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A review of emissions and concentrations of particulate matter in the three metropolitan areas of Brazil

Marina Torres Pacheco[a, b], Magno Marcos Miotto Parmigiani[a, c], Maria de Fatima Andrade[d], Lidia Morawska[f], Prashant Kumar[a, g, *]

[a]*Department of Civil and Environmental Engineering, Faculty of Engineering and Physical Sciences, University of Surrey, Guildford GU2 7XH, United Kingdom*

[b]*Department of Production Civil Engineering, Federal Centre for Technological Education of Minas Gerais, Belo Horizonte, 30.421-169, Brazil*

[c]*Department of Civil Engineering, Federal University of Rio Grande do Sul, Porto Alegre, 90.035-190, Brazil*

[d]*Department of Atmospheric Sciences, Institute of Astronomy, Geophysics and Atmospheric Sciences, University of São Paulo, São Paulo, Brazil*

[f]*International Laboratory for Air Quality and Health, Queensland University of Technology, 2 George Street, Brisbane 4001, QLD, Australia*

[g]*Environmental Flow (EnFlo) Research Centre, Faculty of Engineering and Physical Sciences, University of Surrey, Guildford GU2 7XH, United Kingdom*

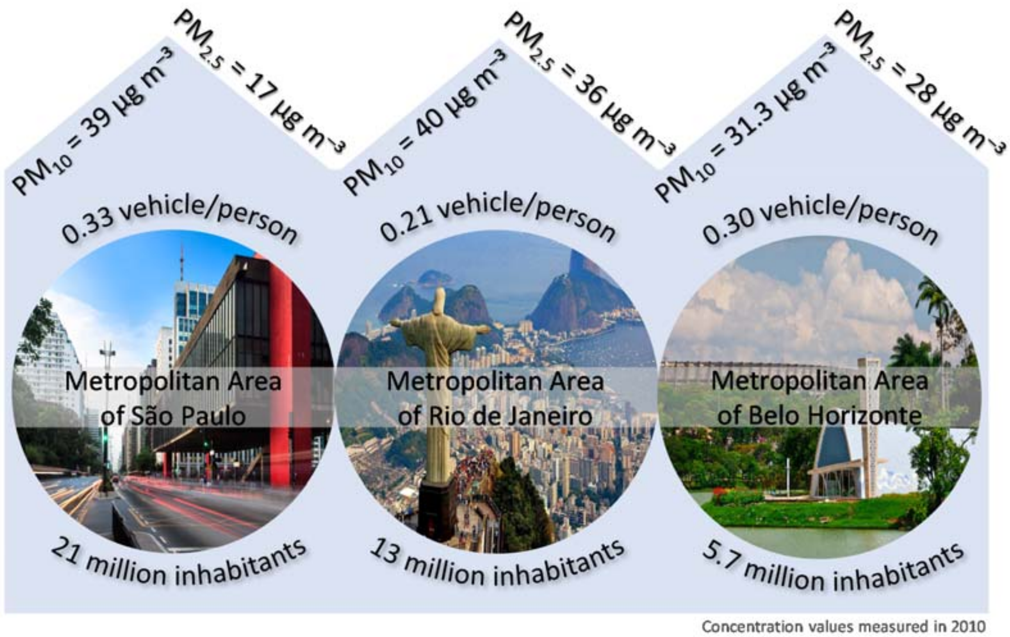
Research highlights

- Emissions and concentrations in three Brazilian metropolitan areas are reviewed
- In 2014, vehicular emissions contributed 34% of the total Brazilian $PM_{2.5}$ emissions
- Non-exhaust sources could represent 90% of the Brazilian PM emissions from 2020 onwards
- Brazilian cities have relatively relaxed guidelines of PM concentration compared with WHO
- Studied metropolitan areas showed up to 3-fold high concentrations above WHO guidelines

[*]Corresponding author. Address as above. *Tel.: +44 1483 682762; Fax: +44 1483 682135;*

E–mail addresses: P.Kumar@surrey.ac.uk, Prashant.Kumar@cantab.net

Graphical abstract



ABSTRACT

We critically assessed numerous aspects such as vehicle fleet, type of fuel used in road vehicles, their emissions and concentrations of particulate matter ≤2.5 µm ($PM_{2.5}$) and ≤10 µm (PM_{10}) in three of the most polluted metropolitan areas of Brazil: the Metropolitan areas of São Paulo (MASP), Rio de Janeiro (MARJ) and Belo Horizonte (MABH). About 90% of the Brazilian LDVs run on ethanol or gasohol. The HDVs form a relatively low fraction of the total fleet but account for 90% of the PM from road vehicles. Brazilian LDVs normally emit 0.0011g (PM) km^{-1} but HDVs can surpass 0.0120g (PM) km^{-1}. The emission control programs (e.g., PROCONVE) have been successful in reducing the vehicular exhaust emissions, but the non-exhaust vehicular sources such, as evaporative losses during refueling of vehicles as well as wear from the tyre, break, and road surface have increased in line with the increase in the vehicle fleet. The national inventories show the highest annual mean $PM_{2.5}$ (28.1µg m^{-3}) in the MASP that has the largest vehicle fleet in the country. In general, the PM_{10} concentrations in the studied metropolitan areas appear to comply with the national regulations but were up to ~3-times above the WHO guidelines. The current Brazilian air quality standards are far behind the European standards. There has been a progress in bringing more restrictive regulations for air pollutants including PM_{10} and $PM_{2.5}$ but such steps also require suitable solutions to control PM emissions from motor vehicles and mechanical processes.

Graphical abstract

Keywords: Particulate matter; Vehicular emissions; PM concentrations; Air quality standards; Brazilian metropolitan areas

1. Introduction

Air quality deterioration is a major environmental concern in many modern cities, including urban centres of Latin America (Onursal and Gautam, 1997; Park and Kim, 2005). Amongst several pollutants capable of affecting air quality, particulate matter (PM) is one of the main pollutants showing the greatest impact on human health (Heal et al., 2012). The PM is commonly divided into coarse and fine particles (Miranda et al., 2011). Fine particles comprise mass concentration of particles with an aerodynamic diameter equal to or less than 2.5 µm ($PM_{2.5}$) whereas coarse particles include diameters ranging from 2.5 to 10 µm ($PM_{2.5-10}$) (Mariani and de Mello, 2007). The scope of this article includes both the fine and coarse particles, which together are referred to as PM_{10}.

Vehicular emissions are a significant source of PM, especially fine particles (Onursal and Gautam, 1997). Over the past few years, an increase in motor vehicles and industrial activities in urban areas have worsened the atmospheric contamination by PM; such conditions give rise to numerous public health problems (Faiz et al., 1996; Fernández et al., 2000). Exposure to elevated levels of PM can have severe impacts on human health, and in particular cause respiratory and cardiovascular diseases and premature deaths (Lanki et al., 2006; Saldiva et al., 1994). Thus, PM has become an important matter due to its impact on health and the environment (Jimoda, 2012), resulting in the formulation of guidelines and standards by international bodies and local governments and to protect public health. While the threshold values help in safeguarding public health, exposure to pollutants could still affect human health at the PM concentrations below the levels recommended by the air quality guidelines (Baldauf et al., 2009; Kumar et al., 2015; Martins et al., 2009). Regarding climate change, $PM_{2.5}$ in many Brazilian cities is characterised by high fraction of carbonaceous compounds that have a direct impact on the radiative balance, and the inorganic fraction, on the cloud microphysics processes (Miranda et al., 2011).

During the last decade, developing countries such as Brazil have been facing rapid economic growth that has triggered an increase in the vehicle fleet and air pollution in urban centres, especially in the capitals and their metropolitan areas (Faiz et al., 1996; Miranda et al., 2011). Three Brazilian metropolitan areas – Metropolitan Areas of São Paulo (MASP), Rio de Janeiro (MARJ) and Belo Horizonte (MABH) – located in the southeast region of the country, are noteworthy for their economic importance. Together, these account for ~60% of the national Gross Domestic Product (IBGE, 2012). Moreover, these are the most populated regions in Brazil, concentrating 12% of the national vehicle fleet and a large number of industries. For that reason, we have selected these three regions as a focus of this review article.

The countries in South America were colonized by Spanish and Portuguese. The urban settlements followed the style of European towns with the buildings and important houses concentrated in the central part of the cities. The growth of the periphery of the cities in South

America was motivated by the access to jobs and public services. This resulted in an increase in commuting time, with direct effects on both times lost during the commute and excessive emissions of pollutants. The spatial distribution of the vehicles indicates that the oldest cars (comprising most of the high-emitters) are usually concentrated in the periphery of the Brazilian cities (CETESB, 2016). Many other countries in South America, Africa, and Asia are facing similar urban transition.

Monitoring of air pollutants is very uneven in the Latin America countries, and usually restricted to the large cities (Clean Air Institute, 2012; UNEP, 2016). Some countries do not have their own air quality standards. The importance of the evaluation of air quality in these emerging cities is important to protect the health of their inhabitants. For example, Romieu et al. (2012) described the results of the ESCALA Project (Estudio de Salud y Contaminacion del Aire en Latinoamerica) that considered cities in Brazil, Chile, and Mexico. For most cities, they found that PM_{10} concentrations were associated with increased mortality from cardiopulmonary, respiratory, cardiovascular, cerebrovascular-stroke, and chronic obstructive lung diseases.

The discussion presented here is focused on the three biggest metropolitan areas of Brazil, which are undergoing fast and disorganised growth, and face challenges in implementing regulations for air pollution emissions from mobile sources. Despite the historical and local characteristics, the trajectory of the urbanization growth in the Brazilian cities can be viewed as a representative example of cities in developing countries, since Brazil is the largest country in Latin America.

The MASP is the largest metropolis in Brazil, with a population of more than 21 million inhabitants; this includes 39 cities covering a surface area of approximately 8000 km² (IBGE, 2015; Miranda et al., 2011). The biggest and most important city is São Paulo, which is the capital of the state of São Paulo. This region presents a tropical climate with wet (October to April) and dry (May to September) seasons; January and June are the hottest and coldest months of the year, respectively (Miranda et al., 2011).

The second largest Brazilian metropolis is the MARJ, with the most relevant city being Rio de Janeiro, which is also the capital of this state and a place for 2016 Olympic Games. This metropolitan area has almost 13 million inhabitants distributed throughout 21 cities across a surface area of ~6800 km² (IBGE, 2015; Miranda et al., 2011). February and July are the hottest and coldest months in the MARJ, respectively. The MARJ is also characterised by a tropical climate with wet (January to April) and dry (December) seasons. The city of Rio de Janeiro also presents a local topography that, when combined with factors such as unplanned land use, sea breeze circulations and the existence of the Guanabara Bay, contributes to an irregular dispersion and distribution of pollutants in the area (Onursal and Gautam, 1997; Soluri et al., 2007).

The MABH is the third largest metropolis in Brazil. Belo Horizonte is its main city and the capital of Minas Gerais state. The MABH has 5.7 million inhabitants distributed throughout a surface area of ~9500 km², which is divided into 34 cities (IBGE, 2015; Miranda et al., 2011). The region is also characterised by a tropical climate with wet (October to March) and dry (April to September) seasons; January and July are the hottest and the coldest months of the year, respectively. The region has a topography characterized by mountains that determine the circulation of the pollutants.

One of the major differences between these metropolitan areas and other well-known cities elsewhere in the world is the incentive of the Brazilian governmental for the use of biofuels (such as ethanol, gasohol, and biodiesel) in the national vehicle fleet. The use of biofuels is effective in reducing PM emissions from vehicles due to a more efficient combustion process (Aparicio et al., 2014; Kousoulidou et al., 2008). Considering the (i) importance of these cities to Brazil, (ii) the influence of vehicles and fuels on PM emissions, (iii) the effect of meteorological conditions on PM concentrations, and (iv) the consequences of PM levels on public health, we set to review the emissions and concentrations of PM in those metropolitan areas in order to evaluate their air quality conditions and exceedances of PM levels in comparison with the national and the World Health Organisation (WHO) guidelines.

2. Traffic fleet and fuel used

There were approximately 42.6 million vehicles in the traffic fleet of Brazil by the end of 2015 (SINDIPEÇAS, 2016). Amongst the three major metropolitan areas in Brazil, the MASP has the largest traffic fleet, comprising more than 7 million vehicles, followed by the MARJ with 2.7 million vehicles, and the MABH with 1.7 million vehicles (IBGE, 2015). This traffic fleet is divided into three main categories: light-duty vehicles (LDVs), heavy-duty vehicles (HDVs) and motorcycles. Here, HDVs are referred to the vehicles with more than 8501 lbs of Gross Vehicle Weight Rating (GVBR); all of them run on diesel with 8% biodiesel. LDV are mostly the passenger cars with GVWR <10000 lbs.

LDVs comprise ~85% of the actual fleet in the MASP, MARJ and MABH whereas motorcycles and HDVs represent ~12% and 3% of the total vehicles in these areas, respectively. The vehicle fleet has changed considerably over the past decade in the study regions (IBGE, 2015). For example, the traffic fleet in MASP, MARJ and MABH have increased by 73%, 77% and 117%, respectively, in 2015 from the 2005 levels. Table 1 presents the characteristics of the fleet, population and transport modes of the three cities.

The sales of new passenger vehicles in Brazil have grown at an annual average rate of 10% between 2003 and 2013 and, since then, the country has been considered as the fifth leading market in the world for sales of new passenger automobiles (Posada and Façanha, 2015). The state of São Paulo is the major consumer of new vehicles (LDVs and HDVs) in Brazil, representing ~27% of the total sales that occurred in 2015 (ANFAVEA, 2016). At second and third positions were the states of Minas Gerais and Rio de Janeiro, with participation in sales of 13.2% and 7.7%, respectively (ANFAVEA, 2016). The selling of new vehicles has decreased by 27% compared with April 2016 to the same month in 2015.

The Brazilian Automotive Industry Association (ANFAVEA) estimates that 88% of the new LDVs sold in 2015 were flex-fuel vehicles. The flex-fuel vehicles were introduced in Brazil in 2003. Since then, they have gained popularity due to their market availability and attractive ethanol price (Nogueira et al., 2015). Flex-fuel vehicles can run on either ethanol or gasohol, which is a blend of gasoline with a proportion of about 25–27% of ethanol (Kumar et al., 2016). The majority of the Brazilian LDV fleet runs on ethanol, gasohol or compressed natural gas whereas the HDVs run on bio-diesel (Andrade et al., 2010). For example, only 3% of the LDVs in the MASP used diesel as a fuel in 2014 (CETESB, 2015). Such a small proportion contrasts with other megacities in the world (e.g. London and Delhi) where a great portion of the LDVs run on diesel (Kumar et al., 2015).

Table 1 shows the number of vehicles that run on different types of fuel in the studied metropolitan areas, and other characteristics related to the choice of transport mode and number of trips by passengers cars, buses, bicycles and metro in the three cities. The National Department of Transportation (DENATRAN) reported that the flex-fuel cars composed ~34% of the MASP vehicle fleet in December 2015. For the MARJ, this proportion was equal to 32% of the total fleet, compared with 52% in the MABH. Gasohol-driven vehicles still comprise the majority of the traffic fleet in the MASP and MARJ, 51% and 39% of their total fleets, respectively (DENATRAN, 2015). Conversely, gasohol vehicles comprise the second largest portion of the MABH fleet, representing 37% of all automobiles (DENATRAN, 2015).

About 40% of the vehicle fuel used in all Brazilian cities is ethanol (Anderson, 2009), which was introduced in the 1920s with its use accelerating after the government created the Brazilian National Alcohol Program (PROALCOOL) in 1975 (Anderson, 2009). The implementation of the PROALCOOL Program was divided into two phases. The first phase intended to increase the percentage of ethanol in the gasohol blend up to 20% nationwide; the second phase concentrated on providing 100% hydrous ethanol as a fuel for LDVs (Onursal and Gautam, 1997). The main goal of the PROALCOOL Program was to stimulate the use of domestic sources and reduce the dependence of the country on imported petroleum (Rosillo-Calle and Cortez, 1997).

In search of environmental-friendly alternatives, the Brazilian government has also encouraged the use of biodiesel as a fuel for HDVs (Nogueira et al., 2015). The current projections show that biofuels will address to 30% of the total Brazilian vehicle fuel demand by 2030 (Schmitt et al., 2011). According to the Brazilian Law No. 13.033 that was implemented from September 2014 onwards, all types of diesel must be blended with at least

7% of biodiesel, with a likely increase in the proportion of up to 27.5% of biodiesel in the subsequent years (Corotti et al., 2015). Furthermore, the commercialisation of S500 (diesel fuel with 500 ppm of sulphur content as a maximum limit) commenced in 2005 in all the metropolitan areas of the country for reducing emissions of sulphur content emitted by diesel vehicles. Due to the creation of new national regulations (CONAMA Resolution Nos. 315 and 403(CONAMA, 1990)) that intended to restrict emission rates, a diesel fuel with 10 ppm of sulphur content (S10) was introduced to the market in 2012 (Nogueira et al., 2015).

Brazilian diesel engines, fuels, and technical standards are far behind the European standards; this is one of the reasons why Brazilian buses and trucks running on diesel fuel emit 200-times more sulphur to the atmosphere than the European vehicles (Nogueira et al., 2015). There is a movement in Brazil to launch a new phase of PROCONVE with a focus on the HDVs that are responsible for most of the fine particle emissions in the urban areas. In contrast, the use of biodiesel in diesel blends can help reducing this sulphur content and the PM emissions (TransLink, 2006), and conversely increase the emission of nitrogen oxides (Nogueira et al., 2015). Therefore, it is important to analyse whether the use of biofuels is capable of decreasing pollutant emissions and improving air quality.

3. Meta-analysis

The urbanization of the metropolitan areas in Brazil follows a pattern of growth similar to that seen in other South American cities. The urban transition began much earlier in Brazil than in Asia and Africa. The United Nations reported that, as of 1950, 36% of the population of Brazil lived in urban areas, compared with 15% in Africa and 17% in Asia (Martine and McGranahan, 2010). The deterioration of air quality poses a challenge to all of the cities in Brazil. Here, we analysed three major metropolitan areas (capital cities) in Brazil (São Paulo, Rio de Janeiro and Belo Horizonte), each of which has different characteristics in terms of topography and meteorological patterns (Table 1). Using the findings of various authors (Miranda et al., 2011; Andrade et al., 2010, Perez-Martinez et al. 2015, Carvalho et al., 2015) and governmental reports from state environmental protection agencies, we carried out an analysis of PM concentrations and its main sources for the three major capital cities under study. The analysis of data from the São Paulo State Environmental Protection Agency, the Rio de Janeiro State Environmental Institute, the Minas Gerais State Foundation for Environmental Engineering and other sources related to pollutant emissions and concentrations showed that, despite the successful implementation of emission control programs, vehicle emissions constituted the dominant source of PM in these cities due to an substantial increase in the number of on-road vehicles (IBGE, 2015). The same is true for non-exhaust vehicular sources such as the evaporative losses during refueling as well as brake, tyre and road surface wear emissions; none of which has as yet been regulated. This trend is expected since the number of vehicles and vehicle fuel consumption is increasing. These observations indicate that it is necessary not only to establish new emission standards for pollutants emitted by vehicular exhaust, but also to implement policies to reduce the number of on-road vehicles and to establish emission standards for non-exhaust vehicular sources. Figure 1 compares the three regions in terms of PM concentrations and the average emission factors of the vehicle fleet, which is defined as the mass of pollutant emitted per kilometer driven (CETESB, 2015). Notably, the mean concentrations of PM_{10} have not responded to the decrease in emission standards for on-road vehicles. Table 1 shows that the participation of public transportation has increased in recent years (to 37% in São Paulo, to 49% in Rio de Janeiro and to 31% in Belo Horizonte), as a result of policies establishing separate lanes for rapid bus lines (in São Paulo and Rio de Janeiro), an increase in the number of rapid transit lines (in Sao Paulo and Rio de Janeiro) and integration of the train, rapid transit and bus systems (in all these cities). However, the increased participation of public transportation is insufficient, given the high numbers of people commuting every day.

4. Emissions from road vehicles associated health impacts and standards

The Brazilian population was estimated to comprise more than 200 million inhabitants by the end of 2015 (CIA, 2015). This population is distributed throughout 26 states whose capitals have been severely impacted by poor air quality conditions (Miranda et al., 2011). Vehicular emissions are the main source of PM, especially $PM_{2.5}$, in Brazil (Miranda et al., 2011). On average, in 2014, the $PM_{2.5}$ contribution made by the road traffic was equal to 34% nationwide, staying ahead of sources such as industry, domestic fuel burning and natural sources (Karagulian et al., 2015). In Belo Horizonte, only 17% of the ambient $PM_{2.5}$ is attributed to vehicular emissions whereas this proportion in São Paulo and Rio de Janeiro increases to 40% and 50%, respectively (Andrade et al., 2010). In other megacities, the traffic fleet contributes significantly to the PM emissions. For example, around 80% of the PM emissions in Central London in 2012 were related to exhaust and non-exhaust vehicular emissions (e.g., tyre and brake wear). Emissions from construction sites, waste incineration, and construction machinery were other common sources of pollution in London (GLA, 2012). Diesel vehicles, which are normally represented by HDVs in Brazil, present greater PM emissions compared with gasoline-driven automobiles (Onursal and Gautam, 1997; Vela et al., 2015).

In 2004, a study carried out in São Paulo showed that HDVs contributed 6-times more emissions of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ compared with LDVs (Sánchez-Ccoyllo et al., 2009). Nowadays, this ratio is higher due to the greater reduction in the emission factor of PM by the passenger cars in comparison to the diesel vehicles (CETESB, 2015). The contribution of HDVs to air pollution is also seen in cities like Delhi where commercial HDVs accounted for 32 to 43% of the total PM_{10} emissions registered between 1991 and 2011 (Nagpure et al., 2016). Whilst HDVs are the major responsible source for PM emissions in the MASP, LDVs dominates the emissions of carbon monoxide (CO) and hydrocarbons (HC) (Kumar et al., 2016). The Brazilian government has limited the sales of diesel passenger cars since the 1970s, however, diesel commercial LDVs are on sale in the country and ~65% of total exhaust $PM_{2.5}$ emissions come from new LDVs (Dallmann and Façanha, 2016).

Exposures to PM emissions can negatively affect human health. Adverse health effects related to inhalation of PM in the Southeast Brazilian Region have already been documented in the literature (De Oliveira et al., 2011; Wikuats et al., 2014). Fine particles are intrinsically associated with respiratory and cardiovascular diseases and daily mortality (Brook et al., 2010; Laden et al., 2000; Pope and Dockery, 2012). However, coarse particles are also capable of adversely affecting human health (Heal et al., 2012). It is estimated that about 1750, 950 and 360 individuals in the MASP, MARJ and MABH, respectively, are hospitalised annually due to respiratory diseases related to PM_{10} emissions (Marcilio and Gouveia, 2007). In these metropolitan areas, ~5% of the deaths associated with respiratory diseases seen in the elderly (≥65 years old) and children (≤5 years old) were attributed to the ambient PM_{10} concentrations (Marcilio and Gouveia, 2007).

In order to control vehicular emissions, the National Environmental Council (CONAMA) created the Air Pollution Control Program by Motor Vehicles (PROCONVE) in 1986 (IBAMA, 2011), which has considerably reduced pollutant emissions from the LDVs and HDVs (Pérez-Martínez et al., 2015). Tables 2 and 3 show the evolution of PROCONVE emission standards for new LDVs and HDVs in Brazil, respectively. From 1996 to 2009, the measures implemented by the Program have helped to reduce the LDV and HDV emissions levels by 90% and 80%, respectively (Carvalho et al., 2014). The air pollution control is exercised by motor vehicle classifications associated with phases that become increasingly restrictive regarding vehicular emissions (IBAMA, 2011).

Even though PROCONVE has helped to decrease PM emissions in Brazil, the limits set by this Program are still high compared with the newest European standards (e.g., Euro VI). The limits for the HDV are presented in Table 3, showing that the last phase of the program P7, similar to Euro 5, was implemented in 2012. The new LDVs being sold in Brazil must follow PROCONVE L6 phase and should not emit more than 0.025 g (PM) km^{-1} (Table 2). This value is comparable to the Euro IV standards implemented in 2005, but 5-times higher than the current Euro VI value (0.005 g/km) adopted in 2013 (Dallmann and Façanha, 2016). Another important aspect is the necessity of controlling the leakage and evaporation of fuel during refuelling of vehicles. In addition, PROCONVE standards do not include motorcycle emission limits. The Brazilian motorcycle fleet has increased 14-times from 1990 to 2008 and doubled between 2005 and 2009 (Estupiñan et al., 2013). Recognising the importance of emissions from motorcycles, the National Environmental Council established the Brazilian Motorcycle Emissions Control Program (PROMOT) in 2003 (Carvalho et al., 2014). However, this program does not include PM emission standards for motorcycles and focuses only on emission rates of carbon monoxide (CO), total hydrocarbons (THC) and nitrogen oxides (NO_x) (IBAMA, 2011). Further amendments in the program need to be considered since it is known that motorcycles can be a great source of PM emissions, especially those powered by two-stroke engines (MECA, 2014).

4.1 Emissions in the MASP

Table 4 summarises the PM emission trends for the three metropolitan areas. According to the São Paulo State Environmental Protection Agency (CETESB), MASP traffic fleet in 2014 was responsible for the exhaust emission of 1.48 ($\times10^6$) kg yr^{-1} of total PM (including $PM_{2.5}$ and PM_{10}). Out of which, LDVs, HDVs, and motorcycles contributed to 0.25 ($\times10^6$) kg yr^{-1}, 1.18 ($\times10^6$) kg yr^{-1} and 0.07 ($\times10^6$) kg yr^{-1} of PM emissions, respectively (CETESB, 2015). Per capita emissions of PM_{10} in MASP was equal to 200 kg yr^{-1} and particle re-suspension contributed to about 25% of the total PM_{10} emissions in 2014 (CETESB, 2015). In general, PM emissions in the MASP have varied substantially throughout the years, as shown by CETESB reports (CETESB, 1986, 1991, 1996, 2001, 2006, 2011, 2012, 2013, 2014, 2015) and condensed in Table 4. For example, MASP vehicle fleet emitted 1.54 ($\times10^6$) kg yr^{-1} of PM through tailpipes in 2013, accounting for ~30% of the total PM emissions in the area (CETESB, 2014). The traffic fleet also contributed with 1.38 ($\times10^6$) kg yr^{-1} of PM exhaust emissions in 2012 whereas this value was 1.40 ($\times10^6$) kg yr^{-1} and 1.74 ($\times10^6$) kg yr^{-1} in 2011 and 2010, respectively (CETESB, 2011, 2012, 2013).

The recent emission values registered in the MASP are comparable to other megacities worldwide such as London. For example, in 2013, the road transport in London contributed with ~2.20 ($\times10^6$) kg yr^{-1} of PM_{10} and ~1.25 ($\times10^6$) kg yr^{-1} of $PM_{2.5}$ (GLA, 2013). Similar to the MASP, re-suspension of particles accounted for ~23% of the total PM_{10} emitted in London in 2013 (GLA, 2013). However, from 2005 to 2010, CETESB has modified its methodology to calculate PM emissions in the MASP. Consequently, the emission values from 2005 and past years are much higher compared with the present values, which appear to be underestimated. This explains the fact that despite the PM emissions in MASP are similar to those in London but the concentrations values in São Paulo are higher.

4.2 Emissions in the MABH

In the MABH, buses and LDVs of the vehicle fleet emitted together an average 58.1 kg h^{-1} of PM_{10} during the morning rush hours in 2014 (IEMA, 2014). The projections by the Energy and Environmental Institute (IEMA) is that, in 2020, the PM_{10} emissions in the area will increase by up to 63.2 kg h^{-1} during the peak hours (IEMA, 2014). According to IEMA, exhaust emissions from car and buses of the MABH fleet in 2014 comprised only 2% and 17% of the total vehicular PM_{10} emissions, respectively, whereas 81% of the vehicular PM_{10} emissions originated from non-exhaust emissions such as tyre, brake and road surface wear. Predictions point out that this proportion is likely to increase to almost 90% in 2020, which means that an emission control may be ineffective in solving this situation (IEMA, 2014). A similar situation was also identified in Delhi, where non-exhaust sources (e.g., tyre and brake wear) account for 66-86% of the total PM_{10} discharges from 2002 onwards (Nagpure et al., 2016). PM_{10} emission trends in the MABH have changed considerably over the years. In 2003, 739 kg h^{-1} of PM_{10} was emitted in the MABH by stationary (e.g., industrial emissions) and mobile (e.g., vehicular emissions) sources, of which vehicles accounted for 95% of the total emissions (~702 kg h^{-1}) (FEAM, 2010). In contrast, LDVs were responsible for 33.9 kg h^{-1} of PM_{10} emissions in 2008 during the morning peak hours; out of which 2.4 kg h^{-1} of PM_{10} were emitted from the tailpipes of cars while 31.5 kg h^{-1} were attributed to wear emissions. In this same year, buses emitted 15.3 kg h^{-1} of PM_{10} through exhaust sources and 6.6 kg h^{-1} through non-exhaust emissions (IEMA, 2014).

4.3 Emissions in the MARJ

In 2010, MARJ's traffic fleet emitted 28.9 ($\times 10^6$) kg yr^{-1} of PM_{10} through exhaust and non-exhaust sources. Out of this, diesel vehicles were responsible for 9.8 ($\times 10^6$) kg yr^{-1} of PM_{10} exhaust discharges whereas the tailpipes of gasoline-driven vehicles and motorcycles emitted 5.4 ($\times 10^6$) kg yr^{-1} and 0.1 ($\times 10^6$) kg yr^{-1} of PM_{10}, respectively. In addition, tyre wear was responsible for the emission of 13.5 ($\times 10^6$) kg yr^{-1} of PM in 2010 (INEA, 2011b). In 2004, mobile sources (e.g., aeroplanes, ships and automobiles) emitted 7.8 ($\times 10^6$) kg yr^{-1} of PM_{10} in the MARJ, a number that corresponds to 42% of the total PM_{10} emissions in the area (INEA, 2009). The main avenue in Rio de Janeiro, known as Avenida Brasil, ranked first by INEA for PM_{10} emission rates. This avenue road contributed 22.9% of the PM_{10} emissions that originated from the traffic fleet (INEA, 2009). MARJ'S recent emissions values are small compared with Delhi's emissions registered in 2010 but high when paralleled with Sydney's PM emissions recorded in 2008 (EPA, 2008; Sahu et al., 2011). In 2010, Delhi's traffic fleet contributed with 30.29 ($\times 10^6$) kg yr^{-1} of PM_{10} and 30.25 ($\times 10^6$) kg yr^{-1} of $PM_{2.5}$ (Sahu et al., 2011). In contrast, on-road mobile sources in Sydney emitted 2.11 ($\times 10^6$) kg yr^{-1} of PM_{10} and 1.55 ($\times 10^6$) kg yr^{-1} of $PM_{2.5}$ in 2008, including exhaust, evaporative and non-exhaust emissions (EPA, 2008).

4.4 Discussions on PM emissions in studied areas

The emissions of PM in the MASP have been decreasing with time (Table 4). From 1985 to 2005, the total exhaust vehicular PM emissions in the area have decreased by about 50%, a fact that can be associated with the implementation of PROCONVE Standards (Tables 2 and 3). Although PROCONVE did not cover PM emissions until 1993 for HDVs and 2006 for LDVs, regulatory limits on other pollutants have led to engine improvements in new vehicles, which helped reduce PM emissions. Moreover, the decrease of PM emissions in the MASP from 2005 onwards may be explained by the popularity of flex-fuel cars and the introduction of biodiesel to the market. No similar conclusions can be drawn for the MARJ since there is very little data available on PM emissions in this area. However, it is noticeable that non-exhaust emissions have a substantial contribution to PM emissions in the region, accounting for approximately 47% of the total vehicular emissions registered in 2010 (INEA, 2011a). The same significance of non-exhaust sources can be perceived in the MABH, where PM_{10} discharges related to emissions from the tyre, brake, and road wear have increased from 38.1 kg h^{-1} in 2008 to 47 kg h^{-1} in 2014 (IEMA, 2014). Although exhaust emissions have been reducing in the MABH, the total PM_{10} emission is increasing due to the mentioned wear emissions and the contributions from the processes. Until now, the regulations are more effectively applied to vehicular emissions with the PROCONVE Standards. The control of non-exhaust emissions in the country is not effective due to the lack of information regarding industrial emissions. Furthermore, the existing reports for the MARJ do not specify the fraction of $PM_{2.5}$ that comprises the total PM_{10} emission whereas MABH reports focus on PM_{10} emissions only. This knowledge gap clearly indicates the need for further studies on non-exhaust PM emissions in these metropolitan areas.

The use of biofuels can also directly influence particles emissions (Kumar et al., 2010). Some studies have shown that the vehicular emission factors of $PM_{2.5}$ have reduced significantly in the MASP over the past few years (Pérez-Martínez et al., 2014). The $PM_{2.5}$ emission factors of LDVs have decreased from 92 mg km^{-1} in 2004 to 20 mg km^{-1} in 2011 (Pérez-Martínez et al., 2014). This reduction is intrinsically related to the PROCONVE program and the growth in ethanol consumption since the introduction of flex-fuel cars in the market. $PM_{2.5}$ emission factors from the HDVs were equal to 588 mg km^{-1} in 2004 and this number decreased to 277 mg km^{-1} in 2011 (Pérez-Martínez et al., 2014). No major engine improvements were made to HDVs at that time and, consequently, the reduction in emissions is usually associated with the addition of biodiesel to the diesel blend and with the introduction of low-sulphur diesel fuels (Sánchez-Ccoyllo et al., 2009).

Some studies also indicated that a full application of ethanol as a transportation fuel would remove ~90% of the total particle mass emissions (Aparicio et al., 2014). According to the Sustainability Report (UNICA, 2010), prepared by the Brazilian Sugarcane Industry Association (UNICA), the substitution of gasoline and diesel with ethanol as a vehicle fuel would save about 875 lives and avoid more than 12,000 hospital admissions in the MASP alone. Nevertheless, increasing the biodiesel proportion in the diesel blend has proved to assist in reducing the PM emissions by –10% for B10 blends to up to –50% for pure biodiesel (Kousoulidou et al., 2008; Randazzo and Sodré, 2010). However, the addition of ethanol in the fuel blends can affect air quality negatively through increased emissions of aldehydes, acetic acids and unburned alcohols (Aparicio et al., 2014; De Melo et al., 2012) whereas the use of biodiesel has also promoted a slight increase in NO_x emissions, according to several studies reviewed by Lapuerta et al. (2008).

5. PM Concentrations

The Brazilian government, along with numerous researchers and academic groups, have been assessing the national air quality conditions in diverse areas of the country. Monitoring stations are distributed throughout the most important Brazilian cities and metropolitan areas, gathering information on emissions and concentrations of various pollutants. The "First Diagnosis of the Brazilian Air Quality Monitoring Network", published by the Energy and Environmental Institute (IEMA) in 2014, shows that PM levels are measured and analysed by the majority of monitoring stations located in the MASP, MARJ, and MABH (IEMA, 2014).

PM concentrations and their main characteristics, such as composition and physicochemical properties of particles, can vary considerably according to meteorological and geographical conditions (Miranda and Andrade, 2005). Winter periods are characterised by low dispersion of pollutants due to the thermal inversion events and less rain. These Metropolitan Regions in the south-east of the country are characterised by rainy summer and dry winters. This aspect has led to the implementation of the "Winter Operation" in the MASP since 1976; this operation involves adopting preventive and corrective actions that aim to control the pollution levels during the winter and develop a quality database for fixed and mobile sources emissions, especially the levels of sulphur content in the atmosphere. These solutions are an effort to prevent PM concentrations to keep getting worse during winter time over the years (CETESB, 2015).

Road vehicles and industries are the dominant sources contributing together ~40% and ~60%, respectively, of primary PM_{10} and $PM_{2.5}$ in the MASP (CETESB, 2015). In the past, concentrations of PM_{10} in the MASP were well above $PM_{2.5}$ levels; however, the gap between those two has been closing, meaning that combustion emission is dominating mechanical emissions with time. A large number of tall buildings and concrete surfaces helps the thermal inversion phenomenon, causing the formation of air pollution episodes with high concentrations of PM (Kukkonen et al., 2005). The biomass burning of sugarcane season (May to November) contributes to the urban pollution, such as the MASP (Kumar et al., 2016). Those two characteristics, together with the sea-salt particles transported from the seashore, showed notable contributions to the PM concentrations registered in the MASP region (Miranda et al., 2011; Souza et al., 2014).

In the MARJ, some regions that incorporate important Brazilian enterprises (e.g. petrochemical complex and mining and metallurgical facilities) showed higher PM concentrations than other regions; emissions from industries not only intensify total PM emissions but also increase the portion of sulphur content within $PM_{2.5}$ discharges originated from the oil refineries (Miranda et al., 2011). Yet, road traffic emissions and oil combustion are major sources of the total PM mass emission (~52 to 75%) registered in the MARJ. Industries, soil dust, and sea spray are other great sources of atmospheric pollution in the area (Godoy et al., 2009).

Numerous industries related to metallurgical, mining activities and automobile manufacturers are also located in the MABH; this area represents 66% of the total mining activities performed in Minas Gerais State (FEAM, 2012). Moreover, a chain of mountains located on the East side helps to flank the region, preventing the pollution spread along with the climate profile. As in the other metropolitan areas, vehicular emissions are together with industrial the major sources of air pollution in the MABH (FEAM, 2012; Miranda et al., 2011). Until now, the MABH operates nine governmental monitoring stations that constantly characterise PM_{10} concentrations, whilst only one station monitors $PM_{2.5}$ levels. In fact, the majority of the available data regarding $PM_{2.5}$ concentrations in Brazil are provided by academic studies (Miranda et al., 2011). This aspect is certainly a weakness in the Brazilian air quality management, especially with the established hazards related to fine particles (Fernández et al., 2000; Heal et al., 2012; Martins et al., 2009). This is also due to the absence of air quality standards for $PM_{2.5}$ in Brazil, being São Paulo State the only exception.

5.1 $PM_{2.5}$ concentrations

Figure 2 illustrates the ambient concentrations of $PM_{2.5}$ in the MASP, MARJ and MABH between June 2007 and August 2008 (Miranda et al., 2011). This study covers all three metropolitan areas and we chose to discuss it because all the measurements were conducted concurrently utilising the same monitoring methods, which allows meaningful comparison. Despite not being a recent publication, Miranda et al. (2011) is one of the few published studies that present a good analysis of $PM_{2.5}$ concentrations in the studied metropolitan areas. As discussed in Section 5, national reports and inventories regarding $PM_{2.5}$ levels in Brazilian cities could not be found in the literature.

It was registered in the MASP the mean $PM_{2.5}$ concentration of 23 μg m^{-3} in the summer, 35.5 μg m^{-3} in the winter, and a mean average for the whole period of 28.1 μg m^{-3}. Figure 2a illustrates the variation of $PM_{2.5}$ levels in three-month intervals throughout the period analysed (June 2007 to August 2008), with a 35.7 μg m^{-3} peak between June and August 2008 and the lowest value of ~20 μg m^{-3} between December 2007 and February 2008 (Miranda et al., 2011). A higher concentration value can be seen during the winter season (June to August), demonstrating the impact of meteorological conditions, as was also observed in cities like Delhi (Kumar et al., 2015).

As seen in Figure 2b, the MARJ showed a slight increase in mean values of $PM_{2.5}$ between June 2007 and August 2008 during winter (23 μg m^{-3}) compared with summer (15.8 μg m^{-3}) periods. The winter in the area is characterised by lower temperatures (21.1 ºC in winter compared to 25.4 ºC in summer), lower relative humidity (78% in winter and 83% in summer) and less precipitation (139.2 mm accumulated during winter and 560.8 mm accumulated in summer), resulting in higher concentrations of $PM_{2.5}$ (Miranda et al., 2011). The annual mean for the whole period was 17.2 μg m^{-3} and its variation throughout the year illustrated in Figure 2b shows concentrations of $PM_{2.5}$ ranging from a 19.9 μg m^{-3} peak (June 2008 to August 2008) to a minimum of 12.4 μg m^{-3} (December 2007 to February 2008) (Miranda et al., 2011).

In the MABH, the annual mean $PM_{2.5}$ concentration was 14.7 μg m^{-3} (Figure 2c). During the winter season, the mean concentration increased up to 18.5 μg m^{-3} whereas it dropped to 14.5 μg m^{-3} during the summer. The variation measured through the period showed a peak of 17.7 μg m^{-3} during June to August 2007 and the lowest concentration of 9.0 μg m^{-3} between December 2007 and February 2008. In general, $PM_{2.5}$ concentrations in the MASP were almost twice as large as those registered in the MARJ and MABH for the same period (Figure 2).

The São Paulo State Environmental Protection Agency (CETESB) constantly monitors $PM_{2.5}$ levels in the MASP and presents results in annual reports. In general, the overall annual mean concentrations of $PM_{2.5}$ vary according to the regions where the monitoring stations controlled by CETESB are located inside the metropolitan area. In 2015, the Congonhas Airport region presented $PM_{2.5}$ levels around 20 μg m^{-3} whereas the Marginal Tietê, a motorway with intense traffic, showed values of 22 μg m^{-3}, the highest among all stations in the MASP. The daily mean concentration of $PM_{2.5}$ for all monitoring stations in the MASP was equal to 20 μg m^{-3} in the same year. The maximum daily mean concentration of $PM_{2.5}$ was recorded about 60 μg m^{-3} considering all the stations (CETESB, 2015). In contrast, Delhi usually presents the worst conditions during traditional festivals, when annual mean $PM_{2.5}$ levels reached to 278 μg m^{-3} in 2009 (Perrino et al., 2011). Beijing, on the other hand, normally register its worst conditions during the haze days; for example, in 2012, annual mean concentrations of $PM_{2.5}$ reached to 143 μg m^{-3} during such period (Zhang et al., 2016).

Compared with the previous data, this shows that the concentrations inside metropolitan areas vary between regions, increasing in areas with high traffic, industries and during periods of intense activity such as festivals. The time used for the analysis also change the results significantly, as time passes, new emission may be found in these areas and higher vehicle fleet and industries cause a change in the concentrations. However, numerous interventions such as improvements in engines of road vehicles, new types of biofuels, stricter emission standards and a better monitoring system have helped to reduce the annual $PM_{2.5}$ concentrations in the MASP throughout the years. For example, the annual mean on 2007/08 in the whole MASP had a concentration of 28.1 μg m^{-3} whereas the worst region inside MASP in 2015 showed no more than 22 μg m^{-3} during the year, resulting in an even lower mean for the whole metropolitan area.

Measurements made at the University of São Paulo (USP) campus have shown a decrease in the overall annual mean $PM_{2.5}$ concentrations from 19 μg m^{-3} in 2012 to 12 μg m^{-3} in 2015, as summarised in Table 5 (CETESB, 2015). In 2013, the state of São Paulo established new PM guidelines, being the pioneer Brazilian state to set limits for $PM_{2.5}$ concentrations. This fact may explain the reductions identified in 2013 compared with the previous year. The station located at USP campus started measuring $PM_{2.5}$ levels in 2012, explaining why earlier data could not be found.

Another study carried out in the MARJ from 2003 to 2005 showed that the regions with the highest $PM_{2.5}$ concentration in the area were sampling sites located close to industrial emissions, as well as areas with heavy vehicular traffic (Godoy et al., 2009). Up to date, the State governments and the local institutes have not published monitoring reports regarding

$PM_{2.5}$ concentrations in the MARJ and MABH. Hence, all the available data are solely based on previously published studies (Mariani and de Mello, 2007; Miranda et al., 2011).

5.2 PM_{10} concentrations

Several reports regarding PM_{10} concentrations in the MASP, MARJ and MABH were found in the existing literature. The annual mean concentrations of PM_{10} throughout the years in the MASP, MARJ and MABH are summarised in Table 6. In 2015, annual concentrations in the MASP showed slightly increased values in areas of intense road traffic, such as Osasco and Parelheiros, with a concentration of 40 µg m^{-3} for both regions. The 24h-limit analysis in the short-term measurement showed a maximum concentration of 111 µg m^{-3} and 121 µg m^{-3} for the same regions (Parelheiros and Osasco), respectively (CETESB, 2015). According to CETESB monitoring results, annual mean PM_{10} concentrations have stayed almost constant throughout recent years in the MASP, varying from 33 µg m^{-3} in 2009 to 31 µg m^{-3} in 2015 (CETESB, 2009, 2015). Slightly increased values were registered in the area in 2010 and 2011 when annual mean concentrations reached to 39 µg m^{-3} and 38 µg m^{-3}, respectively (CETESB, 2010, 2011). These values are well below those encountered decades ago. In 1990, the annual mean PM_{10} concentration in the MASP was approximately 60 µg m^{-3} (CETESB, 1991). The difference between previous and recent PM_{10} levels results from the implementation of PROCONVE standards, which reduced vehicular emissions, and the creation of CONAMA guidelines in 1990, which set stricter limits to PM_{10} levels in Brazil (Carvalho et al., 2014).

Similar to the behaviour of $PM_{2.5}$, PM_{10} concentrations also vary according to meteorological conditions. Carvalho et al. (2014) have documented the variation in monthly average values of PM_{10} concentrations in the MASP from 1996 to 2009 As expected, the highest concentration values were reached in the winter, especially in August (~65 µg m^{-3}) whereas December and January (summer time) presented the lowest PM_{10} levels (~35 µg m^{-3}) from 1996 to 2009.

In the MARJ, PM_{10} levels have reduced considerably in recent years. In the Botafogo district station, monitored by State Environmental Institute (INEA), PM_{10} concentrations have decreased from 40 µg m^{-3} in 2010 to 34 µg m^{-3} in 2014. Some industrial regions inside the MARJ (São Cristovão) presented worst scenarios in 2014, registering annual concentrations 1.5-times greater (~69 µg m^{-3}) than the average in the area (INEA, 2015). In an extended analysis over the metropolitan area, between October 2008 and September 2009, annual mean concentrations registered 46.5 µg m^{-3} for the city of Rio de Janeiro and 71.1 µg m^{-3} for Duque de Caxias (Gioda et al., 2011).

In 2014, the maximum short-term measurement registered in the MARJ was ~159 µg m^{-3} and ~63 µg m^{-3} in São Cristovão and Botafogo, respectively. However, the station near the petrochemical complex in Duque de Caxias presented the highest concentration within the metropolitan area, with a maximum 24h-limit value equal to 249 µg m^{-3} (INEA, 2015).

Samples collected at different sites in Rio de Janeiro indicated the presence of elements associated with the automotive traffic, tire rubber and brake abrasion on PM_{10} concentrations (Silva et al., 2008).

In the MABH, the State Environmental Foundation (FEAM) is responsible for monitoring the air quality condition at different stations located in the area. The concentration throughout the years showed an increase in the annual means, from 21.5 µg m^{-3} in 2005 to 31.3 µg m^{-3} in 2010, with a slight reduction in 2012 (30.4 µg m^{-3}) (FEAM, 2012). This increase can be linked to the evolution of the traffic fleet in the region and the development of new industries and roads because of the national economic growth. New infrastructure projects were also carried out during this period due to the 2014 World Cup, an aspect that could show some responsibility for the PM_{10} levels registered in the area.

The latest FEAM report (FEAM, 2012) showed an annual mean PM_{10} concentration of 50.6 µg m^{-3} in the most polluted region of the MABH (Betim). The fact that Betim is the home of a Petrobras oil refinery and an automobile manufacturer explains the pollution levels measured in this area. In 2012, Belo Horizonte, the largest city in the MABH, had an annual mean concentration of 30.4 µg m^{-3}. Daily short-term concentrations reached the highest values in September 2012 for both regions; ~200 µg m^{-3} in Betim and ~90 µg m^{-3} in Belo Horizonte's centre.

6. Guidelines and exceedances

Air quality management varies from country to country based on the approaches adopted to control health risks. These approaches consider the feasibility of a given guideline, economic considerations, and social factors, depending on the capability of developing air quality solutions. Table 7 summarises all these guidelines according to particle sizes and resolutions. The Brazilian daily mean PM_{10} concentration limits are 3-times greater than WHO (i.e., 150 µg m^{-3} by CONAMA compared with 50 µg m^{-3} by WHO) for a short-term (24h) exposure range and 2.5-times greater (50 µg m^{-3} CONAMA; 20 µg m^{-3} WHO) for the annual mean exposure range. In addition, $PM_{2.5}$ concentrations limits are not covered by the existing Brazilian guidelines.

Some Brazilian states (e.g. São Paulo) are trying to tighten these guidelines towards WHO limits with a three-phased plan; however, the adopted values in the first phase are still far from WHO guidelines. Concentration limits for $PM_{2.5}$ have been set in São Paulo and they are 2-times greater in the annual mean and almost 3-times bigger in the 24h-limit value compared with WHO (CETESB, 2013). It is worth considering that even WHO guidelines are not fully safe and its concentration limits cannot guarantee the prevention of adverse health effects (Martins et al., 2009).

In the MASP, all the $PM_{2.5}$ concentration values analysed surpassed WHO guidelines and the local limits (CETESB, 2013). The registered annual mean of 28.1 μg m^{-3} between June 2007 and August 2008 exceeded the WHO limits in 18.1 μg m^{-3} (~180% higher) and the São Paulo guideline in 8.1 μg m^{-3} (~40% higher). The worst region reported in the MASP was Congonhas Airport in 2015, which surpassed WHO limits by 38 μg m^{-3} with a short-term measurement of 63 μg m^{-3}. The PM_{10} concentrations in 2015 followed the same behaviour when both annual and 24h-limit values were above WHO guidelines. Yet, the values registered in all monitoring stations did not exceed the local recommendations. The difference between the annual PM_{10} concentration and the WHO limits reached 11 μg m^{-3} (i.e., 55% higher than WHO threshold) in the MASP study; this difference was about twice over the Parelheiros area that also showed a 24h-limit concentration by about 142% (121 μg m^{-3}) above the WHO guidelines.

The MARJ showed a higher level of pollutants than the MASP analysis, except for the $PM_{2.5}$ concentrations. Yet, from 2007 to 2008, $PM_{2.5}$ concentration values were greater than WHO limits by 5.8 μg m^{-3} (58% higher) in the whole area and 13 μg m^{-3} (130% higher) during the winter season. Regarding PM_{10} values, in 2014, all stations monitored in the MARJ were in accordance with the national guideline boundary; however, they all reached annual means that surpassed the limit determined by WHO. The annual mean concentration of PM_{10} in the MARJ was 132% higher than the recommended in 2014. In the worst area found in the MARJ (Duque de Caxias), the annual mean of 71.1 μg m^{-3} is ~3-times (355%) above the global guideline's threshold and 21.1 μg m^{-3} beyond the Brazilian limit. When analysing 24h-limits, MARJ's region known as São Cristovão presented average PM_{10} concentrations 3-times greater than the WHO guidelines and one occurrence slightly over the CONAMA limit. Once again, Duque de Caxias area was well above WHO (199 μg m^{-3} exceeded) and CONAMA (99 μg m^{-3} exceedance) limits. In this region, the concentration limit was surpassed more than 8 times in a year; normally, only one exception is accepted by the guidelines. Besides all measurements being beyond the global limit, only one of them was above the national limit.

The MABH was the least polluted area of the three regions in this study. $PM_{2.5}$ concentrations in the region were the closest to the WHO guidelines, exceeding the annual mean limit in 4.5 μg m^{-3} (2007–2008). The winter concentration value (8.5 μg m^{-3}) was 85% higher than the global guideline for $PM_{2.5}$, the closest so far. In 2012, the annual mean of PM_{10} recorded a 10.4 μg m^{-3} exceedance (52% beyond) compared to WHO. In this same year, the difference registered for the 24h-limit value was even higher – 80% above WHO guideline (40 μg m^{-3}). Betim exceeded the annual mean in 30.4 μg m^{-3} (152% beyond WHO limits), whilst the 24h-limit surpassed it in 3-times due to the industrial activity and petrochemical companies located in the region. All values were in accordance with the CONAMA guideline, except the 24h limit, which exceeded 50 μg m^{-3} in 2012.

Tables 8 and 9 presents the concentration values and their compliance with WHO guideline. This result shows how far Brazil is from developing an acceptable management system for air quality and emission's control in comparison with the international guidelines (e.g., WHO). On the other hand, Table 10 brings together relevant mobility and geographical features of different megacities worldwide. It can be seen from the Table 10 that the ridership of buses and metro are related to the public transport usage. Further, the size of public bus fleet and the rail length is related to the demand for public transport, while the car ownership can be associated with the economic development of the cities. However, cities with similar human development index and population can differ in the proportion of private cars, since other factors, such as public transport supply, promotional fares and charging schemes can influence the car ownership (Pan, 2013).

6.1 Comparison with cities worldwide

As discussed in Section 5.1, the studied metropolitan areas (MASP, MARJ, and MABH) have shown reduced $PM_{2.5}$ concentration values compared with other cities in the world, such as Delhi and Beijing. In Delhi, the annual mean concentration of $PM_{2.5}$ was between 110 and 170 μg m^{-3} in 2011 (Apte et al., 2011). In Beijing, the average annual concentration of $PM_{2.5}$ was ~56 μg m^{-3} in 2012 (WHO, 2014). London, however, presented better results compared with the three metropolitan areas, with an annual $PM_{2.5}$ concentration of 16 μg m^{-3} during 2011 (WHO, 2014).

In Delhi, the annual PM_{10} concentration reached 286 μg m^{-3} in 2010, a value that is extremely high compared with the studied metropolitan areas (WHO, 2014). In this megacity, great doses of particle pollution are inhaled from walking, cycling or using public transports, raising a relevant concern regarding public health (Goel et al., 2015). London's annual PM_{10} concentration reached 22 μg m^{-3} in 2011 (WHO, 2014); this reduced value can be attributed to congestion charging schemes and emissions and traffic interventions (Atkinson et al., 2009). Beijing, a highly industrial city, kept its PM_{10} concentrations around 121 μg m^{-3} in recent years, with the worst measurements encountered in suburban areas (Kong et al., 2016).

Figures 3 and 4 compare $PM_{2.5}$ and PM_{10} levels in the studied metropolitan areas with other cities in the world according to WHO Database (WHO, 2014). In 2011, cities such as London and Stockholm showed concentration values significantly below those measured in Brazil. London is an important megacity that can be compared with São Paulo in size; however, in 2011, London presented an annual PM_{10} concentration value of 22 μg m^{-3}, whilst the MASP registered 35 μg m^{-3} in 2012 (WHO, 2014). In the MARJ (2010) and MABH (2011), the PM_{10} levels according to WHO Database (WHO, 2014) were around 67 μg m^{-3} and 52 μg m^{-3}, respectively. When comparing the annual $PM_{2.5}$ levels, the difference is smaller (16 μg m^{-3} in London in 2011; 19 μg m^{-3} in the MASP in 2012; 36 μg m^{-3} in the MARJ in 2010; 28 μg m^{-3} in the MABH in 2011) (WHO, 2014). Yet, London surpasses WHO limits for $PM_{2.5}$ and PM_{10} (2 μg m^{-3} in exceedance for PM_{10} and 6 μg m^{-3} in exceedance for $PM_{2.5}$). Even though

Brazil does not have the recommended air quality conditions, the atmosphere in the studied metropolitan areas is cleaner than in other megacities, such as Delhi and Beijing. In 2010, the annual PM_{10} concentration in Delhi was 286 µg m^{-3}; in Beijing, PM_{10} levels exceeded MASP values in ~4.5-times (WHO, 2014). Both Delhi and Beijing present concentrations values of $PM_{2.5}$ and PM_{10} above all the guidelines presented in this review. Stockholm, on the other hand, is the only city that complies with WHO limits.

According to WHO Database (WHO, 2014), the annual mean of $PM_{2.5}$ registered in Rio de Janeiro and Belo Horizonte were quite different in comparison with the values gathered in the academic study (Miranda et al., 2011) (Section 5.1). Such disparity can be explained by different approaches regarding the measurements, distinct periods of time, season variations and by the fact that Brazil does not have a policy of control and measurement of these concentrations yet (with the exception of the São Paulo State that recently changed its air quality regulation).

The majority of the cities worldwide surpass WHO guidelines for PM_{10} and $PM_{2.5}$ concentrations (Figures 3 and 4). However, there is a substantial difference on how much each city goes over those guidelines; cities such as Delhi, Beijing, and Mumbai present the worst air quality scenario whereas the other megacities such as London, Paris and Buenos Aires, show better management over their air quality with smaller PM concentration values. Sydney and Salvador (Brazil) had the best results on the list; annual mean PM_{10} and $PM_{2.5}$ concentrations in Sydney did not surpass WHO limits whereas Salvador only exceeded the $PM_{2.5}$ limits. In general, the cities worldwide presented better results on PM_{10} concentrations compared with $PM_{2.5}$ levels. It is worth noting that WHO guideline does not guarantee safeguard from hazard to human health, meaning that values below WHO limits can still cause health problems, as some studies have previously pointed out (Baldauf et al., 2009; Kumar et al., 2015; Martins et al., 2009).

7. Summary, conclusions and future outlook

We comprehensively reviewed the vehicle fleet and the type of fuel used in three Brazilian metropolitan areas (MASP, MARJ, and MABH). Through a meta-analysis of data from published results, the aims were to assess the aspects that influence PM emissions and their ambient concentrations. Recent emissions trends from the MASP and MABH were discussed and their comparisons with national standards and WHO guidelines were performed in order to evaluate exceedances of PM levels. However, parallel information for the MARJ was not available, suggesting a clear need for further studies in this region. Further, the review discussed the PROCONVE emission standards and its positive impact on the vehicular emissions.

The following conclusions are drawn:

- LDVs account for ~85% of the total vehicle fleet in the MASP, MARJ, and MABH, where flex-fuel cars and gasohol-driven vehicles are the predominant types of LDVs. In general, the use of biofuels such as ethanol and biodiesel is increasing with time due to governmental incentives (e.g., PROALCOOL and Brazilian Law No. 13.033) and it is expected to reach to 30% of the total fuel consumption in 2030. The synthesis of the existing studies indicates that motor vehicles, in particular, diesel-driven automobiles, contribute notably to PM emissions in Brazil, accounting from 34-50% of total $PM_{2.5}$ national emissions (Karagulian et al., 2015; Miranda et al., 2011).
- The implementation of vehicular emission control (e.g., PROCONVE standards) and an increased use of biofuels were very effective in reducing the concentration of air pollutants (e.g., CO, NOX, PM) in the urban areas, but similar regulations are needed to control non-exhaust vehicular emissions such as tyre, brake and road surface wear and the evaporative losses of fuel at filling stations. The available studies for the MABH suggest that the non-exhaust sources could compose 90% of the ambient PM_{10} emissions in 2020. Hence, further research is necessary to assess, control and manage non-exhaust emissions.
- The $PM_{2.5}$ concentrations in the MASP were the highest among the three analysed areas due to the largest number of motor vehicles in the region. The MARJ has presented high PM_{10} concentrations; an outcome explained by the local topography of the region and the presence of the Petrobras complex and other plants in the industrial area of the city. The MABH has the smallest number of inhabitants and vehicles compared with the MASP and MARJ; hence, this region presented the lowest $PM_{2.5}$ mean concentrations of all areas. Comparisons between PM_{10} concentrations of the three metropolitan areas could not be performed due to the lack of recent data.
- The national guidelines (i.e., CONAMA; Resolution No. 03/1990) established limits for PM_{10} concentrations that are about 3-times higher than the WHO guidelines. Yet, the PM concentrations in the MASP, MARJ and MABH found to exceed the WHO guidelines by up to 300%. While there is a need to control emissions at the source and halt the increase in the number of on-road vehicles through a shift from private to public transportation system, both the PM emissions and ambient concentration standards also need to be improved to meet the international guidelines. The initiatives such as restricting vehicular circulation in congested areas, improving and increasing the public transport fleet and incentivising the use of bicycles as a transport mode could help in curtailing the pollutant emissions. In addition, the use of biofuels should be studied more carefully in order to evaluate the types of their environmental impacts (e.g., an increase in aldehydes and secondary pollutants (ozone) and nanoparticle emissions) for establishing the appropriate trade-off during their life cycle.

Much of the current focus has been on controlling and monitoring PM_{10} levels in the studied regions. Therefore, there is a lack of studies on $PM_{2.5}$ emission rates and concentrations, clearly emphasising a need for future studies. The MASP uses most up-to-date air quality and emission standards in Brazil, which should be adopted for the other states in Brazil. The development of novel modelling tools to assess the future air quality scenarios and adopting preventive measures accordingly, could prove helpful to manage air pollution in these regions.

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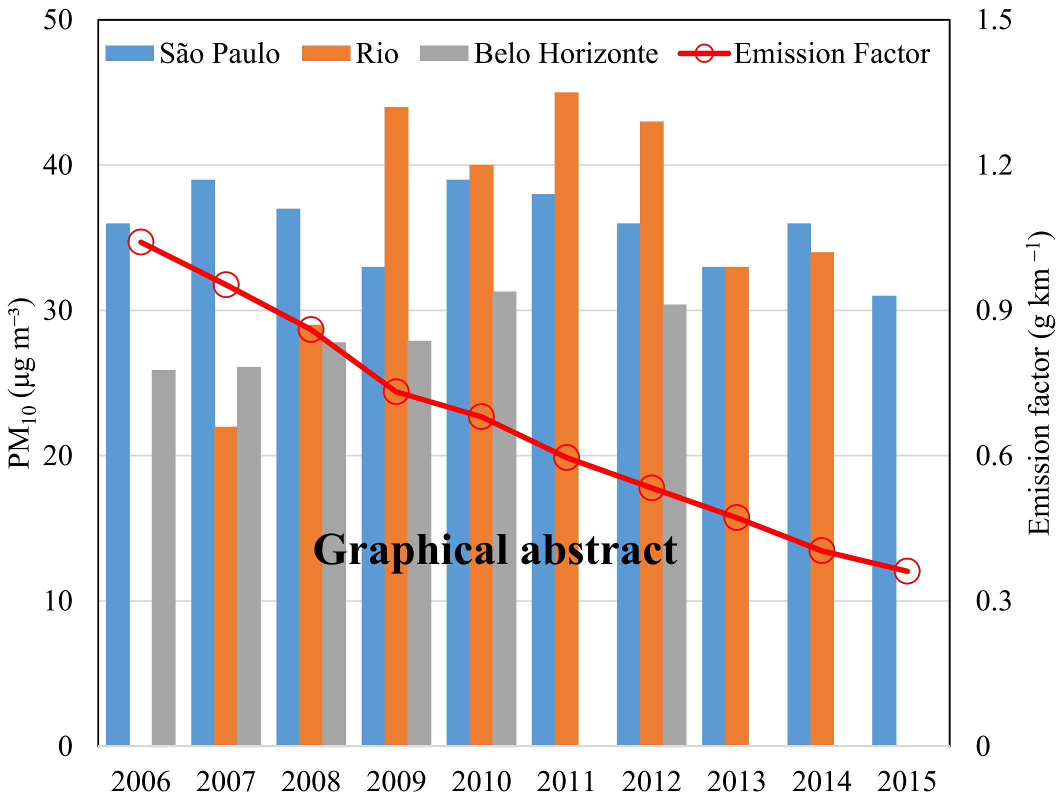

Figure 1. Mean concentration of PM_{10} obtained at the air quality stations in São Paulo, Rio de Janeiro and Belo Horizonte and the average emission factor for light-duty fleet, obtained from CETESB data (2015).

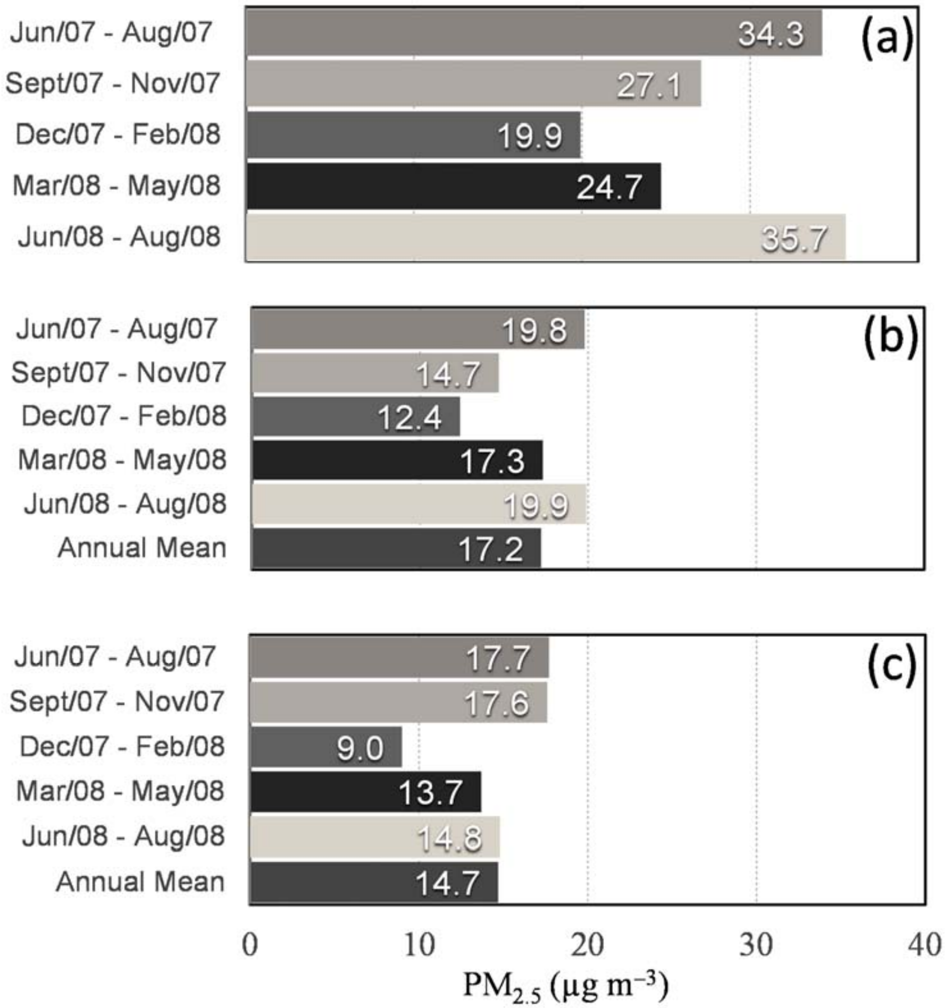

Figure 2. Three-month averages for $PM_{2.5}$ in (a) MASP, (b) MARJ, and (c) MABH (Miranda et al., 2011).

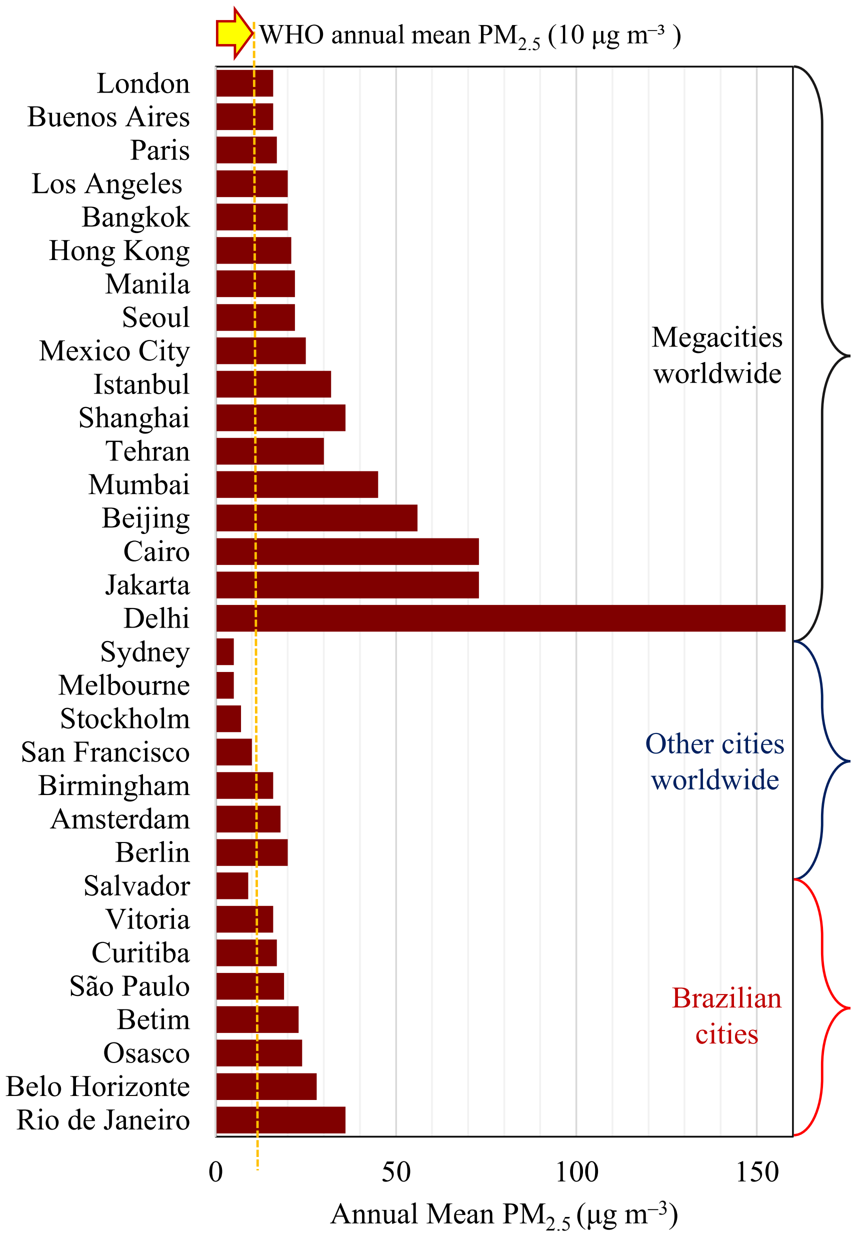

Figure 3. $PM_{2.5}$ concentrations from different cities around the world in comparison with Brazilian metropolitan areas(WHO, 2014). Note: these values were measured from 2010 to 2012 for all cities.

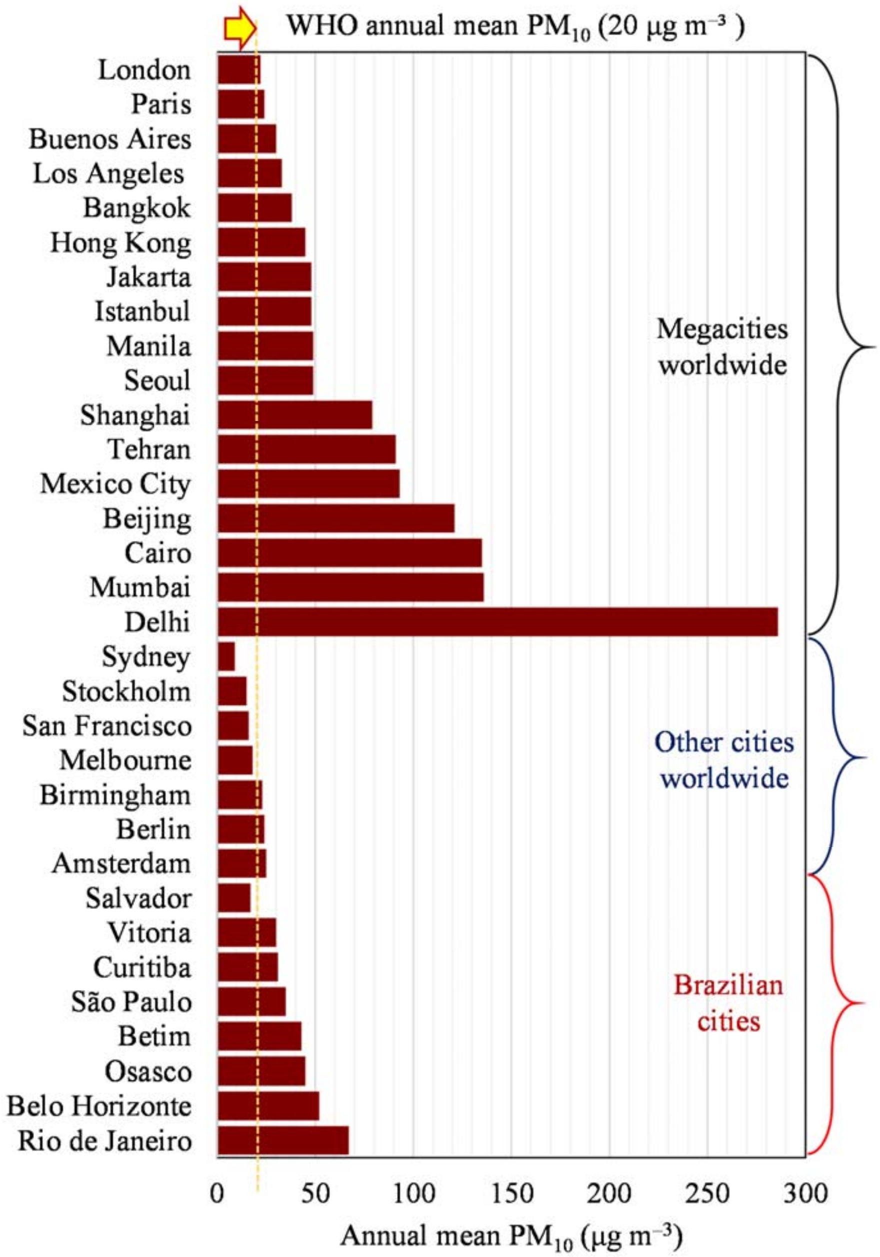

Figure 4. PM_{10} concentrations from different cities around the world in comparison with Brazilian metropolitan areas (WHO, 2014). Note: these values were measured from 2010 to 2012 for all cities.

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Table 1. Characteristics of the three cities along with the number of vehicles according to their fuel type.

Aspects	São Paulo	Rio de Janeiro	Belo Horizonte
Population (annual rate of change, %)[1]	21 Million (1.0%)	13 Million (0.85%)	5.7 Million (1.2%)
Year of foundation	1554	1565	1897
HDI[2]	0.794	0.771	0.774
Geography	50 km from the coast Moderate-high plains	Near the coast Low plains	Far from the coast High plains
Climate[2]	Sub-tropical, 19 C, 1500 mm, moderate seasonality	Tropical, 22°C, 1800 mm, low seasonality	Tropical, 20.5°C, 1430 mm, moderate seasonality
Local sources	Transportation, Industries	Transportation petrochemical industries, refinery	Transportation minery, refinery
Number of Gasohol Vehicles	3,877,098	1,040,321	633,242
Number of ethanol vehicles	495,946	139,801	54,768
Number of Flex fuel vehicle	2,615,055	854,556	885,515
Number of Diesel Vehicles	364,797	124,110	100,092
Number of Electric cars	848	42	29
External sources	Outdoor biomass burning, long range transport	Biomass burning	Biomass burning
total of travel per day (x1000)[4,5,6,5]	43715	22594	13060
% of travels by Public transportation per day [4,5,6]	36.9	48.7	31
% of travels by passenger cars [4,5,6]	31.1	19.5	31
% of travels by foot [4,5,6]	31.4	29.4	37
% of travels by bycicle [3,4,5]	0.6	2.4	1
Major Interventions	Controlling emissions from, vehicles since 1986 with PROCONVE		

[1]IBGE: Growth rate calculated from 2000 to 2010; [2]UNDP (2014); [3]DENATRAN (2015); [4]METRO (2013); [5]SETRANS (2015); [6]BHTrans (2014)

Table 2. PM emission standards by PROCONVE for LDVs.

LDVs PROCONVE PM Emission Standards (IBAMA, 2011)				
		LDVs (passenger cars)	LDVs (commercial <1700 kg)	LDVs (commercial >1700 kg)
Phase	Year	PM (g/km)	PM (g/km)	PM (g/km)
L1[a]	1988 - 1991	-	-	-
L2[a]	1992 - 1996	-	-	-
L3[a]	1997 - 2006	-	-	-
L4	2007 - 2009	0.05	0.08	0.10
L5	2010 - 2012	0.05	0.05	0.06
L6	2013 - Now	0.025	0.03	0.04

[a]PM emission standards were not covered in phases L_1, L_2 and L_3; however, these phases have limited other pollutants' emissions (e.g. CO and hydrocarbons).

Table 3. PM emission standards by PROCONVE for HDVs.

PROCONVE PM Emission Standards for HDVs (IBAMA, 2011)		
Phase	**Year**	**PM (g/kWh)**
P1[a]	< 1990	-
P2[a]	1991 - 1993	-
P3	1994 – 1997	0.40
P4	1998 – 2000	0.15
P5	2004	0.10 or 0.13[b]
P6	2009	0.02
P7	2012	0.02

[a] PM emission standards were not covered in phases P1 and P2; however, these phases have limited other pollutants' emissions (e.g. CO and hydrocarbons).

[b] Applied for unit piston displacement engines of less than $0.75dm^3$ and rotation above 3000 min^{-1}**.**

Table 4. The emission trends of PM for the MASP, MARJ and MABH.

PM Type	Metropolitan Area	Emissions ($\times 10^6$ kg yr^{-1})	Year	Sources	References
PM[a]	MASP	56.60	1985	Traffic fleet[b]	CETESB (1986)
		20.87	1990	Traffic fleet[b]	CETESB (1991)
		40.70	1995	Traffic fleet[b]	CETESB (1996)
		32.50	2000	Traffic fleet[b]	CETESB (2001)
		28.20	2005	Traffic fleet[b]	CETESB (2006)
		1.74	2010	Traffic fleet[c]	CETESB (2011)
		1.40	2011	Traffic fleet[c]	CETESB (2012)
		1.38	2012	Traffic fleet[c]	CETESB (2013)
		1.54	2013	Traffic fleet[c]	CETESB (2014)
		1.48	2014	Traffic fleet[c]	CETESB (2015)
	MARJ	28.90	2010	Traffic fleet[b]	INEA (2011b)
PM_{10}	MARJ	7.80	2004	Mobile sources[c,d]	INEA (2009)
	MABH	702.5[e]	2003	Traffic fleet[b]	FEAM (2010)
		55.80[e]	2008	Traffic fleet[b]	IEMA (2014)
		58.10[e]	2014	Traffic fleet[b]	IEMA (2014)
		63.20[e,f]	2020	Traffic fleet[b]	IEMA (2014)

[a] PM relates to the total particulate matter, being $PM_{2.5}$ a fraction of this total.

[b] Includes exhaust and non-exhaust emissions.

[c] Includes exhaust emissions only.

[d] Includes different means of transportation, such as aeroplanes and ships, but especially automobiles.

[e] These numbers relate to kg h^{-1} emission rates.

[f] Future predictions estimated by IEMA (2014).

Table 5. Annual mean concentrations of $PM_{2.5}$ recorded at the University of São Paulo Campus (CETESB, 2012, 2013, 2014, 2015).

$PM_{2.5}$ ($\mu g\ m^{-3}$)			
2012	**2013**	**2014**	**2015**
19	15	15	12

Table 6. Annual mean PM_{10} concentrations throughout the years in the studied regions.

PM_{10} ($\mu g\ m^{-3}$) - MASP[a]										
2005	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**	**2015**
36	36	39	37	33	39	38	36	33	36	31

PM_{10} ($\mu g\ m^{-3}$) – MARJ[b]										
2005	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**	**2015**
-	-	22	29	44	40	45	43	33	34	-

PM_{10} ($\mu g\ m^{-3}$) - MABH[c]										
2005	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**	**2015**
21.5	25.9	26.1	27.8	27.9	31.3	-	30.4	-	-	-

[a] Annual mean concentrations in the MASP area from 2009 to 2015 (CETESB, 2009, 2010, 2011, 2012, 2013, 2014, 2015)

[b] Annual mean from 2010 to 2014 in Botafogo district, MARJ (INEA, 2013, 2015)

[c] Annual mean concentrations from 2005 to 2012. Due to lack of data, 2011 values were not represented (FEAM, 2005, 2006, 2007, 2008, 2009, 2010, 2012).

Table 7. Air Quality Guidelines and Standards for PM concentrations(CETESB, 2013; CONAMA, 1990; WHO, 2016).

Guidelines ($\mu g\ m^{-3}$)				
		WHO Guidelines	CONAMA Regulation[a]	São Paulo Guidelines For the first phase
$PM_{2.5}$	24h Sample	25	-	60
	AAM[b]	10	-	20
PM_{10}	24h Sample	50	150	120
	AAM[b]	20	50	40

[a] MABH and MARJ both uses CONAMA regulation (CONAMA, 1990)

[b] Annual Arithmetic Mean

Table 8. $PM_{2.5}$ average values at some sites in the three metropolitan areas studied.

$PM_{2.5}$ - $\mu g\ m^{-3}$					
		Summer –AM[a]	Winter – AM[a]	Annual – AM[a]	24h Limit
MASP		23	35.5	28.1	-
Worst Areas	Congonhas Airport	-	-	20	63
	Motorway M. Tietê	-	-	22	-
MARJ		15.8	23	17.2	-
MABH		14.5	18.5	14.7	-

[a] Arithmetic Mean

Table 9. PM_{10} measurements for the three most polluted metropolitan areas studied.

PM_{10} ($\mu g\ m^{-3}$)				
		AAM[a]	**24h-Limit**	**Year**
MASP		31	----	2015
Most polluted areas	Osasco	40	111	2015
	Parelheiros	40	121	2015
MARJ		46.4		2008-09
Most polluted areas	RJ - São Cristovão	-----	159	2014
	RJ – Botafogo	-----	63	2014
	Duque de Caxias	71.1	249	2014
MABH		30.4	----	2012
Most polluted areas	Betim	50.6	200	2012

[a]Annual Arithmetic Mean

Table 10. Mobility aspects of some of the selected cities around the world.

Aspects	**Beijing**	**London**	**Sydney**	**New York**	**Stockholm**

Population (annual rate of change, %)[1]	20 Million (2.0%)	8.3 Million (0.61%)	4.7 Million (0.60%)	8.3 Million (0.77%)	2.1 Million (1.0%)
Year of foundation	1403	43	1788	1624	1252
HDI[2]	0.727	0.907	0.935	0.915	0.907
Geography	Situated at the northern part of the North China Plain	Non-coastal city River Thames running through it	Coastal basin with Tasman Sea to the east and Blue Mountains to the west	Coastal city, located at the meeting of the Atlantic Ocean and Hudson River	Coastal city Situated on the water in Riddarfjärden bay
Climate[2]	Continental monsoon, 11.8°C, 576.9 mm	Oceanic climate, 12.8°C, 601 mm	Humid subtropical, 18.5°C, 1222.7 mm	Humid subtropical, 12.9°C, 1268.5 mm	Humid continental, 6.6°C, 531 mm
Local sources	Coal burning, transportation, industries	Transportation, construction sites	Transportation, industries	Transportation, power plants	Transportation
External sources	Biomass burning	Waste incineration	Biomass burning	Construction sites	Wood burning
Public bus fleet size[1]	21,628	7,500	2,213	4,344	2,114
Rail length (km)[1]	456	436	329	373	110
Public bus daily ridership ('000)[1]	13,788	6,397	625	1,825	814
Metro daily ridership ('000)[1]	6,008	3,641	829	4,521	874
Private Vehicles ('000)[1]	2,862	2,535	2,626	1,777	829

[1]Pan, 2013; [2]UNDP, 2014.