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Road vehicle emission inventory of a Brazilian metropolitan area and insights for other emerging economies

Nara Angélica Policarpo[a], Carla Silva[b], Tâmara Freitas Aragão Lopes[c], Rinaldo dos Santos Araújo[a,c], Francisco Sales Ávila Cavalcante[a], Cira Souza Pitombo[d], Mona Lisa Moura de Oliveira[a,c,*]

[a] *State University of Ceará, Av. Dr. Silas Munguba 1700, 60.714.903 Fortaleza, Brazil*
[b] *Instituto Dom Luiz, Faculdade de Ciências, Universidade de Lisboa, Portugal*
[c] *Technology Federal Institute of Ceará, Av. Treze de Maio 2081, 60.040-531 Fortaleza, Brazil*
[d] *Department of Transportation Engineering, São Carlos School of Engineering, University of São Paulo, Av. Trabalhador São-Carlense, 400, São Carlos, SP 13566-590, Brazil*

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ABSTRACT

The vehicle fleet in the Ceará state has grown 180% over the last ten years. The growth of the resulting emissions is unknown in view of the expansion of this fleet in the greater Fortaleza Metropolitan Area (FMA). The largest fleet in the FMA is in the Fortaleza city itself, where flex fuel vehicles predominate (~30%). Flex fuel motorcycles increased significantly (greater than 800%) between 2010 and 2015. This paper aims to estimate the road vehicle emissions of carbon monoxide (CO), non-methane hydrocarbons (NMHC), aldehydes (RCHO), nitrogen oxides (NO_x), and particulate matter (PM) from the main road vehicle fleets of Fortaleza and its metropolitan area using a macrosimulation, bottom-up method, between 2010 and 2015. The results showed that road vehicle emissions of CO, NMHC and RCHO increased mainly by Otto cycle vehicles increase due to the introduction of flex fuel vehicles; however, the NO_x and PM emissions noticeable reduction is also a result of emission policies that seed the introduction of new technologies. In 2015, more than 70,000 tons of CO (21.2 ton/1000person), 8000 tons of NMHC (2.5 ton/1000person), 290 tons of RCHO (0.09 ton/1000person), 15,000 tons of NO_x (4.4 ton/1000person) and 600 tons of PM (0.2 ton/1000person) were emitted in the region under study. Comparing with other Brazilian regions, FMA emit higher levels of pollutants per inhabitant than the state of São Paulo and the state of Rio de Janeiro but lower levels than Porto Alegre city.

1. Introduction

Passenger and goods transport in Brazil is done mainly by road, and is highly dependent on fossil fuels. In 2014, the transport sector in Brazil consumed more than 60% of oil products (e.g. gasoline and diesel), and the road segment represents roughly 70% of the total energy consumption. Therefore, this sector is one of the main sources of urban air pollution, creating problems to the environment and to human health due to its combustion (MME, 2015; Morishita et al., 2006; Progiou and Ziomas, 2011; Silva et al., 2006; Souza et al., 2013; Zhang and Batterman, 2013). Ethanol biofuel blended with gasoline has been increasing since 2003 and since 2015 the gasoline sold has 27% bioethanol in it. The ethanol use reduces the oil dependence because it is produced by

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[*] Corresponding author at: LACEEMA/CCT/UECE, State University of Ceará, Applied Physical Sciences, Av. Dr. Silas Munguba, 1700, Fortaleza, Ceara CP: 60.714.903, Brazil.

E-mail address: mona.lisa@uece.br (M.L.M.d. Oliveira).

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endogenous sugar cane, and represents roughly 20% of the energy consumption in this road sector.

Fortaleza is the capital of the state of Ceará in northeastern Brazil, and has the seventh largest vehicle fleet in the country (387/1000 inhabitants) (DENATRAN, 2016). Currently, the Fortaleza Metropolitan Area (FMA), which includes 19 municipalities around the city of Fortaleza (including Fortaleza), is the sixth largest metropolitan area in Brazil (Cassiano et al., 2016; Cavalcante et al., 2009; IBGE, 2016). According to data from the Traffic State Department of Ceará (DETRAN-CE), the vehicle fleet of Ceará grew about 180% over the last ten years. In 2015, the FMA road vehicle fleet had more than one million vehicles, of which 75% were in Fortaleza itself (DETRAN-CE, 2017). This substantial increase in the road vehicle fleet has become an important source of urban air pollution in the region and, as well as result of this growth; there has been an increase in the number and size of traffic jams as well as ever increasing emissions of pollutants into the atmosphere. This scenario, which is common in large urban centers, is striving the need to carrying out studies concerning emissions (Cassiano et al., 2016a; Cavalcante et al., 2009; Schifter et al., 2005; Souza et al., 2013; Vivanco and Andrade, 2006), especially when considering atmospheric emission inventories as a management tool to improve the air quality of the local population. Besides, a monitoring network of air quality does not exist in FMA, and regarding data reported in literature information is scarce and limited to a couple of pollutants for the region (Cassiano et al., 2016b; Cavalcante et al., 2016; Rocha et al., 2016).

Several studies carried out in Brazil have pointed out a reduction of emissions with the gradual implementation of programs such as PROCONVE (Program for Control of Air Pollution by Automotive Vehicles) and PROMOT (Air Pollution Control Program for Motorcycles and Similar Vehicles) (CETESB, 2016; IBAMA, 2011; Réquia et al., 2015; Souza et al., 2013; Szwarcfiter et al., 2005; Ueda and Tomaz, 2011). Internationally, the guidelines of the EURO standards have, in particular, been a reference for the introduction of such regulatory policies worldwide (Cai and Xie, 2007; Jing et al., 2016; Tang et al., 2016). In Brazil, the current phases of PROCONVE/PROMOT are L6 for Otto cycle vehicles, P7 for Diesel cycle vehicles and M4 for motorcycles, as presented in the literature (DieselNet, 2016; IBAMA, 2011; MMA, 2013).

At present, Brazil follows the EURO V standard, which since January 2012 foresaw the implementation of the exhaust aftertreatment system, namely SCR – Selective Catalytic Reduction (Oliveira et al., 2011), for heavy diesel vehicles (i.e. buses and trucks). The technologies put into automotive vehicles, especially exhaust gas aftertreatment systems, and have been able to significantly reduce the emissions of polluting gases from vehicles over the years (Faiz, 1993). Gases such as carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2), particulate matter (PM), aldehydes (RCHO) and non-methane hydrocarbons (NMHC) are the main ones that have been reduced. However, these measures are still not sufficient. Consequently, road vehicle emission reductions are still being studied by the automotive industry, which is seeking technologies to lower emissions levels even further, since environmental standards are becoming increasingly restrictive (Aguiar et al., 2015; Elfasakhany, 2016; Lewtas, 2007; Ouyang et al., 2014; Shahir et al., 2015a, 2015b; Suarez-Bertoa et al., 2015; Szklo et al., 2005; Iodice et al., 2016; Zhu et al., 2014).

The first step to quantify the urban air pollution in cities is usually through a mobile source emissions inventory. Road vehicle emission inventories have been made in various large urban centers of Brazil (e.g. Porto Alegre – RS, Rio de Janeiro – RJ, São Paulo – SP, Vitória – ES) and around the world (e.g. Buenos Aires, London, Mexico City, New York, India, China, Norway). These measures have been taken by environmental agencies and other segments of the society in order to evaluate emissions and propose controls through public policies (Arriaga-Colina et al., 2004; CETESB, 2016; Cooper et al., 2014; D'Avignon et al., 2010; Fujita et al., 1992; López-Aparicio et al., 2017; MMA, 2013; Ozan et al., 2011; Schifter et al., 2005; Souza et al., 2013; Susilo et al., 2007; Venegas et al., 2011). An inventory of road vehicle emissions of air pollutants compiles fleet data over a given period in a given region. This has been shown to be a low-cost and fast-response tool that is extremely effective in assisting air quality models in urban centers (CETESB, 2016; D'Avignon et al., 2010; EPA, 2017; López-Aparicio et al., 2017; Pu et al., 2015), since detailed registration through monitoring networks is expensive (Réquia et al., 2015; Righi et al., 2013) and, according to Vormittag et al. (2014), there are only 252 monitoring stations in Brazil, which encompasses only 1.7% of Brazilian cities. In addition, estimated road vehicle emissions from inventories can be used as the input to a database for pollutant dispersion models (Jing et al., 2016; Tang et al., 2016) and, therefore, air quality forecasts (He et al., 2016).

There are two usual methodological approaches: the bottom-up (BU) and the top-down (TD). The former refers to using emission factors per vehicle category in a specific region/city and eventually aggregated to give a National perspective and the latter usually refers to spatially aggregated data (Nationally wise) disaggregated to provide insight on a specific region/city. This is also used when looking to the energy consumption methodologies (Horowitz and Bertoldi, 2015).

An extensive literature recommends the use of the bottom-up method used by the United States Environmental Protection Agency (EPA, 1994; Singer, 1998; Perugu et al., 2017), which has been shown to be effective in estimating vehicle emissions (Colvile et al., 2001; Cook et al., 2006; Pu et al., 2015; Righi et al., 2013; Wang et al., 2009; Zhu et al., 2014). This method uses data of the local fleet in circulation, combined with pollutant emission factors and annual mileage of vehicle (CETESB, 2016; MMA, 2013; Souza et al., 2013; Teixeira et al., 2008; Wang et al., 2009). Emission factors are one of the complex parameters that are acquired through models validated for emission measurements and driving cycle tests, and include vehicle characteristics, vehicle classification and age, fuel type, as well as emission control policies among other characteristics (Cassiano et al., 2016a; Cook et al., 2006; Gallus et al., 2016; Jing et al., 2016; Lawrence et al. 2016; Oduro et al., 2016; Pu et al., 2015; Schifter et al., 2005; Song et al., 2016). In addition, the emission factor is an indicator of emission control coming from the technological evolution of motor vehicles (CETESB, 2016). In the last years, emission factors are extensively acquired through the COPERT (COmputer Program to calculate Emissions from Road Transport) (Jing et al., 2016; Song et al., 2016). Based on Tier 3 approach (EEA, 2016), COPERT is the most complete bottom-up method since it accounts all type of exhaust and non-exhaust road vehicle emissions such as hot-running, cold-start, urban, rural, and highway emissions.

In China, for example, Tang et al. (2016) published estimates for the years 2006–2010, as the total number of vehicles increased from 14.5 million in 1999 to 78.1 million in 2010 – an increase of 437% in 12 years, due to the fast development of the Chinese economy. They concluded that gasoline vehicles are the largest emitters of pollutants such as CO and VOC (Volatile Organic Compounds), while Diesel cycle vehicles are the highest responsible for emissions of NO_x, $PM_{2.5}$ (fine particulate matter), PM_{10} (inhalable particulate matter) and BC (black carbon).

Studies of this nature have been practically non-existent in the northeast of Brazil, which is surprising since Fortaleza is the seventh largest capital of Brazil. Additionally, this study employs a useful, low-cost and no time-consuming tool to estimate vehicular emissions in any city compared with the usage of monitoring stations. Such estimates are crucial on the prediction of pollutant release trends by road transports, as well, to capture the effects of new public transport solutions such as BRT and the increase of biofuels in the road transportation sector such as ethanol up to 100% in volume blends. Recent efforts have been made to develop computational tools aiming to integrate inventory regional data with worldwide databases (Alonso et al., 2010). This will also be the basis for air pollution dispersion models.

The present study is the first inventory of road vehicle emissions for Fortaleza and five of the major towns in its metropolitan area with the largest road vehicle fleets for the years 2010–2015. Using a macrosimulation bottom-up method this research aimed to estimate the road vehicle emissions of CO, NMHC, RCHO, NO_x, and PM, considering the main categories of vehicles and types of fuels used in Brazil (i.e. gasoline, hydrated ethanol, and diesel).

2. Methodology

Air pollution concentration data is essential for mitigating the source of emissions and protecting human health. Ideally, this data could be obtained from air pollution monitoring stations that usually developing countries do not have. Other way is to gather on-board emission factors from real world driving but is a time consuming and expensive procedure. Therefore a first and cheaper approach of getting pollutant levels is by using simulation models with actual road transport data. Therefore, the methods described hereafter could be replicated for other developing countries and a sense of the uncertainty of the data is provided. For PM emissions the scope is only the tailpipe. So no brakes, tires and pavement wear are considered.

2.1. Characterization of the studied area

The state of Ceará, located in northeastern Brazil, has 184 municipalities and had a fleet of 2,828,433 vehicles in December 2015. Of this total, 1,370,303 vehicles belong to the Fortaleza Metropolitan Area (FMA), which encompasses 19 municipalities, including the capital Fortaleza (DETRAN-CE, 2017). Therefore, the FMA vehicle fleet corresponds to almost 50% of the total number of vehicles in the state. The present study aims to estimate the road vehicle emissions from 2010 to 2015 for the FMA which includes the city of Fortaleza itself and the five main cities with the major vehicle fleets, namely Caucaia, Maracanaú, Maranguape, Eusébio, and Pacajus (see Table 1). The inventory work was based on the main pollutants from the exhaust of automotive vehicles: CO, NMHC, RCHO, NO_x and PM, as reported in the literature (CETESB, 2015; Fujita et al., 1992; Schifter et al., 2005; Song et al., 2016; Souza et al., 2013; Tang et al., 2016; Wang et al., 2009; Zhu et al., 2014).

2.2. Estimation of emissions

The automotive fleet increase and the current patterns of consumption, among other factors are the cause of increasing emissions of air pollutants. In urban traffic, emissions of pollutants and fuel consumption are impacted according to characteristic traffic events, such as overtaking, traffic jams, sudden stops, type of road, pavement and driving style (moderate, aggressive), among others (Cassiano et al., 2016a; Chatterton et al., 2015). Furthermore, the emission factors (EF), that came from Environmental Sanitation Technology Agency of the state of Sao Paulo (CETESB) (2015), applied in the present research were not specific by each of these traffic events. This is a limitation of most macrosimulation models e.g. the COPERT software, which is able only to account for different average speeds in urban rural and highway segments (EEA, 2016). In addition, the use of COPERT software to estimate road vehicle emissions would be helpful to validate the results obtained for FMA during the studied years. On the other side, Smit et al. (2017) concluded that COPERT underestimated emission results by a factor of 7–37% according to pollutant type. Alonso et al. (2010) summarize the main macrosimulation models used in regional emissions inventories. All bottom-up approaches with emission

Table 1
FMA main urban areas and their characteristics in 2015.

Urban areas	Road vehicle fleet[a]	Population[b]	Motorization index (vehicles/1000 inhabitants)	Territorial area[b] (km^2)
Fortaleza	1,009,695	2,609,716	387	314.93
Caucaia	79,163	358,164	221	1,228.51
Maracanaú	61,629	223,188	276	106.648
Maranguape	22,523	125,058	180	590.873
Eusébio	20,414	51,913	393	79.005
Pacajus	20,322	69,877	291	254.636

[a] DENATRAN (2015).

[b] IBGE (2016).

factors taken from EPA or from average speed correlations (COPERT).

The authors opted to use the so called “bottom-up approach”, which means that specific emissions factors (in g/km) for each powertrain technology are used and up-scaled to account for total mileage driven and total number of vehicles of each technology to give a g/year overall fleet value. The bottom-up method adopted has been widely used by authors worldwide in an attempt to estimate emissions in a microscale level of the main pollutants from internal combustion engine road vehicles (CETESB, 2016; Cook et al., 2006; Baidya and Borken-Kleefeld, 2009; EPA, 1994; Huo et al., 2011; Jing et al., 2016; MMA, 2011, 2013; Souza et al., 2013; Szwarcfiter et al., 2005; Wang et al., 2009; Wills and La Rovere, 2010). According to Palacios et al. (2001), the bottom-up approach is mainly applied when local detailed data from road transport are known. Concerning the spatial resolution of the bottom-up method, emission allocation over the studied area only could be determined through geostatistical computational tool (Réquia et al., 2015; Tang et al., 2016). The method calculates the emissions considering the following variables: number of vehicles circulating in the region under evaluation, average annual distance traveled for each type of vehicle and the emission factor. These variables encompass three large data sets that are introduced in Eq. (1), resulting in the total amount of pollutant emitted in the year studied.

$$E_{\mathrm{C,F,P,Y}} = \left(\sum_{\mathrm{C}} \sum_{\mathrm{F}} \mathrm{Fc}_{\mathrm{C,F,Y}} \times \mathrm{IUadj}_{\mathrm{C,F,Y}} \times \mathrm{EF}_{\mathrm{C,F,P,Y}}\right) \cdot 10^{-6} \tag{1}$$

where E is the vehicle emission for the base year [ton/year], Fc is the circulating fleet disaggregated per model-year [number of vehicles], $\mathrm{IU}_{\mathrm{adj}}$ is the adjusted intensity of use [km/year], and EF is the emission factor [g/km]. Eq. (1) calculates road vehicle emissions according to vehicle category (C), fuel type (F), pollutant (P) and base year (Y) (Colvile et al., 2001; Schifter et al., 2005; Pu et al., 2015; Vivanco and Andrade, 2006).

2.2.1. Circulating fleet (Fc): Fortaleza city and FMA fleets

The data of the road vehicle fleet used to calculate the emissions was provided by the DETRAN-CE. The original data showed the category of the vehicle, type of fuel and year of manufacture of the vehicle (model-year), for each of the six FMA urban centers studied. The estimation of the emissions considered the following vehicle categories: cars, light commercials, motorcycles, which are Otto cycle vehicles (gasoline, ethanol, and flex fuel) and light commercial vehicles, trucks, and buses, which are Diesel cycle vehicles.

In order to calculate the road vehicle emissions from the fleets of flex fuel vehicles (i.e. cars, light commercials, and motorcycles), it is necessary to know the percentage of vehicles that choose to use gasoline and the percentage of those that opt to use ethanol. In Brazil, the choice of gasoline or ethanol is based on the ratio of the prices of these fuels on the market (Dominutti et al., 2016; Goldemberg et al., 2008), which is an extremely dynamic criterion. However, in the present study, we adopted the equation statistically adjusted by Rosa (2011). The fuel prices and their percentages calculated and applied to the flex fuel vehicle fleet are shown in the supplementary data (Appendix A).

The truck fleet was subdivided into the subcategories of: semi-light, light, medium, semi-heavy and heavy, according to the intensity parameters of use and emission factor that are specific for these subclasses (DNIT, 2008). In the present study, these subclasses were defined from statistical sampling carried out by Lopes (2016) and grouped as trucks and heavy-duty trucks.

In studies such as these, it is recommended to apply scrap functions to simulate the percentage of vehicles that cease to circulate for various reasons (e.g. mechanical deterioration, theft, accidents, and abandonment). This strategy makes the calculation of vehicle emissions of the actual circulating fleet more realistic (Schifter et al., 2005; Souza et al., 2013; Wills and La Rovere, 2010). This paper applied scrap functions, derived from statistical models, which included a Gompertz function (Eq. (2)), specific for Otto cycle vehicles, and a renormalized logistic function (Eq. (3)) for Diesel cycle vehicles (CETESB, 2016; Lopes, 2016; MMA, 2013).

$$\mathrm{S(t)} = 1-\mathrm{e}^{-\mathrm{e}^{[a+b(t)]}} \tag{2}$$

$$\mathrm{S(t)} = \left[1 + \mathrm{e}^{a\cdot(t-t_0)}\right]^{-1} + \left[1 + \mathrm{e}^{a\cdot(t+t_0)}\right]^{-1} \tag{3}$$

In Eqs. (2) and (3), S(t) is the fraction of vehicles in circulation, t is the age of the vehicle [years], a, b and t_0 are coefficients adjusted to the vehicle category as obtained in the literature (MMA, 2013; Souza et al., 2013).

Fig. 1 shows the scrap curves for the Otto and Diesel cycle fleets in the region under study. About 80% of the vehicle fleet is less than 10 years old, showing that it is a young fleet. This data is in agreement with the road vehicle fleets of other major cities in Brazil (Réquia et al., 2015).

The average age of a vehicle fleet assists in the evaluation of the emissions profile of the fleet, as well as indicating in which phase of PROCONVE/PROMOT it was manufactured (CETESB, 2016). Eq. (4) calculates the average age of a given fleet for a specific year based on the number of vehicles by model-year (Shibuya et al., 2015).

$$\mathrm{A_n} = \frac{\sum_{i=0}^{n} (\mathrm{Fc_i} \times \mathrm{A_i})}{\sum_{i=0}^{n} \mathrm{Fc_i}} \tag{4}$$

where $\mathrm{A_n}$ is the average age of the fleet in a base year [year], $\mathrm{Fc_i}$ is the number of vehicles in a given fleet disaggregated by model-year [number of vehicles], and $\mathrm{A_i}$ is the age of the fleet given by model-year of the vehicles [year]. Fig. 2 shows the fleet evolution by fuel type for Fortaleza and FMA from 2010 to 2015.

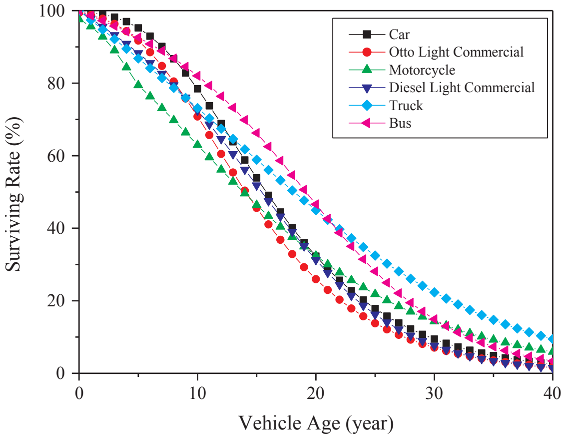

Fig. 1. Scrap curves for the Otto and Diesel cycle fleets.

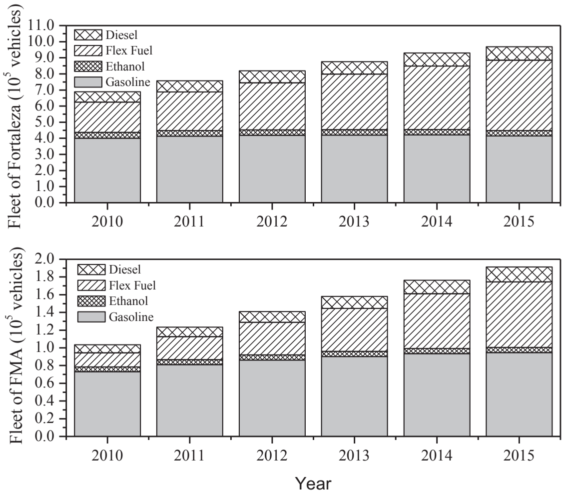

Fig. 2. Fleet evolution by fuel type for Fortaleza and FMA from 2010 to 2015.

2.2.2. *Intensity of use (IU)*

The intensity of use (IU) is the parameter that estimates the average annual distance traveled by the vehicles (CETESB, 2013; MMA, 2013; Souza et al., 2013). In the present study, this parameter was taken from the CETESB (2015), and they are defined as the reference values (IU_{ref}), but they require adjustment to local scenario. Thus, the IU obtained using Eq. (5) is referred to as the adjusted IU (IU_{adj}).

$$IUadj_{C,F,Y} = IUref_{C,F,Y} \times \frac{Cobs_{C,F,Y}}{Cest_{C,F,Y}} \tag{5}$$

where IU_{adj} is the adjusted intensity of use [km/year], IU_{ref} is the reference intensity of use [km/year], C_{obs} is the observed fuel consumption [L/year] and C_{est} is the estimated fuel consumption [L/year]. All these parameters vary depending on the category of the vehicle, the type of fuel and the base year.

The observed fuel consumption (C_{obs}) in Fortaleza and in each FMA municipality is the result of the linear interpolation of the volume of fuel sold annually in Ceará, and in Brazil as well, according to the ANP (National Petroleum Agency) (ANP, 2016), as shown in Fig. 3.

In order to obtain the estimated fuel consumption (Cest), Eq. (6) was applied (CETESB, 2015; MMA, 2013; Souza et al., 2013; Wills and La Rovere, 2010). The Cest value is the circulating fleet (Fc – number of vehicles) multiplied by the reference intensity of use (IUref – km/year) and divided by the autonomy of the vehicle (R – km/L), which is the distance a vehicle covers per liter of fuel. The autonomy values for all fleets were also taken from the database of the Brazil's, specifically of CETESB (2015).

$$Cest_{C,F,Y} = \sum_{F} (Fc_{C,F,P,Y} \times IUref_{C,F,P,Y}) \div R_{C,F,Y} \tag{6}$$

The Fc in Eq. (6) is the same as in Eq. (1). Thus, the road vehicle emissions were estimated for 16 categories of the Fortaleza and

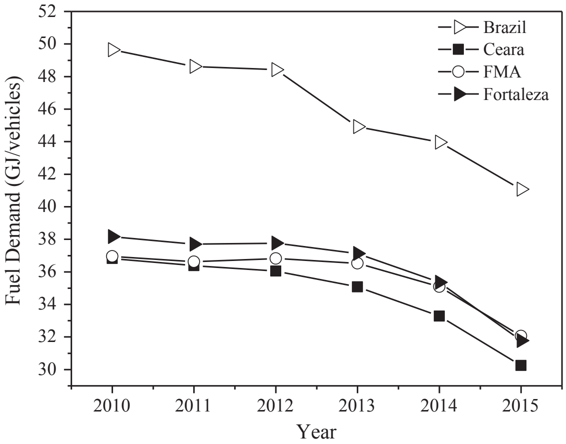

Fig. 3. Fuel demand for Brazil, Ceará, Fortaleza and FMA over studied years.

FMA fleets for six years (2010–2015); which were: gasoline passengers cars, gasoline light commercials, gasoline motorcycles, ethanol passengers cars, ethanol light commercials, gasoline/flex fuel passengers cars, ethanol/flex fuel passengers cars, gasoline/flex fuel light commercials, ethanol/flex fuel light commercials, gasoline/flex fuel motorcycles, ethanol/flex fuel motorcycles, diesel light commercials, diesel microbuses, diesel buses, diesel trucks and diesel heavy trucks.

2.2.3. Emission Factor (EF)

The Emission Factor (EF) is the mass of pollutant emitted by a vehicle when driven over a certain distance, usually expressed in g/km. The emission factors are typically determined through driving cycles for light-duty, heavy-duty and motorcycles. In the case of Brazil, such tests are conducted by vehicle manufacturers or importers that communicate EF values for CETESB. Thus, CETESB have been certified technical agent of government, publishes EF on its road vehicle emission report annually (CETESB, 2013, 2016; MMA, 2013; Rosa, 2011), and these results include all the vehicle categories studied in this paper. Emission factors for air pollutants vary according to the pollutant analyzed the vehicle category, the type of fuel and the model-year of the vehicle. In addition, factors such as accumulated mileage, conditions of use, maintenance status and vehicle driving style, in addition to environmental conditions, directly influence the emission of pollutants. Thus, it is recommended to add a correction factor to the EF, which is called the deteriorated emission factor (EF_{det}) (Jing et al., 2016; MMA, 2011). In this paper, the EF_{det} was only used for cars and light commercials of the Otto cycle, since, according to CETESB (2016), there is insufficient data for Diesel cycle vehicles and motorcycles to determine a deterioration factor. Therefore, the uncorrected EF was adopted for these categories (see the supplementary data in the Appendix B).

2.3. Validation of results

First the established spreadsheet model was replicated for São Paulo to compare with existing inventories and confirm that the model is well set. After, real world on-board measurements were used to compare with flex-fuel vehicle data with pure ethanol (E100) and gasohol (E27). Finally, for Fortaleza metropolitan area, the validation of results was done by comparing estimates with allocated per capita Nacional emissions. The existing Brazilian national inventory reports the emissions of the all country without spatial disaggregation by state or municipal district. It shows the emissions of the following pollutants: carbon monoxide (CO), nitrogen oxides (NO_x), non-methane hydrocarbons (NMHC), aldehydes (RHCO), particulate matter (PM).

As earlier pointed out, regional inventories of road vehicle emissions are usually reported as an outstanding policy instrument regarding air quality. The present study appears as the first inventory of the city of Fortaleza and its Metropolitan Area. In order to confirm that the model is well set it was applied in an already inventoried region, São Paulo, which uses the same bottom-up

Table 2
Model assessment for the state of São Paulo.

Parameters	CO	NMHC	RCHO	NO_x	PM
Reported emissions[a] (tons/year)	148,867	29,851[b]	583	18,725	75
Calculated emissions (tons/year)	137,217	13,486[c]	560	17,230	69
Error[d] (%)	7.8	54.8	3.9	8.0	8.1

[a] CETESB (2015).
[b] Exhaust, evaporative, and fueling emissions.
[c] Exhaust emissions.
[d] The error percentage stands for (calculated emissions-reported emissions)/reported emissions * 100%.

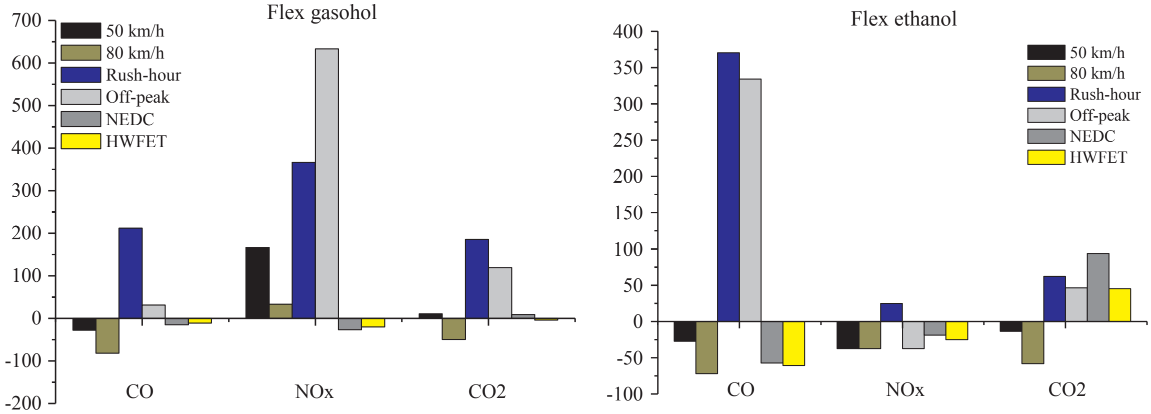

Fig. 4. Percentage differences from CETESB values for a PROCONVE L6 car.

approach. The results obtained were compared to the ones reported by CETESB (2015) and both of them are shown in Table 2. Data concerning gasoline car fleet of the state of São Paulo are available at CETESB website. It is important to note that for NMHC emissions reported by CETESB also included evaporative and fueling emissions to the total value presented in the table. Because of that, it was found a 54.8% of difference between the reported and calculated values. Thus one can conclude the model is well set.

Other aspect is the deviation between published values and real driving values, in actual vehicle use. CETESB data is based on dynamometer tests on UDDS – Urban Dynamometer Driving Schedule driving cycles. Real driving is not equal. Referring to a previous study (Cassiano et al., 2016b), the authors used on-board equipment data to monitor NO_x, CO and CO_2 instantaneous emissions (proportional to energy consumption, due to a combustion relation). Also simulate a flex fuel (PROCONVE L6) vehicle in several driving cycles such as NEDC and HWFET. Additional monitored data on rush hour, off-peak hour and average 50 km/h and 80 km/h trips were performed and the results are depicted in Fig. 4 as percentage differences from CETESB values, for NO_x, CO and CO_2.

Fig. 4 also gives an indication on how real driving emissions could be apart from those published. And of course this will be a source of inaccuracy at the inventory level. CO_2 emissions could be −60% to 190% different; CO −90% to 370%, and NO_x −40% to 630%. This way of validating the emission factors is time consuming and expensive because it implies several real runs, on-board equipment maintenance, calibration, and available drivers. Therefore it is not recommended for developing countries. It is more suited to countries/cities with more resources e.g. Madrid (Ariztegui et al., 2004; Vedrenne et al., 2016).

The air quality monitoring stations usually are also scarce or inexistent in such countries, for example, Brazil has only 1.3 monitoring stations per 1 million inhabitants as opposed to Germany that has 23 (data from 2013) (Réquia et al., 2016). The MMA document (MMA, 2011) shows the first Nacional inventory of emissions for Brazil giving overall emissions from historic data 1980–2009 and projections up to 2020. To have a kind of validation of our Fortaleza results we will represent the 2010–2015 prediction against our results divided per the number of vehicles.

3. Results and discussions

Table 3 shows the quantification and variation of road vehicle emissions of the CO, NMHC, RCHO, NO_x and PM pollutants for Fortaleza and FMA, according to our approach. The flex fuel vehicle fleet has been shifty increasing and therefore we compare our estimations of emissions with those of the Nacional emission inventory (MMA, 2013), as shown in Fig. 5.

The differences are on the order of 13–40% which is commonly found in such validation procedures. The research from Zhou et al. (2014) shows deviations up to 40%, from using municipal districts data to predict overall National emissions inventory. This was confirmed in Réquia et al. (2016) when applying the same methodology to Brazil (without distinguishing between gasoline or alcohol for the light and utility vehicles).

The average age of the road vehicle fleet (A_n) is a factor that strongly influences both the intensity of use of the fleet and the emission factors, since they are directly related to the characteristics of the vehicles (CETESB, 2016; Cook et al., 2006; D'Avignon et al., 2010; Jing et al., 2016; Shibuya et al., 2015; Souza et al., 2013). The older fleets are those of cars and light commercials that

Table 3
Total road vehicle emissions for Fortaleza and FMA in 2010 and 2015.

Year	Fortaleza (ton)					FMA (ton)				
	CO	NMHC	NO_x	PM	RCHO	CO	NMHC	NO_x	PM	RCHO
2010	53,92	6,35	12,999	596	196	9,978	1,212	2,396	107	31
2015	60,93	7,228	12,288	495	249	11,933	1,517	2,748	109	42
Change (%)	13.0	13.9	−5.5	−17.1	27.0	19.6	25.1	14.7	1.9	36.7

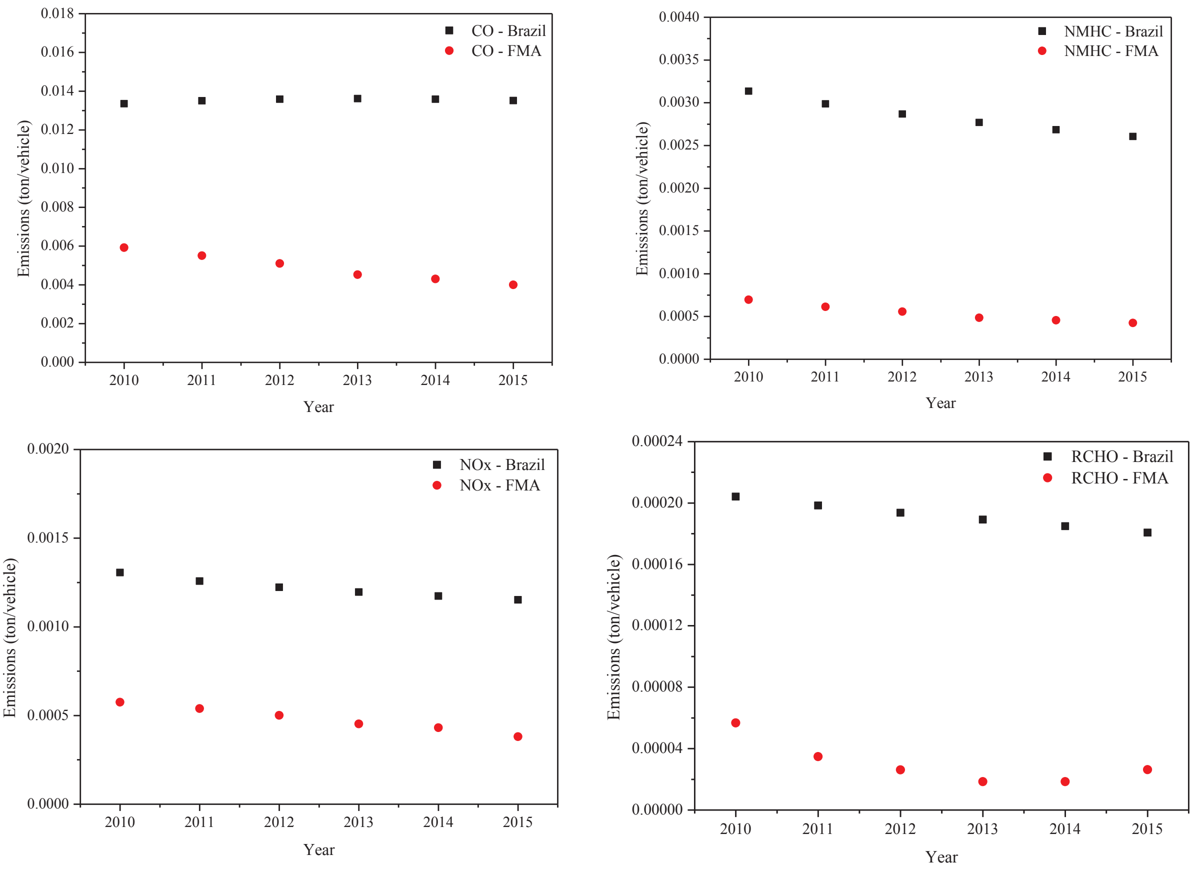

Fig. 5. Comparison between emissions per vehicles for flex fuel cars from Brazil and FMA.

run on ethanol (> 20 years old) and most of the new fleets are flex fuel (< 5 years old) vehicles. According to Shibuya et al. (2015), the age profile of Brazilian fleets in 2014 was: flex fuel vehicles were on average age 4 years old, gasoline vehicles were around 13 years old, and ethanol vehicles were in the 23 year old category. The fleets of Fortaleza and FMA are quite similar with the profile of the Brazilian fleet, with a general average age of 12 years old.

In general, in 2015, Fortaleza and FMA (5 towns) emitted more than 70,000 tons of CO, 8000 tons of NMHC, 15,000 tons of NO_x, 600 tons of PM and 290 tons of RCHO. Fortaleza emitted the most, contributing with values above 80% for all pollutants. Carbon monoxide (CO) was the pollutant with the highest emissions in the studied region, which is in agreement with the literature for other regions in Brazil (Souza et al., 2013; Tang et al., 2016; Ueda and Tomaz, 2011). Additionally, emission patterns by vehicle category and fuel can be seen in Fig. 6.

Table 4 presents some studies using the same method as the present research by base year, accordingly. The largest contributions of CO and NO_x emissions are due to gasoline and diesel fuels, respectively.

Analyzing the estimated vehicle emissions (Fig. 6), by vehicle category and fuel type we have:

(i) The gasoline passenger cars category was responsible for the largest emissions of CO in 2010 (~30%) in Fortaleza, while in the FMA, this category of vehicles and gasoline motorcycles contributed the same percentage. In Fortaleza, ethanol passenger cars stood out as they produced more than 30% of the CO emissions. The increase in emissions from this fleet, despite the 5.5% reduction in the number of vehicles, is due to the increase in the average age from 23 to 28 years old in the towns studied. The same was observed in 2015, the average age of the gasoline passenger cars fleet is much older than that of gasoline motorcycles, contributing to increase CO in FMA.

(ii) Gasoline vehicles emit the most NMHC emissions in Fortaleza and FMA, and especially the gasoline motorcycles that accounted for the highest emissions, both in 2010 (35%) and in 2015 (31%), as reported in studies by Tang et al. (2016).

(iii) The NO_x emissions increased by more than 20% for Otto cycle vehicles in 2015 (with the fleet increase), while these pollutants reduced by more than 10% for Diesel cycle vehicles, as a result of a more retributive legislation in the country and, consequently, the use of exhaust gas aftertreatment technologies for these engines. Approximately 50% and 60% of the NO_x emissions originate from the trucks and buses that circulate in the Fortaleza and FMA, respectively, in 2015. These values are similar to those found by Souza et al. (2013) and Tang et al. (2016).

(iv) The largest reductions were observed in PM emissions (> 15%) between 2010 and 2015 in Fortaleza (Table 3). As with NO_x,

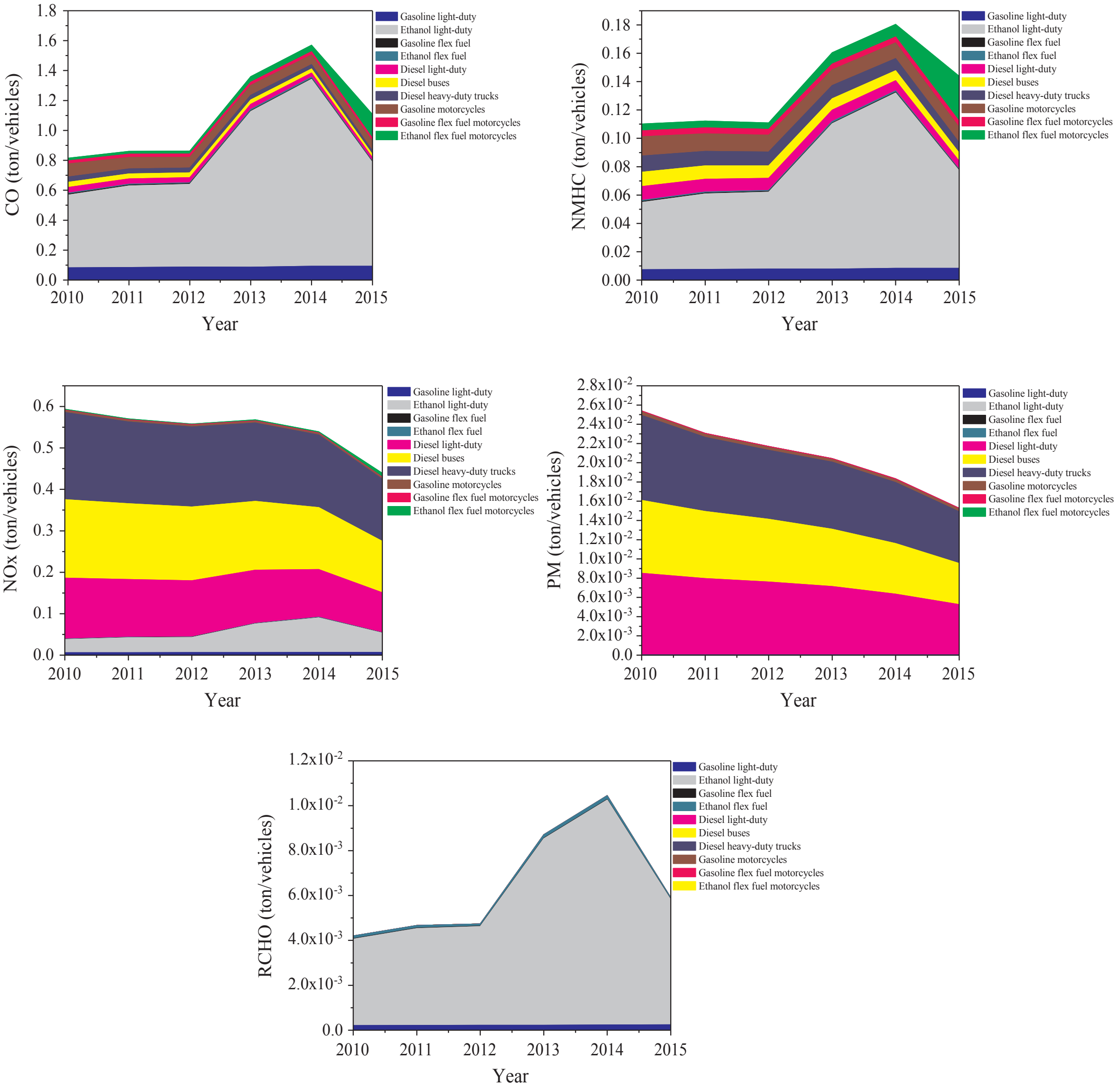

Fig. 6. Vehicular emissions by vehicle category and fuel for FMA between 2010 and 2015: CO, NMHC, NO_x, PM and RCHO.

Table 4
Per capita road vehicle emissions for some different regions in Brazil and worldwide.

Regions	Reference	Year	CO	NMHC	RCHO	NO_x	PM
			(ton/1000person)				
FMA[*]	This research	2015	21.2	2.50	0.09	4.40	0.20
SP[a]	CETESB (2016)	2015	3.00	0.40	0.01	1.20	0.03
RJ[b]	Souza et al. (2013)	2010	5.30	1.00	0.02	3.00	0.05
PA[c]	Teixeira et al. (2008)	2004	13.2	15.8	0.13	23.0	1.60
China	Tang et al. (2016)	2010	24.2	–	–	4.00	–
MCMA[d]	Hernández-Moreno and Mugica-Álvarez (2013)	2010	98.0	–	–	8.10	–
		2015	109.0	–	–	9.30	–

[*] Including Fortaleza City.
[a] SP – state of São Paulo.
[b] RJ – state of Rio de Janeiro.
[c] PA –Porto Alegre City.
[d] MCMA – Mexico City’s Metropolitan Area.

trucks and buses were the main sources of PM (~55% of total emissions in the period studied) in Fortaleza and FMA, and Diesel cycle vehicles were responsible for 89.0% of the total emissions in 2010 and 86.0% in 2015. Concerning inhalable PM, Rocha et al. (2016) monitored the air quality of three areas in the city of Fortaleza and stated that the greater PM concentration was related to higher traffic of Diesel vehicles in neighborhood. The authors registered a Diesel vehicle flow of about three vehicles per minute.

(v) Gasoline motorcycles were responsible for 11% of PM emissions in 2015 in Fortaleza, more than buses (~7%). Pacheco et al. (2017) drew attention to this fact and the need to implement PM emission limits for motorcycles, which so far do not exist in Brazil since PROMOT only controls the emissions of CO, NMHC, and NO_x from these vehicles.

(vi) The aldehydes pollutants directly reflect the expressive increase in the fleet of flex fuel vehicles that run on hydrated ethanol and/or gasoline, the latter fuel in Brazil also has a current average percentage of 27% of anhydrous ethanol in its composition (Dominutti et al., 2016; EPE, 2016). Ethanol vehicles (cars and light commercials) emitted more than 70% of the RCHO pollutants only in 2015, in both Fortaleza and FMA.

The reductions in road vehicle emissions are also due to the strict control of PROCONVE over the years, and is reflected in the gradual reduction of emission factors, such as PM (−14%) and NO_x (−10%) for the period, as well as the initiatives of local environmental agencies to inspect diesel vehicles.

In overall terms, gasoline vehicles emit the most carbon monoxide (CO) and non-methane hydrocarbons (NMHC), while ethanol vehicles emit the most aldehydes (RCHO) and diesel vehicles emit the nitrogen oxides (NO_x) and particulate matter (PM). These conclusions are also seen in the works of CETESB (2016), Cooper et al. (2014), Pacheco et al. (2017), Souza et al. (2013), Tang et al. (2016) and Ueda and Tomaz (2011).

The Otto cycle vehicles predominate in both Fortaleza and FMA. On average, they made up more than 90% of the total fleet between the years 2010 and 2015, and this was strongly supported with the introduction of flex fuel vehicles. These engines emit high amounts of CO, NMHC, and RCHO into the atmosphere when compared to Diesel cycle engines. Between 2014 and 2015, consumption of hydrated ethanol increased substantially (38.2% in Fortaleza and 57% in Ceará) when compared to gasoline consumption, which declined 5.5% in Fortaleza and 1.3% in Ceará. However, as can be seen in Fig. 3, gasoline consumption was much higher than hydrated ethanol. The reduction of emissions in recent years also results from the introduction of aftertreatment systems for the exhaust gases (Aguiar et al., 2015; Oliveira et al., 2011; Cassiano et al., 2016).

Total diesel consumption, as well as gasoline consumption, declined from 2014 to 2015 (see Fig. 3) contributing to the reduction of atmospheric pollutant emissions. One interesting fact is that diesel consumption in Ceará was lower than that of gasoline, while diesel consumption in the capital exceeds that of gasoline. The probable cause of this is the strong presence of diesel vehicles in Fortaleza (especially buses and trucks), while in the rest of the state there is a very significant fleet of motorcycles, increasing gasoline consumption.

In 2015, the total fleet of heavy-duty diesel trucks in Fortaleza and the FMA towns studied (Caucaia, Eusébio, Maracanaú, Maranguape and Pacajus) was more than 5000 vehicles, of which Fortaleza alone accounted for 78% of this fleet. Only 16% of this fleet was fitted with SCR-NO_x systems as of 2012 (ICCT, 2016). This fact reflects directly on the reduction of NO_x and PM emissions for this category.

To have some guidance on how much could be the future emissions moving forward with different fleet growth scenarios, Fig. 7 was drawn, for Fortaleza city. It shows two hypothetical scenarios where the flex fuel vehicles fed by E100 raises 4%/year, stagnating the BRT buses ("what if scenario #1") and other stagnating the light-duty vehicles and doubling the BRT service ("what if scenario #2").

As we can see, both scenarios guarantee similar levels of PM, NO_x and RCHO. If we look to the mobility capacity: usually the light-duty vehicles take one person which means an increase of the mobility for 102,000 people; the BRT capacity is 85 passengers which means an increase of the mobility for 680,000 people. Given this it would be advisable to promote policies that seed mode turnover.

4. Conclusion

In this paper, pollutant emission estimates of CO, NMHC, NO_x, PM, and RCHO from the main categories of road vehicles powered with gasoline, hydrated ethanol, gasoline-ethanol blends, and diesel were calculated using a macrosimulation, bottom-up method, including EPA and laboratory data emission factors. This inventory was based on a five-year period in the city of Fortaleza and its Metropolitan Area (FMA), and it refers to the first road vehicle emission inventory of the state of Ceará. Otto cycle vehicles were the largest emitters of CO, NMHC and RCHO, the latter emitted mainly by vehicles running only on hydrated ethanol; while Diesel cycle vehicles are the main responsible for NO_x and PM emissions.

In general, the CO, NMHC, and RCHO emissions increased from 2010 to 2015, while NO_x and PM emissions decreased in both Fortaleza and FMA. The largest reductions were recorded for PM (~17%) in Fortaleza. The Diesel cycle vehicle emission reduction is largely due to increasingly restrictive legislation over the years, such as PROCONVE and PROMOT (Euro V vehicles sold since 2012). Ethanol as a largely used biofuel in Brazil is causing the aldehydes (RCHO) emissions to increase steeply. This may be a major hurdle regarding air quality. The flex fuel vehicles running on 100% ethanol decreased in 2013 and 2014 due to fuel prices, but the minimum ethanol present in gasoline (the so called gasohol) has been increasing from 20 to 27% by volume.

The validation procedure consisted on comparing the emission factors with on-board real measurements where we found flex fuel vehicle real driving differences in the range of −40% to 630%. Also we acknowledge this is an expensive and time-consuming procedure but gives insights on real driving deviations from dynamometer tests which can be useful to air quality assessments.

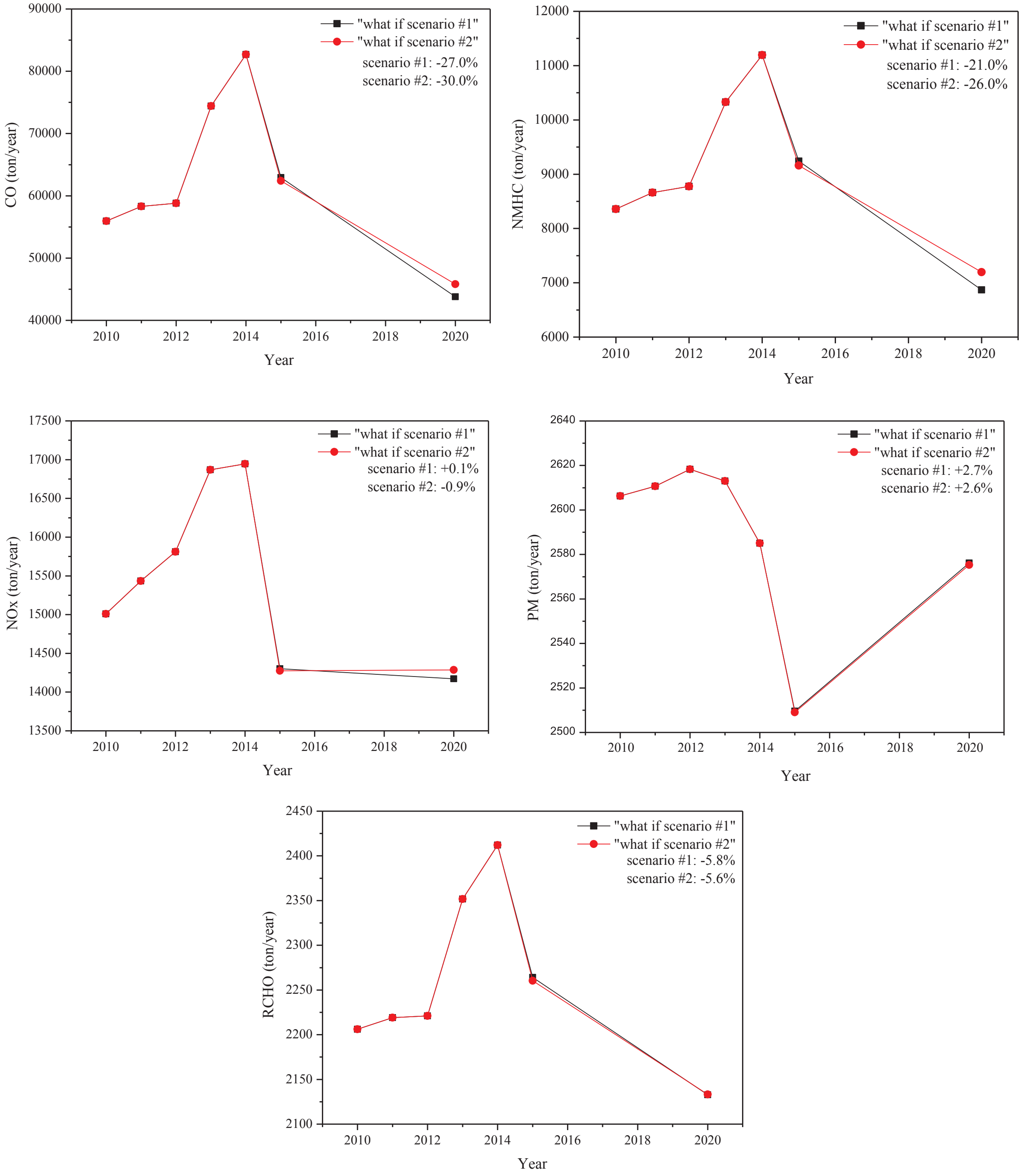

Fig. 7. Total emission trends from Fortaleza fleet for both “what if scenarios”.

National inventory scaled to the number of vehicles was compared against Fortaleza metropolitan area results. A typical deviation between 13 and 40% was found, as common to other development countries efforts to produce emission inventories, e.g. China.

The “what if” scenarios tried to investigate weather policies to increase the flex fuel vehicle fleet fed by E100 or policies to increase the BRT public transport system are preferable. It is quite interesting to notice that both “what if” scenarios will be accountable for similar PM, NOx and RCHO levels.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.trd.2017.12.004.

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