Barreto: supervision on the chromatographic analysis; Antonio G. Ferreira: supervision on the height of the planetary boundary layer and writing – original draft; Kamila V. Mendonça: formal analysis, writing – original draft; Rivelino M. Cavalcante: supervision, writing – review and editing.

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