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Benzene, Toluene, Ethylbenzene and Xylene (BTEX) Concentrations in Urban Areas Impacted by Chemical and Petrochemical Industrial Emissions

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Abstract
The Metropolitan Region of Rio de Janeiro is the second largest urban and industrial region in Brazil. While the south and south-east areas are affected by vehicular emissions, the districts and cities located in the northern area are subjected to industrial emissions and have the poorest air quality of the region. In this study, BTEX concentrations were determined in the District of Irajá, a residential area located in the north of the city of Rio de Janeiro, approximately 25 km from the industrial zone, as well as in the District of Jardim Primavera, in the city of Duque de Caxias. The mean values for total BTEX concentrations were 38.4 ± 11.7 and 44.6 ± 29.3 μg m^{-3}, in Irajá and Jardim Primavera, respectively, which are higher than those previously reported for other areas. The benzene/toluene rates, (approximately 0.5 for both sampling sites), were also higher than typical values that were determined for diesel and gasoline emissions through dynamometer experiments.

Keywords BTEX · Industrial emissions · Petrochemical complex · Chemical industries · Urban pollution

Volatile organic compounds (VOC) play a key role in the chemistry of tropospheric ozone and other photochemical oxidants, and are also involved in the formation of secondary particulate matter (Finlayson-Pitts and Pitts 2000). Additionally, some VOC have been shown to have short- and long-term adverse health effects, in particular compounds such as benzene, toluene, ethylbenzene and xylene – represented by the acronym BTEX (da Silva et al. 2020).

A number of adverse health effects and diseases have been discussed in the literature, such as cancer, aplastic anemia, and possible neurological disorders, accompanied by symptoms of weakness, loss of appetite, fatigue, a state of confusion and nausea, caused by exposure to BTEX (Hadei et al. 2018). Benzene has been linked to leukemia and aplastic anemia and is classified as a Type I carcinogen (WHO 2010; IARC 2018), while ethylbenzene, which may cause respiratory failure, eye irritation, and neurological disorders (US EPA 2016), is classified as a suspected carcinogen (Hadei et al. 2018). Long-term exposure to toluene and xylene may also adversely affect the respiratory system as well as having neurological effects, including asthma and asthmatic symptoms, headaches, nausea, fatigue, nervous disorders and a state of confusion (PHE 2015).

According to the World Health Organization (WHO), there are no safe exposure limits for BTEX, especially for benzene (WHO 2000; 2010), but some guidelines and standards have been laid down for ambient air quality. The European Union (EU) Directive 2000/69/EC has set the permissible mean limits of benzene concentrations in ambient air at an annual average of 5 μg m^{-3} (EU 2000). In addition, the EU Directive 2008/50/EC set an "upper assessment threshold" of 3.5 μg m^{-3} for the annual average of benzene levels which cannot be exceeded for more than three separate years of the previous five years. Otherwise, fixed measurements should be performed to assess the ambient air quality in all the extended urban areas where this value has been exceeded (EU 2008).

In Brazil, the National Air Quality Standards (NAQS) (CONAMA 2018) are also applicable to the so-called "classical" pollutants classified in Group I by the WHO (2015): particulate matter with diameters less than 10 and 2.5 μm (PM_{10} and $PM_{2.5}$), carbon monoxide (CO), sulfur dioxide

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(SO_2), nitrogen dioxide (NO_2) and ozone (O_3). Although the NAQS were revised in 2018, benzene (included in Group II) and other VOC were not included, despite their adverse effects on human health, living beings and materials and the critical role they play in ozone production (Siciliano et al. 2020a). Several results obtained from experiments conducted in urban areas in Brazil have shown that benzene concentrations were often high and exceeded the value of 5 μg m^{-3}, in particular in the city of Rio de Janeiro (Corrêa and Arbilla 2007; Brito et al. 2015; Silva et al. 2016a, b; Siqueira et al. 2017; da Silva et al. 2020; Dominutti et al. 2020) and are very high in gasoline stations owing to their evaporative emissions (Oliveira et al. 2007) and tunnels (Machado et al. 2007; Martins et al. 2006).

The Metropolitan Region of Rio de Janeiro (MRRJ) is the second largest urban and industrial region in Brazil (IBGE 2020). Previous studies have shown that the southern and south-east areas are generally subjected to pollutants from vehicular emissions while the northern and north-east regions are affected by both vehicular and industrial sources (Siciliano et al. 2020b). The main goal of this study is (a) to measure the BTEX levels in the northern and north-east areas (b) to evaluate the BTEX ratios (c) to assess the potential health hazards of benzene and ethylbenzene exposure based on the collected data and (d) to make a comparison with data previously obtained from other areas in the MRRJ.

Materials and Methods

The MRRJ, in southeastern Brazil, comprises 22 towns and cities. As shown in Fig. 1, the south and south-east are flanked by the Atlantic Ocean, with Guanabara Bay to the east, and the remaining tropical rainforest occupies approximately 30% of the urban area. The city of Rio de Janeiro has 6.5 million people and is divided by the Tijuca Massif into the north and the south zones. The city of Duque de Caxias, in the north-east area, with a population of approximately 930,000 inhabitants, is one of the largest industrial zones in the country, with more than 150 chemical, pharmaceutical and petrochemical companies, a refinery which accounts for 80% of the production of lubricants and for the largest natural gas processing facility in Brazil and a thermoelectric plant (Termorio) with an installed capacity of 1058 MW (Petrobras 2021).

Samples were collected in the District of Irajá, a residential area located in the north of the city of Rio de Janeiro, approximately 25 km from the industrial zone, and in the district of Jardim Primavera (JP), in the city of Duque de Caxias. The main features of both locations and the sampling period are shown in Table 1.

BTEX samples were collected, as described in Method TO-15 (US EPA 1999), using evacuated stainless-steel

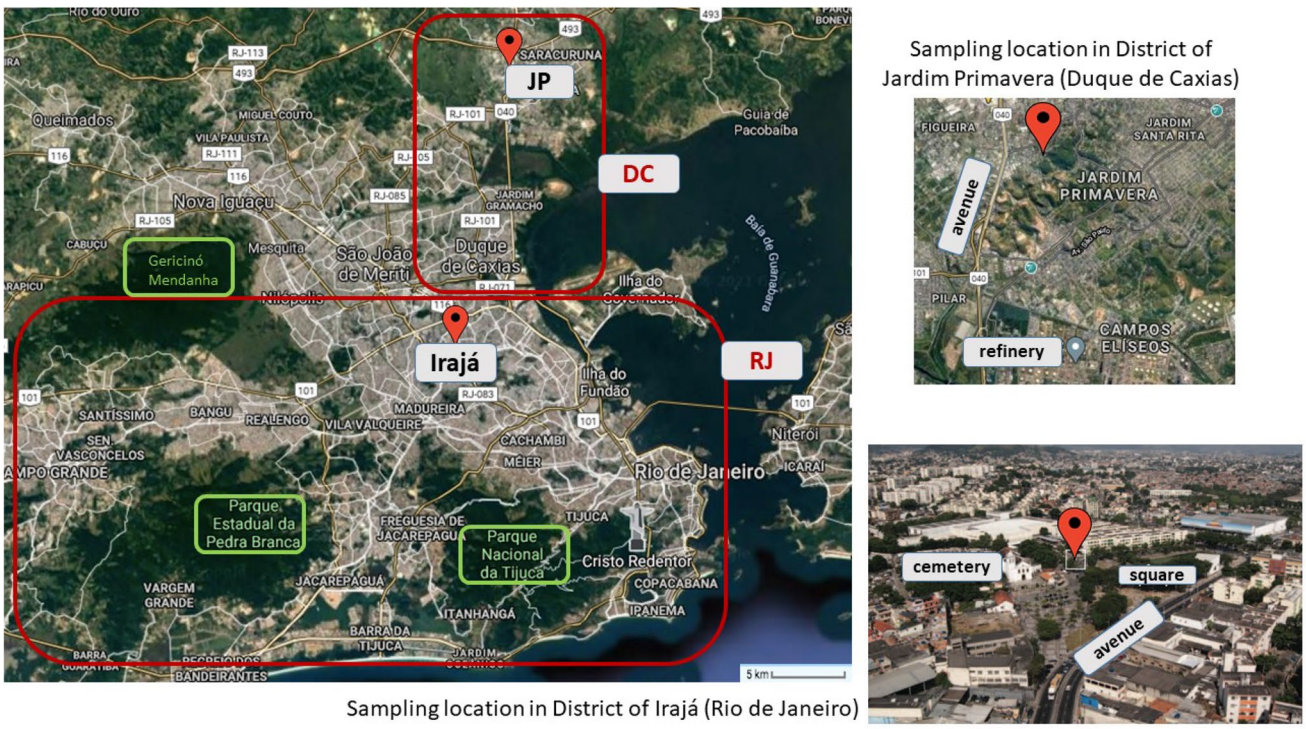

Fig. 1 Map of the Metropolitan Region of Rio de Janeiro showing the sampling areas: Irajá (Rio de Janeiro) and Jardim Primavera (JP, Duque de Caxias). Duque de Caxias (DC) and the city of Rio de Janeiro (RJ) are indicated as well as the mountainous areas (Parque Nacional da Tijuca, Parque Estadual da Pedra Branca and Gericinó-Mendanha Massif)

Table 1 Description of the sampling points: District of Tijuca in the northern area of the city of Rio de Janeiro and district of Jardim Primavera in the industrial zone of Duque de Caxias

Location	Coordinates	Description	Period and number of samples
District of Irajá, Rio de Janeiro	22°49′53″S; 43°19′36″W	Commercial and residential area. The sampling location is approximately 100 m away from two main streets with a constant flow of light and heavy-duty vehicles. The area has two supermarkets (with a steady flow of trucks), a cemetery, a taxi rank and a bus stop. The square hosts cultural events and leisure activities	24 Samples collected from 03/18/2019 to 06/03/2019, during the morning
District of Jardim Primavera, Duque de Caxias	22°41′00″S; 43°16′44″W	Residential, commercial and industrial area next to Campos Elíseos. In a radius of two kilometres, there are 8 chemical plants, 12 logistic warehouses and several gas stations. The sampling point is approximately 1 km away from a natural park (20 hectares) and 200 m away from Washington Luis Avenue where there is a steady flow of light and heavy-duty vehicles	20 Samples collected from 03/26/2019 to 05/03/2019, during the morning

canisters (Entech Silonite™) and flow restrictors (Entech Flow Controller CS1200E with a Silonite® filter), for 60 min, to achieve a final pressure of 1 atm. The canisters had been previously cleaned with a fully automate cleaning system (RM Environmental Systems Inc., model 960, CA, USA) and were considered to be clean if the residual concentrations of the compounds were lower than LOD. The samples were analysed within 72 h of sampling by means of a gas chromatograph with a thermal desorption and mass spectrometer detection (GC/MS/TD) system (Agilent, model GC 7890A, MS 5975C, CA, USA and Markes CIA Advantage, OH, USA) in accordance with the TO-15 Method. The analytical procedure consisted of transferring 500 mL of an air sample from the canister (flow rate of 20 mL min^{-1}) through a cold trap containing carbon molecular sieves (Markes U-T3ATX-2S) at 10°C to retain the organic compounds. Following this, the VOCs were thermally desorbed (300°C) and transferred to a DB-624 gas chromatographic column for separation (60 m × 0.32 mm × 1.80 µm). The carrier gas was He 5.0 (99.999%, ultra-high purity grade) at a constant flow of 2.5 mL min^{-1}. The BTEX were identified by comparison with the compounds of a standard reference mixture (Restek, 100 ppb, p/n 34429, PA, USA) and quantified using selective ion monitoring (SIM) of the most abundant ions, based on an external analytical curve. The curve was constructed in triplicate, at five concentration levels, and the calculated determination coefficients of all compounds were greater than 0.99. All the samples were measured in triplicate, and a difference of less than 25% was regarded as acceptable, as stated in the TO-15 Method. The compounds m- and p-xylene were not resolved under the chromatographic conditions and were reported as (m + p)-xylene. The LOD and LOQ, which were calculated from the standard deviation of the noise, were 0.2 ng and 0.6 ng, respectively, for all of the compounds. The method has been validated in the laboratory and employed in previous research studies (Silva et al. 2016a, b).

A code written in the R language, version 3.3.1 (R 2021) was used to construct the box plots. The air pollutant dispersion was simulated using HYSPLIT (2021). The model was run interactively on the READY website using the Global Data Assimilation System (GDAS) to obtain meteorological data with a 0.5 degree resolution (Stein et al. 2015). This model has been previously used in our laboratory to simulate the dispersion of volcanic ash released by the Puyehue volcano (Chile), smoke originating from industrial and wild fires (Silva et al 2018a) and the degree of pollutant transport within the MRRJ (Siciliano et al 2020b). The results for volcanic ash and fires (both in Rio de Janeiro and São Paulo) closely corresponded with the information from satellite data. There was also a close correspondence of simulated plume dispersion and wind roses calculated from ground-based meteorological observations within the MRRJ.

The risk of adverse health effects from BTEX exposure in ambient air caused by inhalation was assessed by following Hamid et al. (2019) and the EPA Technical Support Documents (NATA 2018; US EPA 1989). Cancer risk (CR) and non-cancer risk or hazard quotient (HQ), were calculated by means of Eqs. 1 and 2:

$$CR = EC \times IUR \quad (1)$$

$$HQ = EC / (RfC \times 1000) \quad (2)$$

where CR is the cancer risk, EC is the exposure concentration (µg m^{-3}), IUR is the inhalation unit risk (µg $m^{-3})^{-1}$,

HQ is the non-cancer risk or hazard quotient, and RfC is the reference concentration (mg m^{-3}).

The IUR and RfC values were obtained from the Integrated Risk Information System, IRIS database (US EPA 2019). In the case of benzene, the IUR value lies in the interval 2.2×10^{-6}–7.8×10^{-6} per µg m^{-3} (US EPA 2019), while for ethylbenzene, the IUR value is 2.5×10^{-6} per µg m^{-3}, according to the information of the Office of Environmental Health Hazard Assessment (OEHHA), California Environmental Protection Agency (CalEPA 2007).The non-cancer risk values for RfCs (benzene, toluene, ethylbenzene and xylene) are 0.03, 5, 1 and 0.1 mg m^{-3}, respectively (US EPA 2019). The ECs, based on chronic exposure via inhalation by adult non-workers for the residential receptor (Bari and Kindzierski 2017; Dutta et al. 2009; Li et al. 2014; Hamid et al. 2019), were estimated on the basis of Eq. 3:

$$EC = (CA \times ET \times EF \times ED)/AT \quad (3)$$

where EC is the exposure concentration (µg m^{-3}), CA is the contaminant concentration in air (µg m^{-3}), ET is the exposure time (h day^{-1}), 4 h for non-workers, EF is the exposure frequency (days $year^{-1}$), 350 days; ED is the exposure duration (years), considered as 24 and 70 years for HQ and CR, respectively; and AT is the average time (h), calculated as: ED in years × 365 days $year^{-1}$ × 24 h day^{-1}. These calculations involve several simplifications and considerations which have been described in the literature (US EPA 1989; 2009).

Results and Discussion

Total BTEX levels varied in the intervals 12.4–54.5 and 20.7–154.3 µg m^{-3} and median values were 40.8 and 37.5 µg m^{-3}, for Irajá and Jardim Primavera, respectively. The mean values were 38.4 ± 11.7 and 44.6 ± 29.3 µg m^{-3}, for Irajá and Jardim Primavera, respectively. The high standard deviation values are related to the ample scope of the data, especially in the industrial zone (JP), as shown in Fig. 2, and often observed in environmental assessments. This fact has also been noted for criteria pollutants (Siciliano et al 2020b; SMAC 2021) and BTEX determinations (Corrêa and Arbilla 2007; Siqueira et al. 2017; Silva et al. 2016a, b; da Silva et al. 2020) and is related to the variability of meteorological conditions affecting transport and deposition processes. These values are higher than recent determinations of BTEX concentrations in urban areas of Rio de Janeiro affected by vehicular emissions: (a) values in the interval 3–13 µg m^{-3} (Siqueira et al. 2017; Carvalho et al. 2020), (b) the mean value, 30.38 ± 26.15 µg m^{-3}, reported for the MRRJ (da Silva et al. 2020), and also (c) values 26.2, 18.1 and 13.7 µg m^{-3} obtained in Beijing in 2006, July 2008 and August 2008, respectively (Xie et al. 2008; Liu et al. 2009).

Individual BTEX concentrations determined in both sampling points are shown in Fig. 2 as box-plots. In both sampling sites, the most abundant aromatic compound was toluene. Benzene concentrations ranged in the intervals from 4.2 to 9.0 and 3.8 to 10.7 µg m^{-3} and the median values were 5.9 and 5.4 µg m^{-3}, for Irajá and Jardim Primavera, respectively. Since the sampling was only carried out in autumn and at irregular intervals, these values cannot be directly compared with the yearly average "upper assessment threshold" (3.5 µg m^{-3}), that was set by the EU Directive 2008/50/EC (EU 2000), but showed the importance of including benzene in the routine measurements of the monitoring stations installed in that area. However, there is no seasonal variation in the industrial activities carried out in the MRRJ and there is a narrow temperature range in Rio de Janeiro, with mean temperatures of 23°C and 27°C in summer and winter, respectively (SMAC 2021). Autumn and winter are characterized by lower rainfall which, in general, leads to higher primary pollutant concentrations, as shown by the data collected at

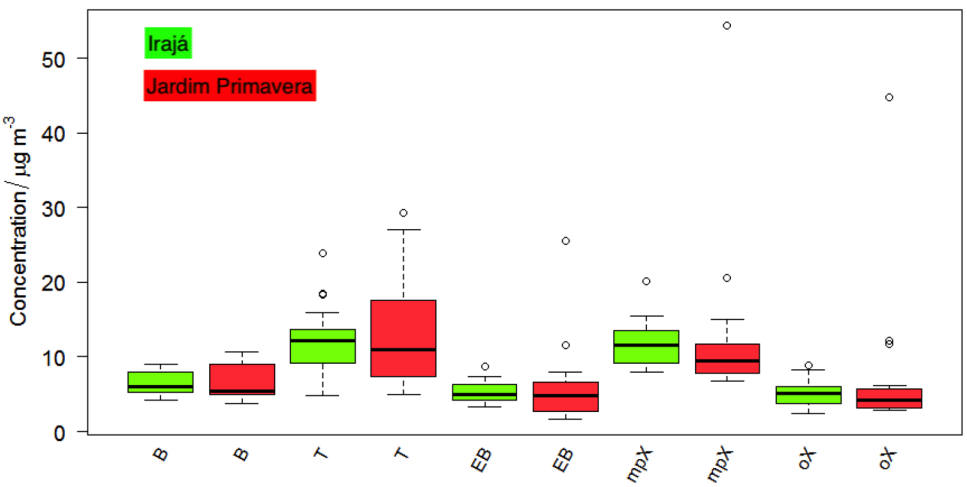

Fig. 2 Concentrations (µg m^{-3}) of benzene (B), toluene (T), ethylbenzene (EB), (m + p)-xylene (mpX) and o-xylene (oX) for samples collected in Iraja (green, N = 24) and Jardim Primavera (red, N = 20)

the automatic monitoring stations (SMAC 2021; DataRio 2021). Previous data obtained in Tijuca, an urban area in Rio de Janeiro where vehicular emissions are the main source of pollutants, showed that VOC concentrations in autumn and winter are approximately 2 and 1.5 times higher than in summer and spring, respectively (Silva et al 2018b). This means that the values reported in this work may reflect the most critical situations found in the city and the highest concentrations.

The mean values (6.4 ± 1.5 and 6.4 ± 2.2 μg m^{-3} obtained in Irajá and Jardim Primavera, respectively) are higher than the reported average for MRRJ, (3.4 ± 3.1 μg m^{-3}), determined in different locations where there are vehicular and mixed sources of emissions (da Silva, et al. 2020), similar to the value of 6.87 μg m^{-3}, determined in Beijing in 2006 (Xie et al. 2008) and higher than the values determined in Beijing in 2008 (Liu et al. 2009).

A Mann Whitney test (non-parametric independent samples, 95% confidence interval) was conducted to determine if samples collected at the two locations had the same distribution pattern. The results showed that all the *p* values were higher than the significance level (0.05) which suggests that, on the basis of this test, there is no reason to assume that the distributions are different. The calculated values were 0.70, 0.95, 0.34, 0.07 and 0.32 for benzene, toluene, ethylbenzene, (m + p)-xylene and o-xylene, respectively.

The benzene/toluene ratios were approximately 0.5 for both sampling sites (calculated by both mean and median values). This value is in the upper range reported by Rad et al. (2014) as characteristic of vehicular emissions in many urban areas around the world (0.3–0.5) and by Hamid et al. (2019) for three urban areas in Malaysia (0.25–0.45). In comparison with previous determinations for Brazil, this is a high ratio considering literature data: (a) the mean value (0.2) calculated for the MRRJ (da Silva et al. 2020), (b) the ratios determined, through dynamometer experiments, for diesel/biodiesel and gasoline (blended with 22% ethanol) -, 0.26 and 0.24, respectively (Corrêa and Arbilla 2006), (c) the values determined by Carvalho et al. (2020) in the interval 0.12 to 0.37 for several areas of Rio de Janeiro affected by vehicular emissions and (d) the mean ratio (0.32 ± 0.11) obtained in São Paulo, Brazil (Martins et al. 2008). Moreover, in a more recent study (Arbilla et al. 2019) based on three representative examples of Brazil´s motor vehicle fleet, ratios in the interval of 0.15–0.26 were determined by using E22 (gasoline with 22% ethanol). The relatively high benzene/toluene ratio is a sign of industrial emissions in the city of Duque de Caxias and air masses being transported to the city of Rio de Janeiro. The atmospheric lifetimes of benzene and toluene are approximately 9.5 and 2 days, respectively. These figures are large enough to allow benzene and toluene emissions from the industrial complex to reach the area of Irajá, prior to their decomposition.

Owing to the complex topography and the influence of the ocean and Guanabara Bay, the wind direction varies during the day with a predominance of north-easterly winds at night and during the morning and southeasterly winds after midday and during the evening. Forward and backward dispersion models were used to simulate the air masses that reached Irajá and also the dispersion of pollutant emissions from the industrial zone of Duque de Caxias, in particular Campos Elíseos. A representative simulation, obtained with the aid of HYSPLIT (2021), is displayed in Fig. 3. Clearly air masses arriving in Irajá during the morning, carry pollutants emitted in the chemical and petrochemical industrial zone, the refinery (District of Campos Elíseos in the city of Duque de Caxias) and also the main roads and highways (shown in Fig. 3). The sampling point in Jardim Primavera is approximately 3.5 km north of Campos Elíseos and is mainly affected by chemical plants and traffic (a significant part of the vehicular fleet is engaged in industrial and commercial activities).

Ethylbenzene and (m + p)-xylene correlations are also used to investigate emission sources. Ambient air samples and liquid fuel samples collected at various locations in 19 cities in Europe, Asia and South America (Monod et al. 2001) have demonstrated that these compounds largely originate from automotive sources in most urban environments. The extent of the BTEX emissions and ratios depends on the nature of the vehicular fleet and fuels. Ethylbenzene and xylene may also derive from solvent evaporation in chemical plants. Since the atmospheric lifetime of ethylbenzene is approximately 40 h, compared with approximately 12 h for xylene (Monod et al. 2001; Finlayson-Pitts and Pitts 2000), (m + p)-xylene/ethylbenzene ratios indicate the contribution of aged masses of air transported from the industrial area. The values calculated in this study (based on median concentrations) were 2.26 and 2.10 for Irajá and Jardim Primavera, respectively. These values are lower than the average (3.03), as well as the interval (2.8–4.6) observed by several authors for urban environments with vehicular emissions as major sources of pollution (Monod et al. 2001) which suggests that the aged air masses transported from the Campos Eliseos area were a significant factor.

Pearson correlation coefficients for BTEX compounds were calculated and are shown in Fig. 4 for both sampling points. A significant correlation ($r > 0.9$) was found between ethylbenzene and (m + p)-xylene in both sites, while the correlation between ethylbenzene and o-xylene was slightly higher in the industrial zone (Jardim Primavera). The correlation between xylene isomers was 0.99 in Jardim Primavera, which suggests the same source, and 0.87 in Irajá. Benzene/toluene coefficient was 0.70 in Jardim Primavera and all the other coefficients were < 0.7 in both sites suggesting several sources, in particular diesel and gasoline vehicles and industrial sources. In general, the correlations were weaker

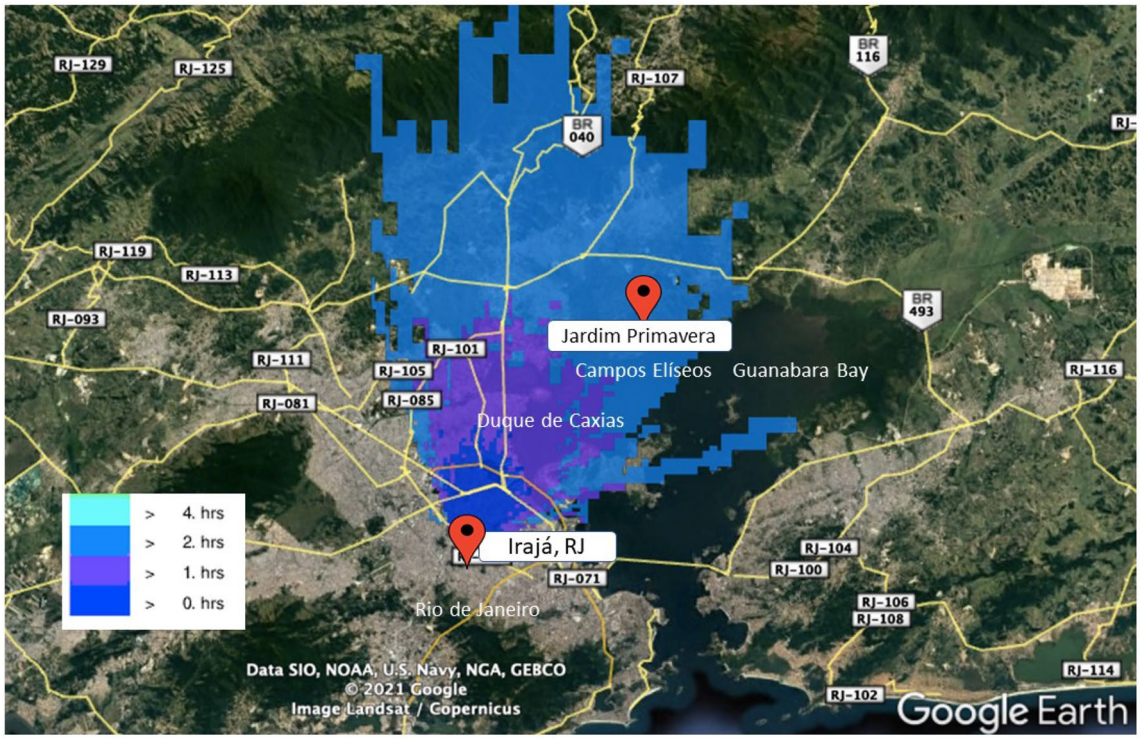

Fig. 3 Backward dispersion model for the air masses reaching Irajá at 9:00 h (local time) on April 16th 2019. Initial time: 5:00 h (local time). Source: HYSPLIT/NOAA/Google Earth

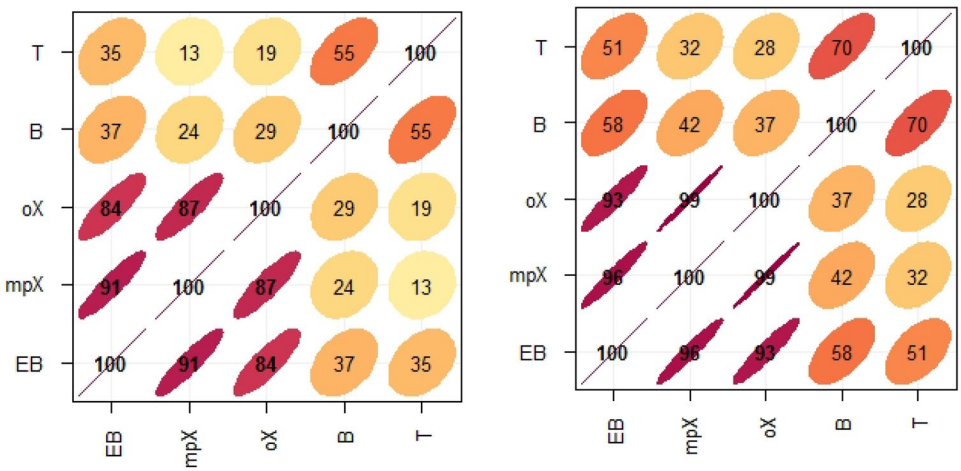

Fig. 4 Correlation matrix for BTEX compounds determined in Irajá (left) and Jardim Primavera (right)

in Irajá suggesting the influence of local sources and also air masses transported from the north and north-east, as shown in Fig. 3.

Cancer risk (CR) values calculated for benzene and ethylbenzene are shown in Fig. 5. Median and individual CR are $> 1 \times 10^{-6}$ for ethylbenzene and for benzene, when based on the minimum and maximum proposed IUR (US EPA 2019), which is regarded as an unacceptable limit and can pose a cancer risk to humans (US EPA 2009). Moreover, the calculated values are for individual compounds, and eventually the joint effects of other species could heighten the threat of the public´s exposure to air pollution. The HQ for all the compounds was < 1, which showed there were fewer non-cancer risks within the studied urban and industrial areas.

Unfortunately, data on cancer cases are not available to allow a further investigation of the adverse effects of the levels of these compound on public health. Nonetheless, in view of the results of this study and the WHO documents (2015), it is highly recommended that BTEX, in particular

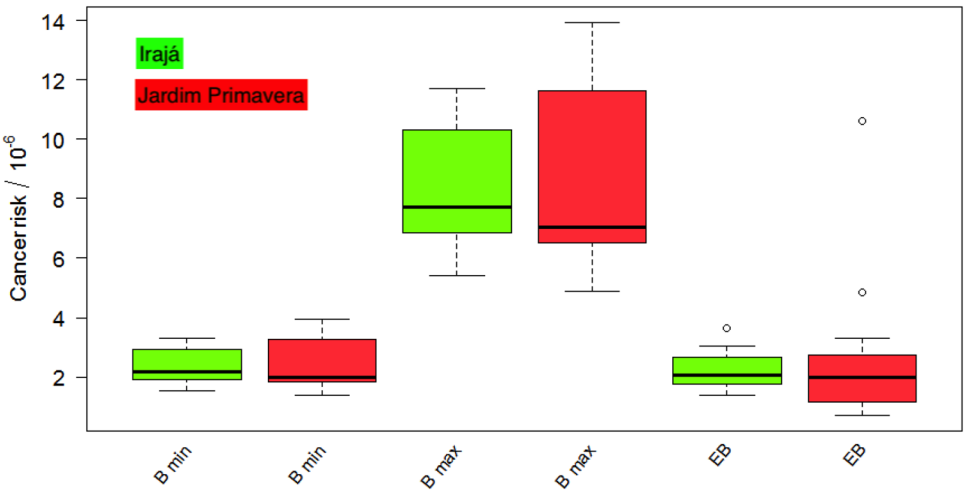

Fig. 5 Benzene (B) and ethylbenzene (EB) health risk estimation for cancer risk (CR) calculated for the sampling locations, Irajá (green) and Jardim Primavera (red). B_{min} and B_{max} stand for the lowest and maximum IUR values proposed for benzene

benzene, should be included within the criteria pollutants regulated by the NAQS.

The comparison of results obtained in this study for samples collected in the northern and northeast areas with previous studies for other regions of the MRRJ, indicates that in the southern area the major contribution to BTEX levels are vehicular sources, as determined by several authors, while in the northern area of the city of Rio de Janeiro and other cities of the MRRJ (northern and northeast areas) the contribution of industrial sources is a significant factor. This finding is a matter of great concern when discussing air quality management for the city of Rio de Janeiro, which is one of the twelve Latin American cities that are members of C40 Cities, a network of 97 of the world's leading cities committed to addressing the problem of climate change (C40 2021). Clearly initiatives to improve air quality should be discussed at a regional level considering all the cities in the metropolitan area, as well as all the sources of pollution (whether vehicular, industrial or natural emissions), and both meteorological and topographical factors.

CR values showed that benzene and ethylbenzene are potentially harmful to humans. Continuous monitoring of BTEX and other hazardous pollutants by the air quality local agencies is essential for a better understanding of the sources of emissions and potential public health risks. More funding for monitoring and control programs is needed, not only to reduce the economic impact of air pollution, but also to ensure the public are given the right to health and made aware of the underlying determinants of health. A suggestion for future work is to conduct a seasonal study, of both the daytime and nighttime, as well as to carry out research in more sampling locations.

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