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ESTIMATE OF PUBLIC HEALTH BENEFITS RESULTING FROM AN AIR QUALITY IMPROVEMENT

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“Qualquer estrada, se seguida exatamente até seu fim, leva exatamente a lugar nenhum. Escale a montanha só um pouquinho, para verificar se é mesmo uma montanha. Do topo, não se vê a montanha.”

Excerto de Duna

Frank Herbert, 1965

RESUMO

O material particulado (MP) é tido como um dos principais poluentes que diretamente afetam a saúde a curto e longo prazo. O objetivo deste trabalho foi avaliar os benefícios na saúde pública decorrentes da melhoria da qualidade do ar nos municípios brasileiros com monitoramento de partículas finas ($MP_{2,5}$; ≤ 2,5 μm de diâmetro aerodinâmico) nas regiões metropolitanas da Região Sudeste do Brasil por meio da modelagem. Para esse último caso, utilizou-se o modelo de transporte químico *Weather Research and Forecasting* (WRF) *model coupled with Chemistry* (WRF-Chem) aplicado para o ano de 2015 e a ferramenta PREP-CHEM-SRC para a geração dos campos de emissões de gases traço e aerossóis utilizando o 2° Inventário Nacional de Emissões Atmosféricas por Veículos Automotores Rodoviários junto com fontes de queimadas. As estimativas dos benefícios na saúde basearam-se em estudos epidemiológicos, aplicando uma função log-normal. Os efeitos a longo prazo avaliaram a mortalidade evitável, pela redução da concentração de $MP_{2,5}$, para todas as causas, causas não-acidentais, doenças isquêmicas do coração, cardiovascular e câncer de pulmão. O efeito na saúde pública a curto prazo foi avaliado com base na estimativa da redução de internações hospitalares públicas por município para doenças respiratórias e do aparelho circulatório, decorrentes da redução da concentração de MP_{10} e $MP_{2,5}$ conforme estudos brasileiros de séries temporais que avaliaram esses poluentes. Os cenários-base foram construídos considerando as concentrações ambientais de $MP_{2,5}$ e aquelas modeladas pelo WRF-Chem (MP_{10} e $MP_{2,5}$), enquanto os cenários-controle consideraram os limites sugeridos pela Organização Mundial da Saúde (OMS). Quase 90% das concentrações anuais de $MP_{2,5}$ nas cidades brasileiras com monitoramento foram superiores à diretriz da OMS (10 μg m^{-3}). A cidade de São Paulo apresentou o maior número de mortes evitáveis, com valores variando entre 28.870 ± 9.770 e 82.720 ± 24.550 para todas as causas, de 2000 a 2017. Em relação à modelagem, nas áreas urbanas estudadas, os veículos podem ser considerados como responsáveis pela maior contribuição para a emissão de MP, e o inventário elaborado se mostrou adequado. O total de mortes evitáveis estimadas para as 102 cidades das regiões metropolitanas da Região Sudeste, relacionadas ao $MP_{2,5}$, foi de 32.000 ± 5.300 devido à mortalidade por todas as causas. Com exceção da região metropolitana de São Paulo, as hospitalizações por doenças respiratórias evitáveis foram maiores para o $MP_{2,5}$ em crianças do que para o MP_{10} considerando todas as faixas etárias. Para doenças do aparelho circulatório, foram estimadas 9.840 ± 3.940 internações evitáveis em idosos relacionadas à diminuição das concentrações de $MP_{2,5}$ em todas as cidades. A exposição humana e os efeitos à saúde são endossados como fatores essenciais para a gestão da qualidade do ar urbano. A OMS ressalta que não é possível estabelecer um limite mínimo de concentração de MP, abaixo do qual não ocorreriam efeitos nocivos à saúde. Portanto, um padrão deve considerar restrições locais e regionais, capacidades e prioridades de saúde pública.

Palavras-chave: Material particulado, monitoramento, modelagem, WRF-Chem, PREP-CHEM-SRC, mortalidade, morbidade.

ABSTRACT

Particulate matter (PM) is considered to be one of the primary pollutants that directly affect short- and long-term health. The objective of this study is to evaluate the public health benefits of improved air quality in Brazilian municipalities with fine particle monitoring ($PM_{2.5}$; ≤ 2.5 μm aerodynamic diameter) and in the Brazilian Southeast metropolitan areas through modeling. For the latter case, it was used the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem) applied for 2015 and the PREP-CHEM-SRC tool for the generation of trace gas and aerosol emission fields. Estimates of health benefits were based on epidemiological studies applying a log-normal function. Long-term effects assessed preventable mortality by reducing $PM_{2.5}$ concentration for all causes, non-accidental causes, ischemic heart disease, cardiovascular disease, and lung cancer. The short-term public health effect was assessed based on the estimate of the reduction in public hospital admissions by the municipality for respiratory and circulatory system diseases, resulting from the reduction in the concentration of PM_{10} and $PM_{2.5}$, according to Brazilian time-series studies that evaluated these pollutants. The base scenarios were built considering the environmental concentrations of $PM_{2.5}$ and those modeled by the WRF-Chem (PM_{10} and $PM_{2.5}$), while the control scenarios considered the limits suggested by the World Health Organization (WHO). Almost 90% of annual $PM_{2.5}$ concentrations in Brazilian cities with monitoring were above WHO guidelines (10 μg m^{-3}). The city of São Paulo had the highest number of preventable deaths, with values ranging from 28,870 ± 9770 to 82,720 ± 24,550 for all causes, from 2000 to 2017. Regarding modeling, in the urban areas studied, vehicles can be considered as responsible for the most considerable contribution to the emission of PM, and the inventory elaborated based on the 2nd National Inventory of Atmospheric Emissions by Road Motor Vehicles proved to be adequate. The total avoidable deaths estimated for the 102 cities in the southeastern metropolitan regions, related to $PM_{2.5}$, was 32,000 ± 5,300 due to all-cause mortality. Except for the metropolitan area of São Paulo, hospitalizations for avoidable respiratory diseases were higher for $PM_{2.5}$ in children than for PM_{10} considering all age groups. For circulatory system diseases, 9,840 ± 3,940 avoidable hospitalizations in the elderly were estimated, related to decreased $PM_{2.5}$ concentrations in all cities. Human exposure and health effects are endorsed as essential factors for urban air quality management. WHO points out that it is not possible to establish a minimum limit of PM concentration, below which no harmful effects to health could occur. Hence, a standard should consider local and regional constraints, public health capacities, and priorities. Therefore, a standard should consider the context of local and region constraints, capabilities, and public health priorities.

Keywords: Particulate matter, monitoring, modeling, WRF-Chem, PREP-CHEM-SRC, mortality, morbidity.

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3BEM	*Brazilian Biomass Burning Emission Model*
ACCENT	*Atmospheric Composition Change: the European Network of Excellence*
ACOM	*Atmospheric Chemistry Observations & Modeling*
ACS	*American Cancer Society*
AEROCOM	*Aerosol Comparisons between Observations and Models*
AFWA	*Air Force Weather Agency*
AGA	*Annual Geometric Average*
Aphekom	*Improving Knowledge and Communication for Decision Making on Air Pollution and Health in Europe*
AVHRR	*Advanced Very High Resolution Radiometer*
BC	*Black Carbon*
BenMAP-CE	*Environmental Benefits Mapping and Analysis Program – Community Edition*
BRAMS	*Brazilian developments on the Regional Atmospheric Modelling System*
CAPES	Coordenação de Aperfeiçoamento de Pessoal de Nível Superior
CCATT-BRAMS	*Coupled Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modelling System*
CCC	*Circular Correlation Coefficient*
CETESB	Companhia Ambiental do Estado de São Paulo
CI	*Confidence Interval*
CGIAE	Coordenação-Geral de Informações e Análises Epidemiológicas
CMAQ	*Community Multi-Scale Air Quality*
CONAMA	Conselho Nacional do Meio Ambiente
CPTEC	Centro de Previsão de Tempo e Estudos Climáticos
DATASUS	Departamento de Informática do SUS
DENATRAN	Departamento Nacional de Trânsito
EDGAR	*Emission Database for Global Atmospheric Research*
ESLR	*Earth System Research Laboratory*

FAA	*Federal Aviation Administration*
FEAM	Fundação Estadual do Meio Ambiente
FNL	*Global Forecast System final*
FS	*Final Standard*
FSL	*Forecast Systems Laboratory*
GAM	*Generalized Additive Model*
GDP	*Gross Domestic Product*
GEIA	*Global Emissions InitiAtive*
GEOS-5	*Goddard Earth Observing System Model, version 5*
GEOS DAS	*Goddard Earth Observing System Data Assimilation System*
GFED	*Global Fire Emissions Database*
GFS	*Global Forecast System*
GLM	*Generalized Linear Models*
GOCART[1]	*Goddard Chemistry Aerosol Radiation and Transport*
GOCART[2]	*Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model*
GOES	*Geostationary Operational Environmental Satellite*
GPAMA	Grupo de Pesquisa em Poluição do Ar & Meteorologia Aplicada
ICD	*International Statistical Classification of Diseases and Related Health Problems*
IEA	*International Energy Agency*
IEMA	Instituto de Meio Ambiente e Recursos Hídricos
IHD	*Ischemic Heart Disease*
INEA	Instituto Estadual do Ambiente
INMET	Instituto Nacional de Meteorologia
INPE	Instituto Nacional de Pesquisas Espaciais
IOA	*Index of Agreement*
IS	*Intermediate Standard*

MA	*Metropolitan Area*
MABH	*Metropolitan Area of Belo Horizonte*
MAF	*Metropolitan Area of Fortaleza*
MAGV	*Metropolitan Area of Great Vitória*
MARJ	*Metropolitan Area of Rio de Janeiro*
MASP	*Metropolitan Area of São Paulo*
MB	*Mean Bias*
ME	*Mean Error*
MEGAN	*Model of Emissions of Gases and Aerosols from Nature*
MODIS	*Moderate Resolution Imaging Spectroradiometer*
MOZART-4	*Model for Ozone and Related chemical Tracers, version 4*
MS	Ministério da Saúde
NCAR	*National Center for Atmospheric Research*
NCEP	*National Center for Environmental Prediction*
NLI	*Night Light Intensity*
NMB	*Normalized Mean Bias*
NME	*Normalized Mean Error*
NOAA	*National Oceanic and Atmospheric Administration*
NRL	*Naval Research Laboratory*
OC	*Organic Carbon*
OMS	Organização Mundial da Saúde
OU	*University of Oklahoma*
PBL	*Planatary Bounday Layer*
PREP-CHEM-SRC	*Preprocessor of trace gas and aerosol emission fields for regional and global atmospheric chemistry models*
r	*Correlation Coefficient*
RADM2	*Regional Acid Deposition Model version 2*
RETRO	*REanalysis of the TROpospheric chemical composition over the past 40 yr.*

Ripsa	Rede Interagencial de Informações para a Saúde
RMSE	*Root Mean Square Error*
RR	*Relative Risk*
SIM	Sistema de Informação sobre Mortalidade
SVS	Secretaria de Vigilância em Saúde
SUS	Sistema Único de Saúde
TMS	*Traffic Management Strategies*
US EPA	*United States Environmental Protection Agency*
VEI	*Brazilian Top-Down Vehicle Emission Inventory*
VERDI	*Visual Environment for Rich Data Interpretation*
WF_ABBA	*Wildfire Automated Biomass Burning Algorithm*
WHO	*World Health Organization*
WPS	WRF *Preprocessing System*
WRF	*Weather Research and Forecasting*
WRF-Chem	WRF *model coupled with Chemistry*

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1. INTRODUCTION

1.1 Background and justification

According to the Brazilian National Environment Policy, pollution is defined as the degradation of environmental quality resulting from activities that directly or indirectly harm the health, safety, and well-being of the population, create adverse conditions for social and economic activities, adversely affects biota, the aesthetic or sanitary conditions of the environment, and release materials or energy in disagreement with the established environmental standards (BRAZIL, 1981).

CONAMA Resolution 491/2018 follows this same idea, and defines the atmospheric pollutant as any form of matter or energy, in quantity, concentration, time or other characteristics, which may become the air inappropriate or harmful to health, inconvenient to public well-being, harmful to materials, fauna, and flora, or harmful to security, use, and enjoyment of the property or normal community activities.

Among air pollutants, particulate matter (PM), ozone (O_3), sulfur dioxide (SO_2), nitrogen oxides (NOx), carbon monoxide (CO) and volatile organic compounds (VOC) are the leading indicators of pollution levels (SEINFELD and PANDIS, 2006; CETESB, 2019). The effects of air pollution exposure, on human health, environmental, animals etc., are the result of pollutants emissions into the atmosphere, and its interactions from physical (dispersion) and chemical (chemical reactions) process. (SEINFELD and PANDIS, 2006; CETESB, 2019). Air quality is, therefore, the product of the interaction between factors such as emissions, topography, and weather conditions, for example.

Atmospheric contamination is an urban problem, and a public health issue. Any factor directly related to population health is an item of social welfare, and also an economic item. For coherent and effective environmental health policies, it is necessary to carry out studies on the relationship between air pollution and health, addressing issues such as epidemiological indicators in environmental health and assessing exposure to air pollution. A reduction in atmospheric emissions and, consequently, a reduction in the concentration of atmospheric contaminants has a direct effect on the morbidity and mortality associated with air pollution (OSTRO and CHESTNUT, 1998; POPE III *et al.*, 2002; BELL *et al.*, 2005; LADEN *et al.*, 2006; JERRET *et al.*, 2009; CESARONI *et al.*, 2013; FANN and RISLEY, 2013; ZHOU *et al*., 2014; HART *et al*., 2015; OSTRO *et al*., 2015; WONG *et al*., 2016; BOWE *et al*., 2018; POPE III *et al*., 2019a; POPE III *et al*., 2019b).

Furthermore, air pollution has become a significant risk factor, since it has the most robust causal associations between long-term exposure to the pollutant, and it reduces life expectancy (APTE *et al*., 2018). The most vulnerable populations are children, the elderly and people who already have respiratory diseases due to their physiological peculiarities (GOUVEIA and FLETCHER, 2000a; GOUVEIA and FLETCHER, 2000b; BAKONYI *et al*., 2004; CASTRO *et al*., 2009; GONÇALVES *et al*., 2012; NASCIMENTO *et al*., 2017; GOUVEIA and JUNGER, 2018; FERNANDES *et al.*, 2020).

According to the World Health Organization (WHO), air pollution has become one of the leading risks to environmental health, causing approximately 4.2 million deaths every year as a result of exposure to ambient air pollution (outdoors), which represents 7.6% of all computed deaths worldwide. Among the main causes are cardiovascular diseases, strokes, chronic obstructive pulmonary disease, and lung cancer, in addition to the increased risk of acute respiratory infections (WHO, 2016a). According to Cohen *et al*. (2017), only the fine particulate matter ($PM_{2.5}$) was the fifth most significant risk factor for mortality in 2015, causing an average of 4.2 million deaths worldwide, an increase of 20% concerning the total deaths in 1990. From a local perspective, Miranda *et al*. (2012) showed that Belo Horizonte (Brazil) exceeded 4.7 µg m^{-3} the annual $PM_{2.5}$ average guideline established by WHO (10 µg m^{-3}) between June 2007 and August 2008, which represented an excess mortality of 500 adults over 45 years old in the evaluated period. Apte *et al*. (2018) showed that in Brazil, there is a decrease of 0.46 years in life expectancy due to $PM_{2.5}$, considering the 2016 annual averages. A limit based on the WHO guideline of 10 µg m^{-3} for the annual average of $PM_{2.5}$ would generate a hypothetical gain of one month in life expectancy (APTE *et al.*, 2018).

Air pollution also leads to an increase in state spending, due to the increase in the number of visits and hospitalizations, in addition to the use of medicines (MIRAGLIA *et al*., 2005; MIRAGLIA and GOUVEIA, 2014; RODRIGUES *et al*., 2015; FERNANDES *et al.*, 2020). Such costs could be reduced by improving air quality. Air pollution can also affect the quality of materials (corrosion), soil, and water (acid rain), in addition to affecting visibility (LISBOA, 2014).

The interaction between pollution sources and the atmosphere will define the level of air quality, which in turn determines the appearance of adverse effects of air pollution on receptors. Air quality monitoring aims to provide data to activate emergency actions during periods of atmospheric stagnation, assess air quality against established limits to protect people's health and well-being, enable a correct territory planning, monitor trends and changes in air quality due to changes in pollutant emissions, in addition to providing information for scientific work (LISBOA and KAWANO, 2007; GOMES, 2010).

However, the air quality monitoring network in Brazil is restricted and unsatisfactory in terms of sample history, territorial coverage, the number of parameters monitored, and the representativeness in measurements. The main reasons are related to the managerial difficulties and the low number of technicians involved, as well as the lack of resources for the purchase and maintenance of equipment and monitoring networks (BRAZIL, 2014). Minas Gerais state, for example, currently present 33 automatic monitoring stations, eight of which are located in the Metropolitan Area of Belo Horizonte (MABH), in the cities of Belo Horizonte (two stations), Contagem (one), Betim (three) and Ibirité (two). The other 25 are located in only eight municipalities. Therefore, monitoring covers less than 12% of the municipalities in MABH and 1.5% of the municipalities in Minas Gerais.

In this context, air quality modeling is a complementary tool for monitoring and often economically viable to estimate the impacts caused by emissions in the atmosphere and receptors, and it is also used for projects of air quality monitoring network. An atmospheric model is a representation of the dynamic, physical, chemical, and radiative processes in the atmosphere, described by partial differential equations that are approximated by finite differences or finite volumes, for example, and resolved. In this sense, air quality models help to understand how air pollutants behave in the environment (JACOBSON, 2005; OKE *et al*., 2017).

The transport distances and the time involved in polluting chemical species can be large enough for chemical and physical transformations to occur. The terrain over any region is generally uneven in terms of roughness and thermal characteristics, besides the possibility of present a complex topography. Consequently, the meteorological parameters that affect transport, diffusion, transformation, and removal processes (dry and wet deposition) are functions of time and space. Therefore, a regional air quality model must include detailed information on all-natural and anthropogenic sources, pollutants transported from other regions, land use and land cover, topography, and regional meteorology, including dispersive characteristics (ARYA, 1999). The model then mathematically (or numerically) simulates the transport and dispersion of the pollutant, and depending on the model, its chemical and physical transformations and removal processes. The result of the modeling is the concentration of air pollutants for a specific period, usually in specific locations of the receptors (VALLERO, 2008).

Many studies that aim to assess the relationship between air pollution and mortality used data based on the concentration of pollutants from air quality monitoring stations (RODRIGUES *et al*., 2015; THURSTON *et al*., 2016), which are representative for a small area around them. Air quality modeling, on the other hand, allows a representation of the entire territory studied (municipality by municipality), and its results may be used as a source for such studies (BOLDO *et al*., 2014; DING *et al*., 2016). In this context, the modeling of meteorological conditions and the air quality of a region permits to assess the current level of pollution, to follow trends, to define responsibilities concerning the levels of pollution, to evaluate a possible impact of future emission sources, in addition to studying scenarios of emission reductions.

In this context, the questions of interest to be answered by the present work were: (i) what are the benefits to human health associated with a policy that improves air quality? (ii) what are the levels of particulate matter in the cities of the four Brazilian Southeast metropolitan areas (Belo Horizonte, Great Vitória, Rio de Janeiro, and São Paulo), especially those that do not have air quality monitoring? (iii) what is the impact on human health attributable to the total levels of particulate matter?

To achieve those answers, at first all available data of $PM_{2.5}$ (until 2017) were used to estimate the avoidable deaths related to an improvement in $PM_{2.5}$ annual concentration. Afterward, to estimate the benefit for all cities of the Brazilian Southeast metropolitan areas, air quality modeling was used. At the end of the work, it will be possible to estimate the benefits that an improvement in particulate matter concentration will bring to the health of the local population, in terms of preventable mortality by $PM_{2.5}$ and hospital admissions by $PM_{2.5}$ and PM_{10}. It is also expected to show that air pollution resulting from emissions from capital cities influences the air quality of the surrounding cities.

1.2 General objective

The general objective of this work is to assess the public health benefits of improving air quality using fine particle monitoring data and applying an integrated modeling system.

1.3 Specific objectives

- To estimate the number of avoidable deaths for Brazilian cities with $PM_{2.5}$ monitoring (until 2017) resulting from the application of the WHO annual guideline (10 µg m^{-3});
- To evaluate the use of the 2nd Brazilian National Inventory of Atmospheric Emissions by Road Motor Vehicles as an alternative to global inventories emissions in the generation of aerosol emission fields for use in the WRF-Chem chemical transport model, creating an inventory of vehicle emissions for the Brazilian municipalities;
- To evaluate the concentration levels of particulate matter in the municipalities of the Brazilian Southeast metropolitan areas, by applying an integrated modeling system to simulate the formation and dispersion of PM;
- To estimate the number of reductions in deaths from various causes and public hospital admissions in the four Brazilian Southeast metropolitan areas resulting from the reduction in the $PM_{2.5}$ and PM_{10} concentration.
- To review the epidemiological studies already carried out for $PM_{2.5}$ in Brazil, discussing the use of monitored and modeled data for this purpose.

1.4 Document structure

This document is divided into eight chapters. The first, Chapter 1 (Introduction), aimed to present the background and justification of this work, with the objectives of the study. Chapter 2 presents a bibliographic review. Chapters 3 to 6 contain the results, written in a format of four scientific articles, as submitted/published.

The first article, entitled "*Excess deaths associated with fine particulate matter in Brazilian cities*", are presented in Chapter 3, and corresponds to the first specific objective. The work consisted of investigating the number of avoidable deaths from various causes in Brazilian cities that monitor $PM_{2.5}$. The article was published in

Atmospheric Environment, v. 194, p. 71-81, 2018. Supplementary material cited in the article can be found in the Appendix I. After the publication of the article, CONAMA Resolution 03/1990 was updated by CONAMA Resolution 491/2018, but the discussion raised and the estimates performed remain valid, since the WHO guidelines were used as the control scenario.

The second article (Chapter 4), "*Top-Down Vehicle Emission Inventory for spatial distribution and dispersion modeling of particulate matter*", corresponds to the second specific objective. It presents the methodology for the construction of the vehicle emission inventory used in the next article to estimate the health benefits. The work was presented at the *Air Pollution Conference Brazil 4th CMAS South America* in 2019, and the article is part of the special issue of the congress in *Environmental Science and Pollution Research* journal.

Chapter 5 corresponds to the article entitled "*Quantifying the impact of particle matter on mortality and hospitalizations in four Brazilian metropolitan areas*", published in *Journal of Environmental Management, v. 270, 110840, 2020*, and addresses the third and the fourth specific objectives. One-year modeling with the WRF-Chem model was carried out, and the number of avoidable deaths from various causes and hospital admissions for respiratory causes was estimated for the 102 cities of the four Brazilian Southeast metropolitan areas. Supplementary material cited in the article can be found in the Appendix II.

Closing the results, Chapter 6 presents the fourth article, entitled "*Fine particles as an indicator of public health in Brazil: from monitoring to modeling*", published in *Air Quality, Atmosphere & Health*. Here, a survey of the epidemiological studies already carried out for $PM_{2.5}$ in Brazil is made, discussing the use of monitored and modeled data for this purpose, which corresponds to the fifth specific objective.

Chapter 7 refers to the final considerations of the work, while Chapter 8 presents the references used. Additionally, in the Appendix is presented the tables cited in Chapters 3 and 5.

2. LITERATURE REVIEW

This chapter provides an overview of current Brazilian air quality standards, a contextualization about air quality modeling, a brief description of the numerical models and programs used, as well as a scientific review, highlighting works that used the proposed tools and epidemiological cohort studies that served as the basis for the development of this research.

2.1 Air quality standards

As defined by CONAMA Resolution 491/2018, air quality standards are concentration values of a specific pollutant in the atmosphere, associated with an exposure time interval, so that the environment and population's health are preserved concerning the risks of damage caused by air pollution. The adoption of air quality standards worldwide arose from the perception that marked increases in mortality and morbidity followed short-term episodes of extremely high levels of air pollutants (WHO, 2006).

The same resolution established intermediate standards (IS), as temporary values to be met in stages, and a final air quality standard (FS), based on the guideline values defined by WHO in 2006. The air pollutants legislated, sampling and their respective national standards are shown in Table 2.1. Only in this legislation updated in 2018 that the fine fraction of the particulate material, with an aerodynamic diameter less than 2.5 µm ($PM_{2.5}$), started to be legislated at a national level (WHO, 2006).

The WHO first published guidelines on air quality in 1987 (WHO, 1987). The most recent update was for the year 2005 (WHO, 2006). The review process is based on scientific studies on the effect of pollution on health and also takes into account the opinion of air quality managers and those responsible for public policies regarding the logic and format of the guidelines, to improve its applicability worldwide. Pollutants of concern are PM_{10}, $PM_{2.5}$, SO_2, O_3, and NO_2, as shown in Table 2.1 (column FS in bold). For particulate matter, the guidelines are based on studies that showed, with 95% confidence, an increase in total mortality, cardiopulmonary and lung cancer in response to long-term exposure to the pollutant, and also increase in mortality associated with short-term exposure.

Table 2.1 - Brazilian national air quality standards.

Pollutant	Sampling Time	IS-1 (µg/m³)	IS-2 (µg/m³)	IS-3 (µg/m³)	FS (µg/m³)
Total Suspended Particles (TSP)	24 hours	-	-	-	240
	AGM	-	-	-	80
Smoke	24 hours	120	100	75	50
	AAM	40	35	30	20
Particulate matter (PM_{10})	24 hours	120	100	75	**50**
	AAM	40	35	30	**20**
Particulate matter ($PM_{2,5}$)	24 hours	60	50	37	**25**
	AAM	20	17	15	**10**
Sulfur dioxide (SO_2)	24 hours	125	50	30	**20**
	AAM	40	30	20	-
Carbon monoxide (CO)	8 hours	-	-	-	9 ppm
Ozone (O_3)	8 hours	140	130	120	100
Nitrogen dioxide (NO_2)	1 hour	260	240	220	**200**
	AAM	60	50	45	**40**
Lead (Pb)*	AAM	-	-	-	0,5

Notes: * Measured in total suspended particles; AGM: annual geometric mean; AAM: annual arithmetic mean. In bold, patterns that match WHO guidelines are highlighted.
Source: CONAMA Resolution 491/2018; WHO (2006).

The intermediate and final air quality standards will be subsequently adopted, taking into account control plans of pollutant emissions and air quality assessment reports prepared by state and district environmental agencies, however, without a deadline. Furthermore, IS-1 values for the main pollutants (PM_{10}, $PM_{2.5}$, and SO_2) are still very permissive compared to their respective final standards and WHO guidelines, as discussed by Siciliano *et al*. (2020).

The implementation of legislation to improve air quality is inherent to each country. It should consider its economic situation and the concentrations that would be observed without anthropic pollution (background concentration). Even with the concentrations of these pollutants below the recommended limit values, the effects on health can occur, as there are still many research gaps (OHANDJA *et al*., 2012).

2.2 Air quality modeling

One of the main objectives of the study of atmospheric behavior of contaminants is to be able to describe their spatial and temporal distribution mathematically after being released into the atmosphere. Atmospheric models can be broadly divided into two types: physical and mathematical. Physical models (or experimental methods) are sometimes used to simulate atmospheric processes using a small-scale representation of a real system and are divided into wind tunnel studies and field experiments. Mathematical models are separated into deterministic models, based on the fundamental description of physical and chemical processes in the atmosphere (dispersion models and chemical transport models), and statistical models, based on statistical data analysis (receptor model, for example) (SEINFELD and PANDIS, 2006).

Atmospheric dispersion modeling refers to the mathematical description of the transport of contaminants in the atmosphere. The term *dispersion* is used to describe the combination of diffusion and advection that occurs within the layer close to the Earth's surface (STOCKIE, 2011). The qualitative aspect of the dispersion theory is to describe the fate of emission to the atmosphere from a point source, line, area, and volume source. Quantitatively, the dispersion theory provides an estimate of a substance concentration in the atmosphere. It also provides specific information about meteorological factors and the characteristics of the emitting sources. Dispersion models include *Eulerian models*, in which the behavior of species is described concerning a fixed coordinate system, with an array of fixed computational cells, and *Lagrangian models*, in which the changes in concentration are described regarding the fluid in motion (SEINFELD and PANDIS, 2006; TIWARY and COLLS, 2010).

Chemical transport models have become widely recognized and are used as tools for regulatory analysis and evaluation of control strategies (ALBUQUERQUE *et al.*, 2019). They are models that simulate changes in pollutant concentrations in the atmosphere using a set of mathematical equations that characterize the chemical and physical processes in the atmosphere. They are applied at multiple spatial scales, from local to global (VALLERO, 2008).

Air quality models require weather and soil data as input data. Meteorological processes, such as horizontal and vertical transport, turbulent mixing, convection, control, or influence the evolution of emissions, gaseous chemical species, and aerosols. Also, the formation of secondary pollutants is directly affected by relative humidity, solar radiation, temperature, and presence of clouds (SEAMAN, 2000). Representative and data from meteorological stations and radiosonde are challenging to be found, especially in the reading format that the models require. Therefore, numerical models that generate meteorological fields, for example, the *Weather Research and Forecasting* (WRF) model (SKAMAROCK *et al*., 2008) and the *Brazilian developments on the Regional Atmospheric Modelling System* (BRAMS) (FREITAS *et al.*, 2017), are an increasingly used alternative.

2.3 Weather Research and Forecasting (WRF)

The *Weather Research and Forecasting* (WRF) model (SKAMAROCK *et al*., 2008) is an atmospheric model designed for both research and numerical weather forecasting. Its development began in the 1990s and was a result of collaboration between the *National Center for Atmospheric Research* (NCAR), the *National Oceanic and Atmospheric Administration* (NOAA), *National Center for Environmental Prediction* (NCEP), *Forecast Systems Laboratory* (FSL), *Air Force Weather Agency* (AFWA), *Naval Research Laboratory* (NRL), *University of Oklahoma* (OU), and *Federal Aviation Administration* (FAA). WRF has become a real community model for its long-term development through the interests and contributions of worldwide user base.

The WRF atmospheric simulation process has two stages. In the first stage, the domains are configured to represent the area of interest, the input data is inserted, and the initial conditions are prepared. The second step is the execution of the forecast model. Figure 2.1 shows a simplified scheme of modeling with WRF.

The simulations with WRF start with the execution of the *WRF Preprocessing System* (WPS) in three stages. The first is the execution of the *geogrid*, in which the model configures the domain horizontally based on the geographical information provided (topography and land use and land cover). The second is the execution of *ungrib*, which is responsible for extracting and reformatting meteorological data for the domain. The third step is the execution of the *metgrid*, in which the meteorological fields are horizontally interpolated. After these steps, the input fields are placed at the vertical levels of the model, and the lateral boundary conditions are generated by executing *real.exe*.

WRF is ready to run (*wrf.exe*) by the forecast component that contains the dynamic resolution and physics packages for atmospheric processes (for instance, microphysics, radiation, and planetary boundary layer). Post-processing (ARWpost) is performed using several free tools (IDV, STEAM, VERDI, and Grads, python, for example).

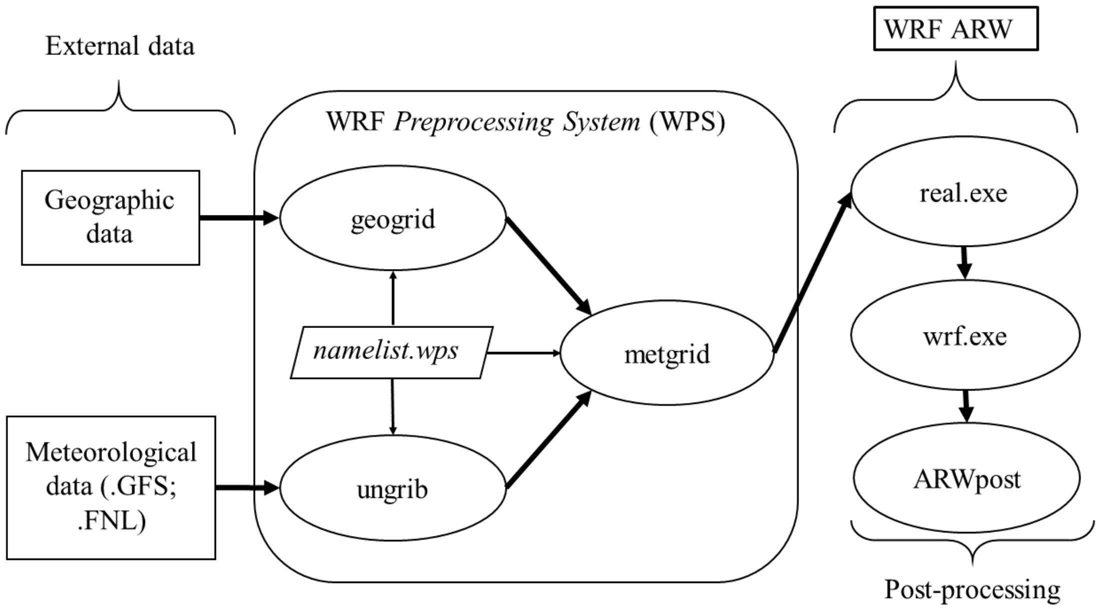

Figure 2.1 - Simplified modeling scheme with WRF.
Source: Adapted from Skamarock *et al*. (2008) and WRF-ARW V3: User's Guide (2017).

An essential factor in modeling with WRF is the choice of appropriate parameterizations to represent the physical processes that occur in the atmosphere, and that can be combined in different ways. The parameterization aims to translate the physical processes mathematically through specific equations. The options range from simple and efficient parameterizations to sophisticated and computationally more expensive parameterizations, and from newly developed schemes to well-tried schemes.

Parameterizations of atmosphere physics are separated into five major groups: (1) microphysics, (2) radiation (subdivided into longwave and shortwave radiation), (3) land occupation models (surface, land, and urban surface layer), (4) parameterization of the planetary boundary layer (PBL), and (5) parameterization of cumulus. Table 2.2 shows the number of parameterizations for each physical process for WRF version 3.9.

Table 2.2 - Number of physics parameterizations present in the WRF v.3.9 model.

Process	**Number of available parametrizations**
Microphysics (mp_physics)	18
Longwave Radiation (ra_lw_physics)	06
Shortwave Radiation (ra_sw_physics)	08
Surface Layer (sf_sfclay_physics)	08
Land Surface (sf_surface_physics)	09
Urban Surface (sf_urban_physics)	03
Planetary Boundary layer (bl_pbl_physics)	13
Cumulus Parameterization (cu_physics)	14

Source: WRF-ARW V3: User's Guide (2017).

The choice of a group of parameterizations that are the most adequate to the conditions of a region is an essential step in the simulation with WRF, and the available computational capacity must be taken into account.

Since the WRF is a mesoscale model, the researches explore the range of topics in mesoscale meteorology and synoptic processes, such as hurricanes (MOON and NOLAN, 2015; KHAIN *et al*., 2016), cyclones (KIM *et al*., 2015; LAKSHMI *et al*., 2017), fronts (CONRICK *et al*., 2016; PASSALACQUA *et al*., 2016), sea breezes (SALVADOR *et al*., 2016a), and extratropical jets (PARISH and CLARK, 2017). The use of the WRF model in regional climate surveys has been increasing in recent years. As examples, the works of Avolio *et al*. (2017) in which the authors evaluated the sensitivity of the PBL parameterizations available in WRF for Calabria region, in southern Italy, in an area characterized by a complex orography close to the sea; Göndöcs *et al*. (2017), who assessed the intensities of urban heat islands and meteorological variables in Budapest, Hungary; Takebayashi and Senoo (2017), who analyzed the relationship between urban size and heat island intensity in three Japanese cities (Tokyo, Osaka and Nagoya); Rafael *et al*. (2019) evaluated different urban surface parameterizations for Portugal.

In Brazil, several studies have used the WRF model. Santiago (2009) performed the simulation of PBL temporal and spatial behavior over the Metropolitan Area of Greater Vitória (MAGV) in Espírito Santo. The results obtained indicated that the model was able to predict the PBL variation patterns analyzed reasonably, but with some significant deviations between the predictions and experimental observations.

Ruiz *et al.* (2010) tested different parameterizations to identify the one that would provide the best estimates of observed surface variables, in a domain that covered much of South America. The authors identified that the surface variables are highly sensitive to the choice of land surface models. The Noah topsoil model well represented the surface temperature, but the dew point temperature was better estimated by a simpler model, which specifies the soil moisture based on climatology.

Zepka *et al.* (2014) presented a lightning forecast method for Brazilian southeastern using a combination of meteorological variables obtained from simulations with WRF, where different convection and microphysical schemes were applied. Through statistical evaluations, the combination of Grell-Devenyi and Thompson microphysical parameterizations described better the convective storms with lightning.

Abreu and Rocha (2015) used the WRF to simulate the subtropical cyclone Anita, which occurred in March 2010 in the southwest sector of the South Atlantic Ocean. Simulations with 24h in advance were more similar to the observations, while that 72h in advance showed more significant errors in precipitation, intensity, and position of the cyclone. The Betts-Miller-Janjic convective parameterization resulted in variables closer to those observed.

Salvador *et al.* (2016b) evaluated the accuracy of parameterizations in detecting the formation and attributes of the Internal Boundary Layer (IBL), which is formed by sea breezes, for MAGV. The simulation that used YSU parameterization for the atmospheric boundary layer for non-local closure, associated with the Noah topsoil model, presented slightly better performance than the other tested combinations.

2.4 PREP-CHEM-SRC

The most crucial component of an air quality model is the sources of air pollutants generated in urban areas or on a regional scale, as well as the pollutants transported from other regions. An inventory of emissions, including information of daily, weekly, and monthly emission variations, is critical for effective air quality modeling (ARYA, 1999; SANTOS *et al.*, 2019; PINTO *et al.*, 2020a). Emission inventories are usually provided by environmental agencies or companies, from single sources, reporting only the annual emission rate (tons per year), mainly for each pollutant legislated in a given latitude and longitude. As the air quality models need, as input data, 3D emissions, spatialized in an area and varying in time, it is necessary to process the primary data and transform them into a suitable format to be able to apply them in the quality models. However, when working on a regional or larger scale in Brazil, it is usually necessary to use global inventories.

The *Preprocessor of trace gas and aerosol emission fields for regional and global atmospheric chemistry models* (PREP-CHEM-SRC) is a software, developed by *Centro de Previsão de Tempo e Estudos Climáticos* (CPTEC/INPE), and made available internationally, to provide grid and trace gas emissions with flexible spatial resolution, various projections, and for use in regional and global air quality models (FREITAS *et al.*, 2011). The emission fields generated can be used by diverse air quality models. However, this was initially developed to be applied to the Brazilian air quality model, the *Coupled Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System* (CCATT-BRAMS) (BELA *et al.*, 2015; OLIVEIRA *et al.*, 2016) and later adapted for use in the American model WRF-Chem (WRF model coupled with chemistry) (ARCHER-NICHOLLS *et al.*, 2015; BELA *et al.*, 2015; GOVARDHAN *et al.*, 2015; IRIART and FISCH, 2016). Due to file format limitations, there is no studies involving the Brazilian emissions processing tool and the *American Community Multi-Scale Air Quality* (CMAQ) model in the literature.

To run PREP-CHEM-SRC, it is necessary to enter emission data from various categories of sources. When the study area covers a vast territory, it is usually necessary to use a global database to perform simulations of atmospheric emissions. For urban and industrial emissions (anthropogenic emissions inventory), PREP-CHEM-SRC can use the *REanalysis of the TROpospheric chemical composition databases over the past 40 yr.* (RETRO), and the *Emission Database for Global Atmospheric Research* (EDGAR). For the South American continent, a regional inventory of urban emissions suitable for local and regional scale applications is also available. This database integrates information from local vehicle emission inventories using socioeconomic data, extrapolating emissions to cities that do not have local inventories, and geographic distribution of emissions in different spatial resolutions (ALONSO *et al.*, 2010; FREITAS *et al.*, 2011). For aerosols, the *Goddard Chemistry Aerosol Radiation and Transport* (GOCART) database are provided.

For biogenic emissions, PREP-CHEM-SRC can use the *Global Emissions InitiAtive* (GEIA)/*Atmospheric Composition Change: the European Network of Excellence* (ACCENT) *Activity on Emission Databases* or the *Model of Emissions of Gases and Aerosols from Nature* (MEGAN) databases. Emissions from biomass burning and plume rise model are provided by the *Brazilian Biomass Burning Emission Model* (3BEM), through satellite fire detection or by *Global Fire Emissions Database* (GFED) (FREITAS *et al*., 2011). Emissions from the use of biofuels and the burning of agricultural waste are also available, according to the methodology developed by Yevich and Logan (2003).

The PREP-CHEM-SRC system uses the database developed by Mastin *et al*. (2009) to determine ash emission fields during volcanic eruptions. Volcanic SO_2 emissions are provided by *Aerosol Comparisons between Observations and Models* (AEROCOM) (FREITAS *et al*., 2011).

The emissions generated by PREP-CHEM-SRC are made for speciation RADM2 and GOCART (ARCHER-NICHOLLS *et al*., 2015). The *Regional Acid Deposition Model version 2* (RADM2) (CHANG *et al.*, 1989) is widely used in atmospheric models to predict concentrations of oxidants and other air pollutants and includes 59 chemical species and 157 reactions. Among the aerosol modules, the *Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model* (GOCART) (CHIN *et al.*, 2000), a bulk aerosol scheme for reactive species, has particle size information for non-reactive species (dust and sea salt). No secondary organic aerosol is considered in this approach. Therefore, only the total mass of aerosol compounds is known. GOCART includes 14 defined aerosol species and a 15th variable representing unspecified aerosol contributions (P25). The 14 species of aerosols defined are: sulfate; hydrophobic and hydrophilic organic carbon (OC1 and OC2 respectively); elemental hydrophobic and hydrophilic carbon (BC1 and BC2 respectively); dust in five particle sizes (effective radii of 0.5, 1.4, 2.4, 4.5, and 8.0 µm, referred to as D1, D2, D3, D4, and D5, respectively); and sea salt in four particle size distributions (effective radii of 0.3, 1.0, 3.25, and 7.5 µm for dry air, referred to as S1, S2, S3, and S4, respectively) (PENG *et al*., 2017). Due to its simplicity compared to other aerosol schemes, GOCART is numerically efficient.

Stuefer *et al*. (2013) used the PREP-CHEM-SRC to determine the necessary volcanic eruption parameters in a study that sought to include ash and SO_2 emissions from volcanic eruptions in WRF-Chem model. PREP-CHEM-SRC provided the location of volcano to the nearest domain cell and the emission parameters (mass eruption rate, pollutant plume height, and duration). This information was then used within WRF-Chem to determine the vertical distribution of the eruptive mass.

França *et al*. (2014) estimated the annual emissions associated with the practice of burning pre-harvest sugarcane in São Paulo state (Brazil) based on remote sensing maps and emission and combustion factors for burning cane straw. The inventories of sugarcane burning emissions for São Paulo state from 2006 to 2011 were built within the PREP-CHEM-SRC using the 3BEM database. A comparison between five annual inventories built from different approaches showed general agreement regarding the spatial location of emissions in São Paulo state.

Govardhan *et al*. (2016) generated the emissions of precursor gases and aerosols for India in simulations with WRF-Chem using PREP-CHEM-SRC. Chemical emissions from three different databases were used: RETRO for different precursors and greenhouse gases; EDGAR for CO, NO, NH_3 and VOC emissions; and GOCART for black carbon (BC) and organic carbon (OC), in addition to SO_2.

Mataveli *et al*. (2019) aimed to characterize and find trends in $PM_{2.5}$ from fires in the Brazilian *Cerrado* region between 2002 and 2017, using the PREP-CHEM-SRC emissions pre-processing tool and the MODIS data set for this purpose. Spatially, it was found that each cell in the 0.1-degree grid emitted, on average, 0.5 t km^{-2} $year^{-1}$ of $PM_{2.5}$ associated with fires, but values of up to 16.6 t km^{-2} $year^{-1}$ could be observed in a single cell.

2.5 WRF model coupled to Chemistry (WRF-Chem)

Chemical transport models structure the atmosphere as a volume modeled with a three-dimensional grid with a defined number of cells. Each cell can be seen as a box. The boxes are stacked on top of each other and differ in height. Shorter boxes represent the air parcels closest to the ground surface. The model calculates the concentrations of pollutants in each cell, simulating the movement of air into and out of cells by advection and dispersion. The model also includes algorithms to simulate the vertical mixing of pollutants between layers, the introduction of emissions from sources in each cell, as well as sets of chemical reactions, equations of pollution precursors, and meteorology, especially the solar radiation received in each cell (VALLERO, 2008).

Among the chemical transport models, WRF-Chem (Grell *et al*., 2005), developed by *National Center for Atmospheric Research* (NCAR)/*Earth System Research Laboratory* (ESRL), is one of the most cited in the literature and widely used by the academic community, mainly because it is an open-source model. The chemical component of the model treats a variety of coupled physical and chemical processes, such as advection, diffusion, dry deposition, gas-phase chemistry, emissions, distribution, parameterization, aerosol chemistry, and photolysis rate. Both WRF and WRF-Chem use the same transport, grid, time step, and physics schemes, for example.

The WRF-Chem is an online model, resolving the meteorology and the chemical transport at the same time. Therefore, important information about atmospheric processes with smaller time scale than the output time of the meteorological model (wind speed and direction, rainfall, and cloud formation, for example) are considered, besides the radiative aerosol feedbacks and cumulus radiation feedback (GRELL *et al*., 2005).

The air quality modeling with WRF-Chem, as shown in Figure 2.2, with PREP-CHEM-SRC, follows several steps: preparation of files for the generation of the meteorological field for the study domain; entry of emissions inventory; establishment of initial and boundary conditions; meteorological modeling; and transport and chemical reactions of pollutants by WRF-Chem (*wrf.exe*).

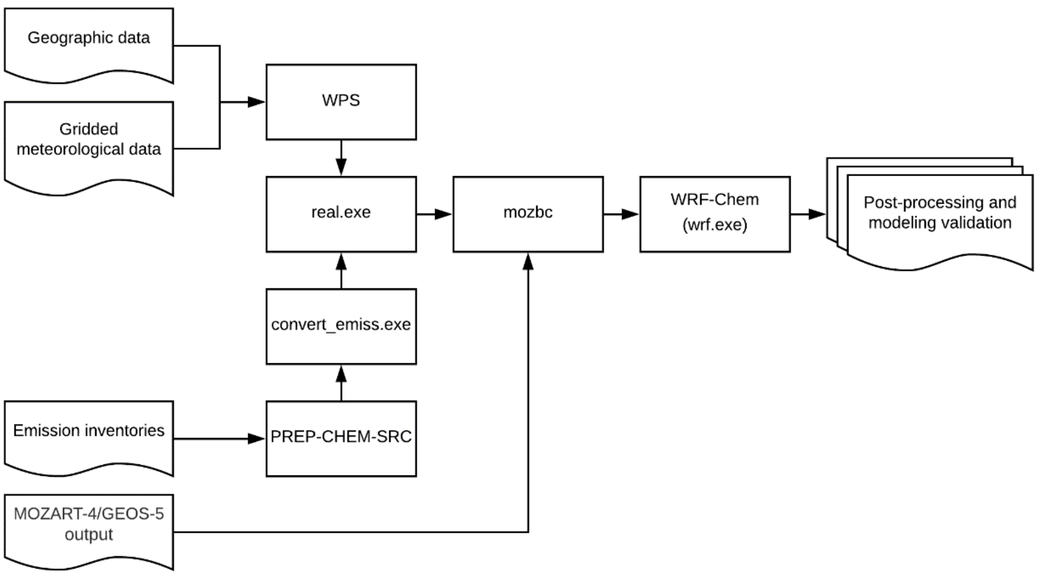

Figure 2.2 - Simplified schematic of the modeling with WRF-Chem.

The *real.exe* is the pre-processor responsible for creating meteorological initial and boundary conditions, as well as the initial chemical conditions for each cell in the domain and the chemical boundary conditions if a fixed profile is used (constant in space and in time). If it is intended to use chemical variable conditions in time and space (dynamic in time and not uniform in space), it is necessary to use external tools, such as *mozbc*. The *mozbc*, developed by NCAR/*Atmospheric Chemistry Observations & Modeling* (ACOM), is responsible for creating time-varying chemical lateral boundary conditions and initial conditions for WRF-Chem from the output of global models, such as the *Model for Ozone and Related chemical Tracers, version 4* (MOZART-4)/*Goddard Earth Observing System Model, version 5* (GEOS-5), being the most used tool in WRF-Chem for this purpose (GAVIDIA-CALDERÓN *et al*., 2018).

Ideally, these initial and boundary chemical conditions should be based on observations, but this is not feasible due to a large number of species and the vast spatial domains manipulated by these models (HOGREFE *et al*., 2017). Therefore, initial concentrations are often based on estimated climatic conditions or global-scale models, while boundary conditions are often derived from global-scale models or predicted concentrations with a larger modeling domain. Problems in defining these conditions generally cause inconsistencies in results and are sometimes not sufficient to adjust the modeling of the study area (BORGE *et al*., 2010; PFISTER *et al*., 2011; GAVIDIA-CALDERÓN *et al*., 2018; PEDRUZZI *et al*., 2019).

The initial conditions influence depends on the geographical domain, and the chemical species, with its influence, decreases with the simulation time. Therefore, it is necessary to start the simulation a few days before the period intended to be evaluated (spin-up).

Jiménez *et al*. (2007), in a case study in the northeast of the Iberian Peninsula, evaluated that a two-day spin-up period was sufficient to reduce the impact factor of the initial conditions to 10% or less for O_3. Hogrefe *et al*. (2017), using a 12 km horizontal grid spacing over the continental USA, showed that a ten-day spin-up period, commonly used in regional scale applications, may not be enough to reduce the effects of initial conditions to less than 1% of surface ozone average concentrations. Twenty days was considered an adequate period, although, in the simulated summer, a period of 30 days was not enough to reduce the effects of the initial conditions by less than 1% on the southwestern portion of modeling domain due to the mass circulation of air.

Unlike initial conditions, boundary conditions, especially those against the wind, continue to affect predictions throughout the simulation. Uncertainties in the predictions of pollutant concentrations in air quality models applied on an urban scale as a result of uncertainties in lateral boundary conditions can be reduced by applying a model on a larger scale (regional scale, for example) to provide the boundary conditions for the urban scale model, in a technique called *nesting* (SEINFIELD and PANDIS, 2006). Alternatively, Samaali *et al*. (2009) and Borge *et al*. (2010) also suggest: (a) extending the modeling domain enough to include all emission sources that affect the atmospheric composition, which could produce a vast domain and result in a high computational expense, along with the need of emission data for the entire domain; and (b) implicitly include the effect of relevant sources through concentration values/profiles at the domain boundaries.

There are several studies involving the use of WRF-Chem model. Hoshyaripour *et al*. (2016) used the model to predict ozone concentrations at ground level in São Paulo. Vara-Vela *et al*. (2016) quantified the impact of vehicular emissions on the formation of fine particles in the Metropolitan Area of São Paulo (MASP). Gavidia-Calderón *et al*. (2018), verified the influence of the use of two chemical boundary conditions in WRF-Chem: the standard model (fixed profile) and other dependent on time (output of the MOZART-4/GEOS-5 model), in the ozone formation in MASP. Vara-Vela *et al.* (2018) investigated the impact of biomass burning sources on aerosol over MASP. Franco *et al.* (2019a) studied the impact of different representations of urban landcover descriptions on meteorology and pollutant concentrations in MASP. Kedia *et al*. (2019) studied the impact of aerosols on the convective and non-convective distribution of rainfall in India. Gueye and Jenkins (2019) verified the influence of the horizontal grid spacing (100, 50, and 18 km) on the PM_{10} concentration for 2012 over West Africa. Sha *et al*. (2019) applied the WRF-Chem model to simulate the chemical components of $PM_{2.5}$ in Nanjing (China). Zhang *et al*. (2020) assessed $PM_{2.5}$ and O_3 concentrations Beijing-Tianjin-Hebei region (China) under emission control scenarios, where it was shown that the application of the new vehicle emission standards from the region and improving fuel quality are effective policies.

2.6 Environmental Benefits Mapping and Analysis Program – Community Edition (BenMAP-CE)

The *Environmental Benefits Mapping and Analysis Program – Community Edition* (BenMAP-CE) (SACKS *et al*., 2018) is an open-source computer program that calculates the number and economic value of deaths and illnesses related to air pollution. The *United States Environmental Protection Agency* (US EPA) and its partners developed the software to attend the analysis needs of different users such as researchers, public policy analysts, and decision-makers. BenMAP-CE estimates the benefits of improvements in human health, such as reductions in the risk of premature death, heart attacks, and other adverse health effects. These analyses are a critical component of air quality policy assessments (US EPA, 2018).

To estimate health effects, BenMAP-CE first determines the change in ambient air pollution. It is used user-specified air quality data (data modeled or generated from air pollution monitoring) for two scenarios, usually, one representing the current conditions and another future, with reduced concentrations of pollutants. Then the relationship between pollution and specific health effects is applied, often referred to as the health impact function or the concentration-response (C-R) function (Equation 2.1) with the exposed population. In this way, different scenarios simulating changes in air quality can be obtained.

$$\Delta Y = Y_o \cdot \left(1 - e^{-\beta \cdot \Delta Q}\right) \cdot Pop \qquad (2.1)$$

where ΔY represents the change in the population's health response; Y_o is the incidence of the evaluated effect for the base case; β is the estimated effect; ΔQ represents the change in air quality, and; Pop is the exposed population.

The change in air quality is the difference between the initial level of air pollution (base scenario) and the level of air pollution after some change (control scenario). Epidemiological studies do not report the C-R function, but instead, some measure of the change in the population's health response associated with a specific change in concentration of pollutants. The most common measure reported is the relative risk (RR) associated with a given change in the concentration of pollutants. When the epidemiological studies use the Cox proportional hazards model or log-linear model to estimate the RR, the value of β can be calculated according to Equation 2.2. The health incidence rate is an estimate of the average number of people who die (or suffer from an adverse health effect) in a given population in a period. Figure 2.3 summarizes the necessary steps in BenMAP-CE.

$$\beta = \ln(RR)/\Delta Q \tag{2.2}$$

In Equation 2.2, ΔQ refers to the change in air quality that the epidemiological study used to estimate the RR, and which is commonly equal to 10 µg m^{-3}.

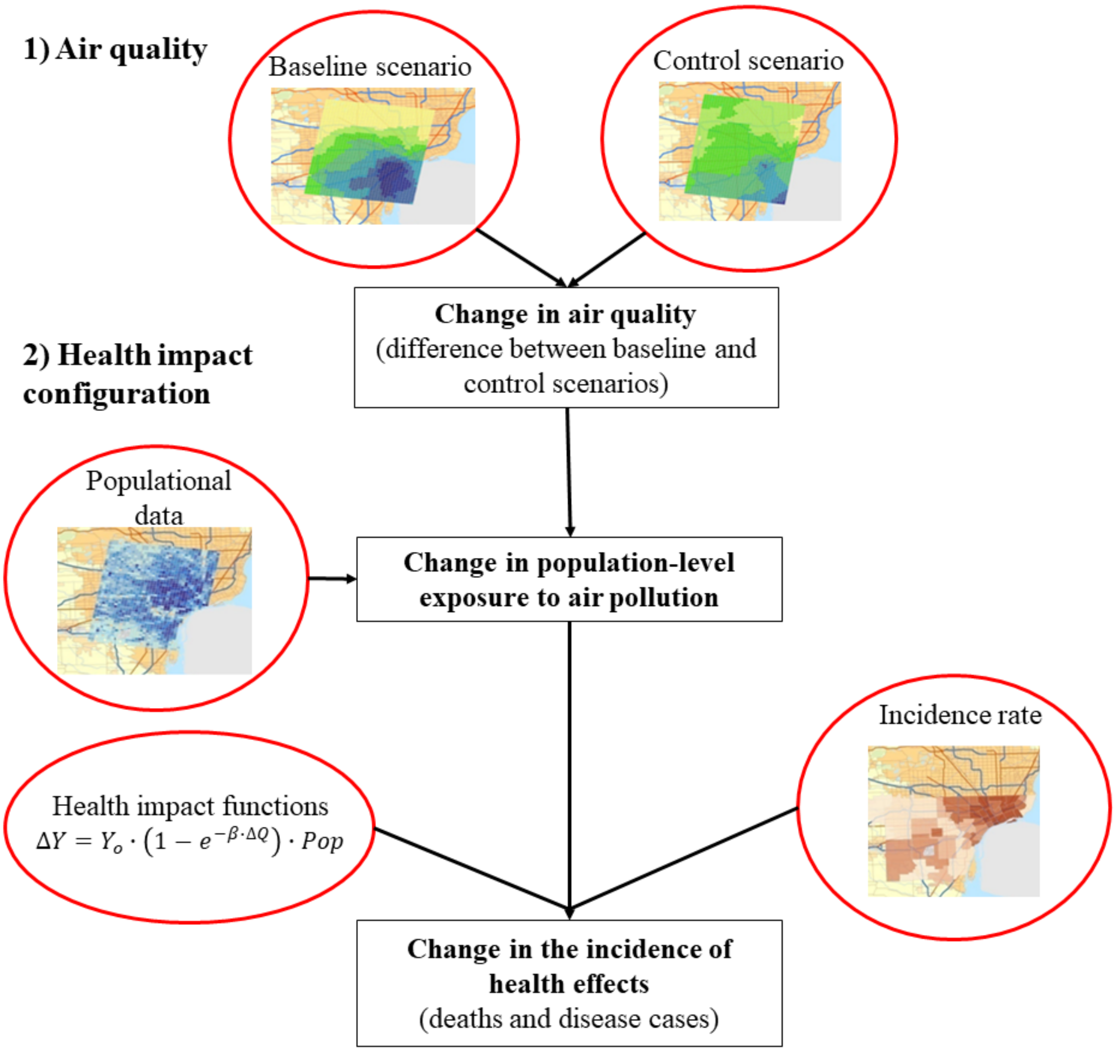

Figure 2.3 - BenMAP-CE flow diagram.
Source: Adapt from US EPA (2018).

For the application of Equation 2.1 in BenMAP-CE, a shapefile of the interest domain, the incidence of the assessed cause, population data, a function of impact on health, baseline, and control scenarios for the evaluated pollutant are needed. For the United States and China, these databases, except the scenarios, are already available in BenMAP-

CE, after its installation. For other locations, these information needs to be inserted into the program.

As stated earlier, epidemiological studies are sources of estimates of the effect of a pollutant on health. The relationship between changes in air pollution levels in the short-term and changes in various indicators of population or individual's health is studied in time series, panels, and case schedule studies. The estimation of chronic health effects associated with air pollution is carried out through cohort studies, which examine the risk of a health outcome (e.g., death) concerning medium to long-term exposure to air pollution, usually comparing people living in different geographic locations (EFTIM and DOMINICI, 2005; WHO, 2006).

Cohort studies generally provide higher estimates of pollution effects than time-series studies, indicating that long-term exposures have a more significant effect than short-term exposures (EFTIM and DOMINICI, 2005). The disadvantages in carrying out this type of study are logistical difficulties, high cost of implementation, monitoring of study populations over long periods with great potential for losses, and a large number of individuals generally needed. Also, as exposure is generally considered to be an average for the entire city, it is necessary to assess different locations to ensure adequate exposure variability (WHO, 2006).

Most cohort studies in the air pollution literature focused mainly on mortality and provided the complete estimates of the deaths number attributable to exposure to pollution and the extent of the average reduction in life expectancy. Therefore, they might be considered more suitable for health impact assessment (COHEN *et al*., 2004; CHEN *et al*., 2008). Vodonos *et al*. (2018) conducted a meta-regression study applied to cohort studies referring to $PM_{2.5}$. A total of 53 studies were selected from 29 cohort studies that provided 135 estimates of the quantitative association between mortality risk and exposure to $PM_{2.5}$. Of the total studies evaluated, 39 studies (18 cohorts) were from North America, eight studies (six cohorts) from Europe, and six studies (five cohorts) from Asia. These numbers highlight the lack of such a study for South America. Therefore, any estimate of avoidable mortality associated with an improvement in air quality in Brazil should use international studies. Subsequently, some cohort studies regarding $PM_{2.5}$ are presented.

The study by Pope III *et al*. (2002) aimed to assess the relationship between long-term exposure to air pollution from fine particles and all-cause mortality, cardiopulmonary, and lung cancer. Vital status and cause of death data were collected by the *American Cancer Society* (ACS) as part of the Cancer Prevention II study, which enrolled approximately 1.2 million adults in 1982. The risk factor data for approximately 500,000 adults were linked with air pollution data for metropolitan areas across the United States and combined with vital and cause of death data of December 31, 1998. The authors also obtained $PM_{2.5}$ data in 116 metropolitan areas collected from 1999 and in the first three quarters of 2000. Each increase of 10 $\mu g\ m^{-3}$ in fine particle concentration was associated with an average increase in the risk of mortality of 6% (Confidence Interval - 95% CI: 1.02-1.11), 9% (1.03-1.16) and 14% (1.04-1.23) for all causes, cardiopulmonary and lung cancer, respectively (considering the average of two periods).

Pope III *et al*. (2004) evaluated long-term exposure to air pollution by fine particles associated with specific cardiopulmonary diseases. Vital status, risk factor, and cause of death data were collected by the American Cancer Society as part of the Cancer Prevention II study and were linked to air pollution data from metropolitan areas in the United States. Long-term $PM_{2.5}$ exposures were most strongly associated with mortality attributable to ischemic heart disease, arrhythmia, heart failure, and cardiac arrest. For ischemic heart disease, the RR (95% CI) found was 1.18 (1.14-1.23), for an increase of 10 $\mu g\ m^{-3}$ in $PM_{2.5}$. The study distinguishing smokers, ex-smokers and people who never smoked. They concluded that although smoking is a much higher risk factor for mortality from cardiovascular disease, exposure to fine PM imposes effects that appear to be at least additives, if they are not synergistic with smoking.

The *Harvard Six Cities* adult cohort study (DOCKERY *et al*., 1993) showed a long-term association between $PM_{2.5}$ and mortality, evaluating a period between the mid-1970s and 1990s, involving an American population of 8,111 adults from six cities. Laden *et al*. (2006) extended the *Harvard Six Cities Study's* analysis time by eight years, encompassing a period of reduced concentrations of air pollutants. The authors found an increase in the assessed mortality associated with each increase of 10 $\mu g\ m^{-3}$ in $PM_{2.5}$. The RR (95% CI) were: 1.16 (1.07-1.26) for all causes; 1.28 (1.13-1.44) for cardiovascular; 1.27 (0.96-1.69) for lung cancer; and 1.08 (0.79-1.49) for respiratory diseases.

Part of the cohort study by Krewski *et al*. (2009) consisted of assessing the relationship between $PM_{2.5}$ and mortality, with approximately 500 thousand participants in areas with adequate information for the year 2000 in the United States. The RR (95% CI) found for the period analyzed from 1999-2000 were 1.03 (1.01-1.05) (all causes), 1.09 (1.06-1.12) (cardiopulmonary), 1.15 (1.11-1.20) (ischemic heart disease) and 1.11 (1.04-1.18) (lung cancer) for an increase in $PM_{2.5}$ concentration of 10 µg m^{-3}.

Lepeule *et al*. (2012) added 11 years of follow-up to the *Harvard Six Cities Study* (DOCKERY *et al*., 1993), covering the period from 1974 to 2009, which ended up incorporating smaller exposures to fine particles. The RR (95% CI) calculated for an increase of 10 µg m^{-3} in $PM_{2.5}$ was 1.14 (1.07-1.22) for mortality due to all causes, 1.26 (1.14-1.40) for cardiovascular and 1.37 (1.07-1.75) for lung cancer. Compared to the study by Laden *et al*. (2006), there is a decrease in RR for all-cause and cardiovascular mortality and an increase in lung cancer.

In the study by Jerrett *et al*. (2009), *American Cancer Society II* cohort data from the *American Cancer Society* was correlated with air pollution data from 96 metropolitan areas in the United States. In models considering only one pollutant, an increase in concentrations of $PM_{2.5}$ was significantly associated with an increased risk of death from cardiopulmonary causes. Considering two-pollutant models, $PM_{2.5}$ was associated with the risk of death from cardiovascular causes, while ozone was associated with the risk of death from respiratory causes.

Katanoda *et al*. (2011) performed an association between long-term exposure of fine particulate matter, suspended particulate matter, sulfur dioxide, and nitrogen dioxide with mortality from lung cancer and respiratory diseases in Japan. The study comprised 63,520 participants living in six areas in three Japanese cities that were enrolled between 1983 and 1985. The RR (95% CI) found for lung cancer, associated with an increase of 10 µg m^{-3} of $PM_{2.5}$ and PM_{10} were 1.24 (1.12-1.37), and 1.16 (1.08-1.25), respectively, after adjusting for confounding factors, such as smoking. Respiratory diseases, particularly pneumonia, were also significantly associated with air pollutants, including SO_2 and NO_2.

The cohort study by Hales *et al*. (2012) was carried out in New Zealand and evaluated the association of PM_{10} with mortality from several causes. For all causes and ethnicities, and all-natural causes, excluding accidental deaths and injuries, the RR (95% CI) found were equal to 1.07 (1.03-1.10). For lung cancer, 1.15 (1.04-1.26), respiratory diseases, 1.13 (1.05-1.21), and cardiovascular diseases, 1.06 (1.01-1.11), all investigated causes considering an increase of 10 μg m^{-3} of PM_{10}.

The study by Crouse *et al*. (2012) was the first national-level cohort study in Canada that aimed to investigate the risk of cardiovascular and non-accidental mortality associated with long-term exposure to fine particles. Exposure estimates for environmental $PM_{2.5}$ from 1991 to 2001 were derived from satellite observations for a cohort of 2.1 million Canadian adults who, in 1991, were among the 20% of the population mandated to provide detailed census data. The RR (95% CI) calculated were 1.15 (1.13-1.16) for non-accidental causes and 1.31 (1.27-1.35) for ischemic heart disease, for each 10-μg m^{-3} increase in $PM_{2.5}$ concentration.

Crouse *et al*. (2015) evaluated the exposure of $PM_{2.5}$ for 16 years in a national cohort of about 2.5 million Canadians, associating with non-accidental mortality and for specific causes in single pollution models and considering the interaction between pollutants. Exposure to $PM_{2.5}$ alone was not enough to fully explain the risk of mortality associated with exposure to environmental pollution, showing, therefore, a synergy between air pollutants. The RR estimates of two and three pollutant models were higher than that of single pollutants. For example, for non-accidental causes, the RR (95% CI) for $PM_{2.5}$ in the single model was 1.035 (1.029-1.041), while modeling $PM_{2.5}$ + O_3, $PM_{2.5}$ + NO_2 and $PM_{2.5}$ + O_3 + NO_2 were 1.038 (1.032-1.044), 1.070 (1.062-1.078) and 1.075 (1.067-1.084), respectively, associated with an increase of 5 μg m^{-3}, 9.5 ppb and 8.1 ppb in $PM_{2.5}$, O_3 and NO_2 concentrations, respectively.

Cesaroni *et al*. (2013) investigated long-term exposure to urban air pollution and mortality in a cohort of more than one million adults in Rome, Italy. The population was registered based on the 2001 Italian census and was followed for nine years. Residential exposures included annual NO_2 (from a regression model based on land use) and annual $PM_{2.5}$ (from a Eulerian dispersion model), as well as the distance to roads with more than 10,000 vehicles per day and intensity of traffic. Long-term exposures to both NO_2 and $PM_{2.5}$ were associated with an increase in non-accidental mortality, with an RR (95% CI) of 1.03 (1.02-1.03) for NO_2 and 1.04 (1.03-1.05) for $PM_{2.5}$, both for each increase of 10 μg m^{-3}. The robust association for 10 μg m^{-3} of $PM_{2.5}$ was found for ischemic heart disease, RR (95% CI) equal to 1.10 (1.06-1.13), followed by cardiovascular disease, 1.06 (1.04-1.08), and lung cancer, 1.05 (1.01-1.10).

The study by Carey *et al*. (2013) was an English cohort, with a total of 835,604 participants aged between 40 and 89 years old. The concentrations of PM_{10}, $PM_{2.5}$, O_3, NO_2, and SO_2 were estimated using dispersion models. The RR (95% CI) found for all causes were 1.07 (0.99-1.16) for PM_{10}, 1.13 (1.00-1.27) for $PM_{2.5}$, 1.20 (1.12-1.28) for SO_2, 1.02 (1.00-1.05) for NO_2, and 0.86 (0.78-0.94) for O_3, associated with an increase of 3.0 µg m^{-3}, 1.9 µg m^{-3}, 2.2 µg m^{-3}, 10.7 µg m^{-3} and 3.0 µg m^{-3}, respectively. The authors also provide estimates for circulatory, respiratory, and lung cancer, with the highest RR for respiratory causes. It can also be seen that the RR value for O_3 shows that there is no direct relationship between the increase in this pollutant and mortality from long exposure. O_3 is more related to mortality from short exposure (BELL *et al*., 2005).

The meta-analysis of 75 cohort studies linking $PM_{2.5}$ to the excess risk of mortality carried out by Pope III *et al*. (2019b) resulted in an average distribution of the effects of cohort studies from North America, Europe, and Asia, indicating robust associations of mortality from $PM_{2.5}$ with heterogeneity. The RR estimated by 10 µg m^{-3} of long-term exposure to $PM_{2.5}$ was 1.08 (1.06-1.11) for all-cause mortality, 1.11 (1.08-1.14) for cardiopulmonary mortality, and 1.13 (1.07-1.20) for lung cancer mortality.

The evidence from an increasing number of epidemiological studies has historically supported critical environmental policy decisions, providing supporting empirical evidence to the establishment of air quality standards. Fann *et al.* (2011) discuss how the results of epidemiological studies can be adequately characterized and applied correctly in risk assessments. Many of the uncertainties inherent in risk assessments are influenced by the methodological choices of the epidemiological study. In this choice, it is necessary to analyze the representativeness of the demographic profile and exposure to air pollution (modifying effects), avoid double counting of impacts, assess the consideration of confounding factors, verify the use of the International Classification of Diseases (ICD), among others.

Among the studies that used BenMAP-CE to evaluate an improvement in air quality associated with a reduction in mortality, Berman *et al*. (2012), who assessed the health benefits of ozone depletion in the United States, is an example. The annual number of avoidable premature deaths related to ozone ranged from 1,410 to 2,480 when the maximum concentrations for the control scenario were 75 ppb. From 2,450 to 4,130 avoidable deaths to 70 ppb, and 5,210 to 7,990 avoidable deaths to 60 ppb. The variability in death estimates results from the use of different epidemiological studies that served as a basis for the analysis.

Punger and West (2013) evaluated the effect that CMAQ grid resolution has on the estimate of avoidable deaths due to $PM_{2.5}$ and ozone in the United States, using BenMAP v. 4.0.44. The concentrations modeled for the finer resolution (12 km), resulted in an estimate of 66,000 deaths from all causes and 21,400 deaths from respiratory diseases per year, attributed to concentrations above the lower concentration thresholds of $PM_{2.5}$ and O_3, respectively. The modeled concentrations of 12 km were scaled to smaller resolutions by simple average projection, evaluating mortality in multiple resolutions from 24 to 408 km. For resolutions less than 100 km, the results obtained with total concentrations of $PM_{2.5}$ were less than 20%, compared to the estimate of the fine resolution, 20-30% less than 100-250 km of resolution, and 30-40% less in resolutions greater than 250 km. By increasing the grid cells size, the estimate of the total national respiratory mortality attributable to ozone was minimally affected, not increasing by more than 6% compared to the estimates for 12 km.

Nowak *et al*. (2013) modeled the removal of $PM_{2.5}$ by trees in ten US cities and estimated the health-related effects using BenMAP. The total amount of $PM_{2.5}$ removed annually by trees ranged from 4.7 tones in Syracuse (New York state) to 64.5 tones in Atlanta (Georgia state). The average improvement in air quality ranged from 0.05% in San Francisco (California) to 0.24% in Atlanta. Mortality reductions were around one person per year per city, but in New York City, there was a reduction in mortality of almost eight people per year. In this same sense, Gopalakrishnan *et al*. (2018) quantified the air pollution removal capacity of grasslands and scrublands in the United States and estimated the human health benefits associated with pollution removal. A total of 6.42 million tons of air pollutants retained by grasslands and undergrowth annually resulted in a health-related monetary benefit of $ 268 million.

Boldo *et al*. (2014) evaluated the association between fine particles and mortality in Spain for 2014 compared to 2007 levels. Taking into account the 2007 population data, between 8 and 15 deaths from all causes per 100 thousand inhabitants could be annual avoided by the reduction expected levels of fine particles, 10 to 30 for non-accidental causes, 1 to 5 for lung cancer and 2 to 6 for ischemic heart disease. Variability is also due to the choice of epidemiological studies (more than one study per cause).

Ding *et al*. (2016), with integrated modeling and monitored data, evaluated the reduction in the number of deaths during the 2010 Guangzhou Asian Games when the local government carried out a series of emission control measures that significantly improved air quality. The results showed that the average concentration of $PM_{2.5}$ in November 2010 decreased by 3.5 μg m^{-3} compared to the same period in 2009, due to the emission control measures, which would prevent 106 premature deaths, 1,869 cases of hospitalization and more than 20 thousand cases of outpatient visits, estimates for all causes of diseases.

Jiang and Yoo (2018) evaluated the spatial resolutions of the CMAQ model on health impact assessment over New York State for the year 2011. At 4 km and 12 km resolutions, $PM_{2.5}$ reproduced measured values with the fractional error of 54.41% for 4 km and 52.28% for 12 km, within recommend performance criteria. In a control scenario considering 0 μg m^{-3}, the annual mortality of all-cause deaths based on the 4-km CMAQ simulation was estimated as 6,187 (95%CI, 4,145-8,253). In contrast with the 12-km CMAQ simulation, the mortality estimates associated with $PM_{2.5}$ were 7,133 (4,778-9,519), 15.3% higher.

Howard *et al*. (2019) showed the benefits in terms of health effects in the adoption of emission control strategies in power plants for the Brazilian Northeast. With the reduction of PM_{10} emissions from 28.15 g/kWh to 0.69 g/kWh, about 168 premature deaths and 16,257 hospitalizations could be avoided annually.

Fernandes *et al*. (2020) estimated avoidable hospital admissions for respiratory system diseases in the four capitals of the Brazilian Southeast (São Paulo, Rio de Janeiro, Belo Horizonte, and Vitória) by meeting the final standards of CONAMA Resolution 491/2018 of all regulated pollutants. In three years, a total of 4,148 preventable hospitalizations were associated with high-level pollutant concentrations.

The literature review has showed the importance of improving air quality around the World. Air quality evaluation provides the policy framework for air quality management and assessment. Regarding the urban air quality management, reduce air pollution is essential to protect human health and the environment in a city.

3. EXCESS DEATHS ASSOCIATED WITH FINE PARTICULATE MATTER IN BRAZILIAN CITIES

3.1 Introduction

Urban populations around the world have increased from 46.5% in the year 2000 to 54.3% in 2016, while in Brazil, the urban population reached 85.9% in 2016 (UN, 2015; THE WORD BANK, 2018). It is associated with the intensification of urbanization processes resulting in the consumption of fossil fuel, deforestation, burning, generation of waste and the degradation of air quality (MCMICHAEL, 2000). Consequently, air pollution has become a public health concern, even when its levels fall short of current legislation (CURTIS *et al*., 2006).

When determining the concentration of a pollutant in the atmosphere, the degree of exposure of the receptors (humans, animals, plants, materials) is measured as the result of the release of this pollutant into the atmosphere from its emission sources and their physical (dispersion) and chemical (chemical reactions) interactions (SEINFELD and PANDIS, 2006). Thus, air quality is the product of the interaction between factors such as emissions, topography and weather conditions.

The World Health Organization (WHO) reported air pollution as the biggest health risk, causing approximately 6.5 million excess deaths globally in 2012, which is 11.6% of all deaths (WHO, 2016b). Among the main causes are cardiovascular diseases, stroke, chronic obstructive pulmonary disease, and lung cancer, in addition to the increased risks of acute respiratory infections (WHO, 2016b). Cohen *et al*. (2017) reported that fine particulate matter less than 2.5 μm ($PM_{2.5}$) was the fifth largest risk factor for mortality in 2015, averaging 4.2 million deaths globally (7.6% of all deaths), an increase of 20% concerning the total deaths in 1990.

In Brazil, Miranda *et al*. (2012) estimated the number of deaths associated with the excess exposure to $PM_{2.5}$ for June 2007 to August 2008, based on experimental campaigns in six Brazilian state capitals for adults over 45 years old. São Paulo presented the worst results with 9,700 premature deaths due to long-term exposure. Rio de Janeiro, Belo Horizonte, Porto Alegre, and Curitiba added other 3,900 deaths that could be avoidable if the annual $PM_{2.5}$ concentrations were reduced to the WHO guideline (10 μg m^{-3}).

An extensive body of epidemiological research has established a strong association between chronic exposures to $PM_{2.5}$ and ischemic heart disease (IHD), cardiovascular, lung cancer, all causes and all non-accidental causes mortality (POPE III *et al*., 2002; POPE III *et al.*, 2004; LADEN *et al*., 2006; KREWSKI *et al*., 2009; CROUSE *et al*., 2012; CESARONI *et al*., 2013; BENTAYEB *et al.*, 2015). The interaction between the sources of pollution and the atmosphere defines the level of air quality, which in turn determines the occurrence of adverse effects of air pollution on its receptors. In the Metropolitan Area of São Paulo, for example, Martins *et al*. (2017) showed that the probability of higher concentrations for CO, NO, NO_2, PM_{10}, and $PM_{2.5}$ were more frequent during the winter, while O_3 episodes occur most frequently during summer. Air quality monitoring aims to provide data to trigger emergency actions during periods of atmospheric stagnation, assess air quality in the light of established limits to protect the health and well-being of people, enable a correct planning of the territory, and monitor trends and changes in air quality due to changes in pollutant emissions.

Exposure to air pollutants is a risk factor for humans and many existing studies that attempt to assess the relationship between air pollution and mortality use pollutant concentration data from air quality monitoring stations (POPE III *et al.*, 2002; POPE *et al*., 2004; LADEN, *et al*., 2006; WONG *et al.*, 2008; KATANODA *et al*., 2011; LEPEULE *et al.*, 2012; HUANG *et al.*, 2012; THURSTON *et al.*, 2016). In Brazil, air quality monitoring is still restricted and unsatisfactory in terms of sample history, territorial coverage, number of monitored parameters and representatively in measurements, due to management difficulties and the low number of technicians involved, as well as lack of resources for the purchase and maintenance of equipment and monitoring networks (BRAZIL, 2014). In addition, the fine particulate matter is not yet nationally legislated.

Until 2017, there were 24 Brazilian cities with $PM_{2.5}$ monitoring. All these cities were in the southeastern region of Brazil. With measurements beginning in the year 2000 in São Paulo city, the concern with this pollutant is increasing, and an annual increase in the number of $PM_{2.5}$ monitoring stations is noticed. For the first time, this study performs an assessment of the number of total avoidable deaths attributable to a reduction in $PM_{2.5}$ concentrations, considering the annual guideline established by the WHO (10 µg m^{-3}) for all 24 Brazilian cities during 2000-2017 years with the available monitoring data. These results may be valuable to consider effective strategies to expanding air quality monitoring in Brazil, to improve air quality and for the adoption of a national standard for $PM_{2.5}$, allowing policymakers to project the population health improvements.

3.2 Materials and methods

3.2.1 Health Effects

The US EPA’s Environmental Benefits Mapping and Analysis Program (Community Edition; BenMAP-CE; v.1.3) (SACKS *et al.*, 2018) is used to facilitate the analyses of health effects. The inputs included a shapefile, the incident rates of the cause evaluated, population data, a health impact function, baseline and control scenarios of the pollutant evaluated. After acquiring the required data, the health effects were estimated using the Equation 2.1 (page 35).

As discussed in Section 3.4, there is no cohort study in Brazil relating to $PM_{2.5}$ mortality. Therefore, the number of deaths was estimated using concentration-response functions based on most cited/used studies of long-term exposure to $PM_{2.5}$ conducted on large cohorts in Europe (CESARONI *et al*., 2013) and North America (POPE III *et al.*, 2002; POPE III *et al.*, 2004; LADEN *et al.*, 2006; KREWSKI *et al.*, 2009; CROUSE *et al.*, 2012), as summarised in Table 3.1. The concentration-response functions that were not already included in BenMAP-CE were added based on β values and their standard errors. Fann and Risley (2013) reported that there are differences between American Cancer Society study (POPE III *et al*., 2002, POPE III *et al*., 2004; KREWSKI *et al*., 2009) and the Harvard Six-Cities Study (LADEN *et al.*, 2006) such as population size, geographic area covered, education level and $PM_{2.5}$ composition. The same may be applied for the studies conducted by Crouse *et al.* (2012) and Cesaroni *et al*. (2013). In order to generate a more comprehensive mortality estimate, it was used different exposure-response functions for each cause assessed, but the results must be interpreted by each function individually due to the differences among the methodology used in each cohort study.

Table 3.1 - Summary of the main features of selected concentration-response functions.

Health outcome	Reference	Age Range	Hazard ratio (95% CI)	β values (STD)
All Causes	Pope *et al*. (2002)	30-99	1.06 (1.02-1.11)	0.005827 (0.002157)
	Krewski *et al*. (2009)	30-99	1.03 (1.01-1.05)	0.002956 (0.000991)
	Laden *et al*. (2006)	25-74	1.16 (1.07-1.26)	0.014842 (0.004170)
Non-Accidental	Cesaroni *et al*. (2013)	>30	1.04 (1.03-1.05)	0.003922 (0.000491)
	Crouse *et al*. (2012)	>25	1.15 (1.13-1.16)	0.013976 (0.000668)
Cardiovascular	Cesaroni *et al*. (2013)	>30	1.06 (1.04-1.08)	0.005827 (0.000963)
	Crouse *et al*. (2012)	>25	1.16 (1.13-1.18)	0.014843 (0.001104)
	Laden *et al*. (2006)	25-74	1.28 (1.13-1.44)	0.024686 (0.006184)
Ischemic Heart Disease	Pope *et al*. (2004)	30 -99	1.18 (1.14-1.23)	0.016551 (0.001938)
	Krewski *et al*. (2009)	30-99	1.15 (1.11-1.20)	0.013976 (0.001989)
	Crouse *et al*. (2012)	>25	1.31 (1.27-1.35)	0.027003 (0.001558)
	Cesaroni *et al*. (2013)	>30	1.10 (1.06-1.13)	0.009531 (0.001631)
Lung Cancer	Pope *et al*. (2002)	30-99	1.14 (1.04-1.23)	0.013103 (0.004280)
	Krewski *et al*. (2009)	30-99	1.11 (1.04-1.18)	0.010436 (0.003222)
	Cesaroni *et al*. (2013)	>30	1.05 (1.01-1.10)	0.004879 (0.002177)

3.2.2 $PM_{2.5}$ data

To estimate the health-related benefits, baseline scenarios were defined considering the annual $PM_{2.5}$ concentrations for all Brazilian cities with representative monitoring data. Figure 3.1 shows the locations of the monitoring sites of $PM_{2.5}$ in 2017 (manual and automatic).

São Paulo was the first city that started monitoring $PM_{2.5}$ in Brazil in 2000 with manual measurements (KUMAR *et al*., 2016; PACHECO *et al*., 2017; ANDRADE *et al*., 2017). After 2001, a single measurement site was expanded to nine sites in São Paulo by 2017, with automatic stations operating since 2005. *Rio de Janeiro* city started the $PM_{2.5}$ monitoring in the middle of 2010 at eight monitoring sites, therefore, a representative annual concentration was available just in 2011. *Belo Horizonte*, capital of Minas Gerais, started monitoring $PM_{2.5}$ in 2013 at a single site in the north of the city. Since this monitoring site is located far from the urban center, the values obtained may be underestimated to represent the entire city. Therefore, the avoidable death values may be higher than those that will be presented. *Vitória* was the fourth capital of a state in Brazil to monitor $PM_{2.5}$. The measurements started in 2015 at a single site. All cities and years with $PM_{2.5}$ concentration values are available in the Appendix, Table S1. For the cities with more than one monitoring station, an average was performed to obtain a single value to represent the city.

In São Paulo state, the automatic stations use Beta radiation method to measure $PM_{2.5}$, while manual stations use gravimetric methods (virtual impaction – dichotomous; or impaction and cyclone), performed for 24 hours every six days (CETESB, 2017). In Rio de Janeiro state, the $PM_{2.5}$ measurements occur with a frequency of six days with a sample of 24 hours. The samples of particulate material are collected in Large Volume Samplers and then analyzed in laboratories by the State Environmental Institute of Rio de Janeiro (INEA, 2016a). In the state of Espírito Santo, the Tapered Element Oscillating Microbalance measurement methodology is used for the continuous measurement of the mass concentration of fine particulate material contained in ambient air (IEMA, 2017). In Minas Gerais, an automatic station monitors the $PM_{2.5}$ concentration in Belo Horizonte using a monitor with Beta radiation method (FEAM, 2016). For criteria of the temporal representativeness of data for the manual stations, half of the daily averages valid for the four-month periods January-April, May-August and September-December were considered, which are the criteria used by São Paulo State Environmental Protection Agency (CETESB).

The control scenario was evaluated considering the maximum annual concentration for $PM_{2.5}$ of 10 µg m^{-3}. This is the lowest level at which total, cardiopulmonary and lung cancer mortality have been shown to increase with more than 95% confidence in response to long-term exposure to $PM_{2.5}$ (WHO, 2006). Therefore, the benefits will only be evaluated if the baseline scenario concentrations are higher than the control scenario.

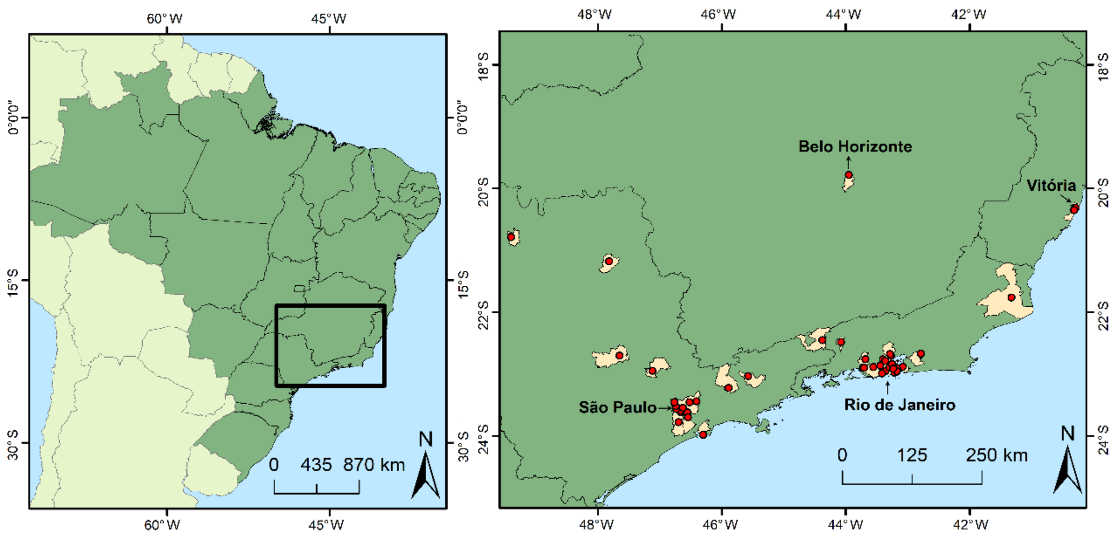

Figure 3.1 - Locations of the monitoring sites of $PM_{2.5}$ in Brazil (2017).

3.2.3 Population and mortality data

Annually population data were obtained from *Departamento de Informática do Sistema Único de Saúde* (DATASUS) for each city by age group from 2000 to 2015. Population data from 2000 to 2013 are preliminary estimates made in a study sponsored by *Rede Interagencial de Informações para a Saúde* (Ripsa), while this data from 2013 to 2015 are preliminary estimates prepared by *Coordenação-Geral de Informações e Análises Epidemiológicas* (CGIAE) of the Secretariat of Health Surveillance (*Secretaria de Vigilância em Saúde* - SVS), Ministry of Health (*Ministério da Saúde* - MS). To estimate the health effects for 2016 and 2017, population data of 2015 was used.

Annually mortality data by age group due all causes (ICD-10: A00-Y98), all non-accidental causes (ICD-10: A00-R99), ischemic heart disease (ICD-10: I20-I25), cardiovascular (ICD-10: I20–I28, I30–I52, I60–I79) and lung cancer (ICD-10: C33-C34) was obtained from DATASUS, which regulates mortality data in the Sistema de Informação sobre Mortalidade (SIM). To estimate the health effects for 2017, the incident rate of 2016 was used.

3.3 Results

3.3.1 Overview of annual $PM_{2.5}$ concentrations

About 89% of the total annual $PM_{2.5}$ concentration analyzed were higher than WHO guideline (10 μg m^{-3}). As for individual cities, Figure 3.2 shows the annual average concentrations of $PM_{2.5}$ in São Paulo city over the period of between 2000 and 2017, showing the annual concentration always above the WHO guideline. There were some periods with a decrease in concentration to 14.6 μg m^{-3} in 2009, followed by periods with increased concentrations to 20.2 μg m^{-3} in 2011. In 2011, there was a decrease in rainfall, with periods of drought and low humidity, probably as a consequence of the planetary scale phenomenon known as La Niña. The winter in 2011, like the previous year, was among the most unfavorable to the dispersion of the pollutants (CETESB, 2012). Plainly, there is no clear trend in $PM_{2.5}$ concentrations over the years, but there has been a steady reduction in concentration values over the most recent years from 19.1 μg m^{-3} in 2014 to 16.1 μg m^{-3} in 2017, mainly because of the reduction in the number of days unfavorable to the dispersion of pollutants during the winter period (CETESB, 2017). Other cities of the São Paulo state, such as São Caetano do Sul and Guarulhos, obtaining annual concentrations higher than São Paulo (18 µg m^{-3} in both cities in 2017). All the concentration values for the 11 cities of São Paulo state are available in SI Table S1 for the years with monitoring data (2000 to 2017).

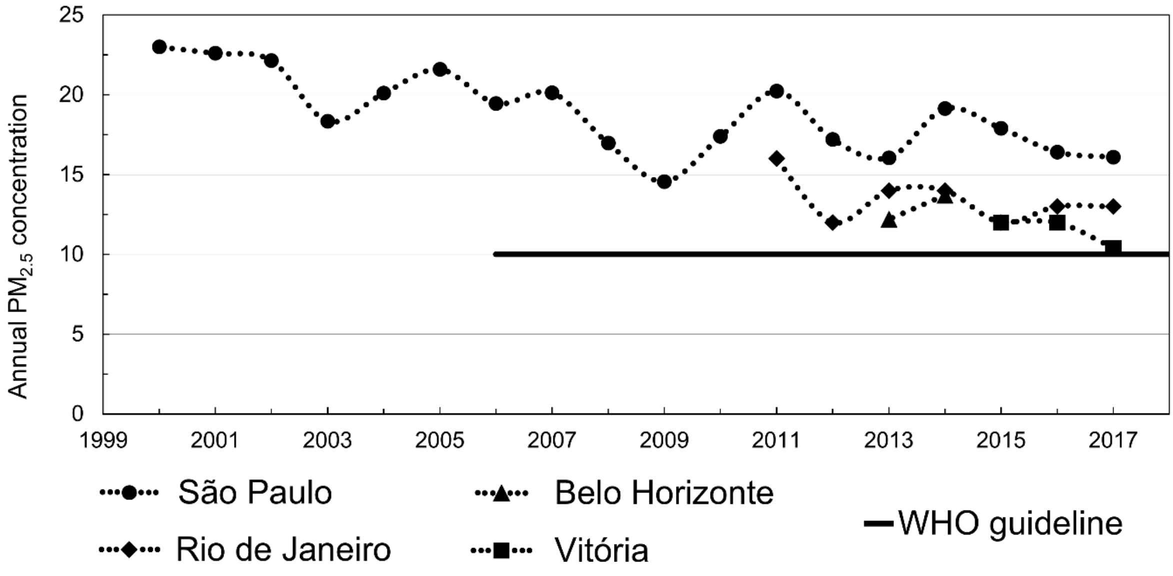

Figure 3.2 - Annual $PM_{2.5}$ concentration for São Paulo, Rio de Janeiro, Belo Horizonte and Vitória over the years.

Figure 3.2 also shows average annual $PM_{2.5}$ concentrations for Rio de Janeiro. In the seven evaluated years, $PM_{2.5}$ levels decreased and increased, with no single tendency, with all concentration values being higher than that recommended by the WHO. For 2016 and 2017, the annual concentration was 13 µg m^{-3}. According to Godoy *et al*. (2018), in Rio de Janeiro, the vehicular contribution to $PM_{2.5}$ ranged from 48 to 70%, with a mean value of 59±9%, during the period from June 2012 to June 2013. Another study carried out from 2003 to 2005 showed that sources related to anthropogenic sources as vehicle traffic and oil combustion, represented about 65% of the $PM_{2.5}$ fraction in Rio de Janeiro (Godoy *et al*., 2009). The other cities of Rio de Janeiro state obtained annual $PM_{2.5}$ concentration values between 8 and 22 µg m^{-3} (Appendix Table S1).

In 2013, the annual $PM_{2.5}$ concentration in Belo Horizonte was 12.2 µg m^{-3} as opposed to 13.7 µg m^{-3} in 2014. These are values below than those reported by Miranda *et al*. (2012) for June 2007 to August 2008 (14.7 µg m^{-3}). Recent monitoring data was not available.

The annual $PM_{2.5}$ concentration in Vitória over 2015-2016 was 12 µg m^{-3}, slightly above the WHO guidelines. In 2017, the annual concentration dropped down a bit more to 10.4 µg m^{-3}. Vila Velha is the other city in Espírito Santo state with a monitoring station. The annual levels for these two first years were 11.4 and 11 µg m^{-3}, while in 2017 the concentration dropped to 9.7 µg m^{-3}, lower than the WHO guidelines.

In 2014, 92% of the world population was living in places where WHO air quality guideline standards were not met (WHO, 2017). Figure 3.3 shows a comparison of annual $PM_{2.5}$ concentration in 2014 among São Paulo, Rio de Janeiro, Belo Horizonte and other cities around the world. In South America, Bogotá (Colombia) and Santiago (Chile) presented annual concentrations higher than those observed in Brazilian cities, exceeding 20 µg m^{-3}. In the Central Valley of Chile, during most of the year, there is a thermal inversion layer, which favors the accumulation of pollution (VALDÉS *et al*., 2012). On the other hand, Montevideo (Uruguay), a coastal city, present a concentration lower than WHO guideline, as Sydney in Australia. In Shanghai and Beijing (China), the average annual concentration of $PM_{2.5}$ was 52 µg m^{-3} and 85 µg m^{-3}, respectively, mainly due to motor vehicles emissions (CHAN and YAO, 2008; LIU *et al*., 2014). Paris (France) and Singapore presented values similar to São Paulo, with an annual $PM_{2.5}$ concentration of 18 µg m^{-3} (WHO, 2016c).

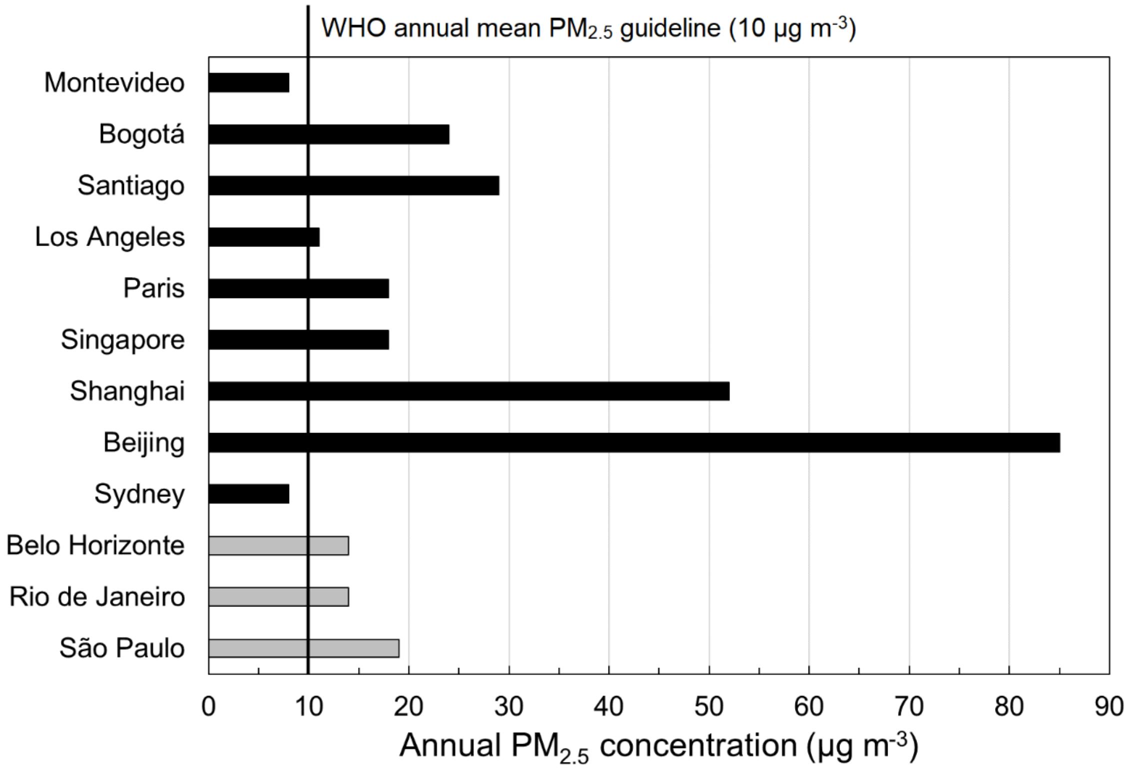

Figure 3.3 - Annual $PM_{2.5}$ concentration in 2014 from different cities around the world (WHO, 2016c). The cities covered in our assessment are presented in grey color.

3.3.2 Population and mortality overview

São Paulo is the city with the largest population in Brazil (7.7 million of inhabitants over 25 years in 2015). From 2000, it was observed an increase in the first ten years for all age groups. After 2010, just the age group 25 to 29 years old presented a decline, while the other groups still growing up in population. In total numbers, the population increases 30% from 2000 to 2015, reaching 7.7 million of inhabitants in 2015. When the total number of deaths is observed, the age groups until 49 years presented a decrease of 34.4% from 2000 to 2016, while the deaths for the group formed with people with more than 80 years went up 90.5%. However, the population of this group more than duplicated in this period and, therefore, the incident rate (deaths/population) presented a decrease of 19%. Figure 3.4 shows the incident rate for all causes over the years. For the younger groups (<44 years), the incidence rate is lower than 0.005.

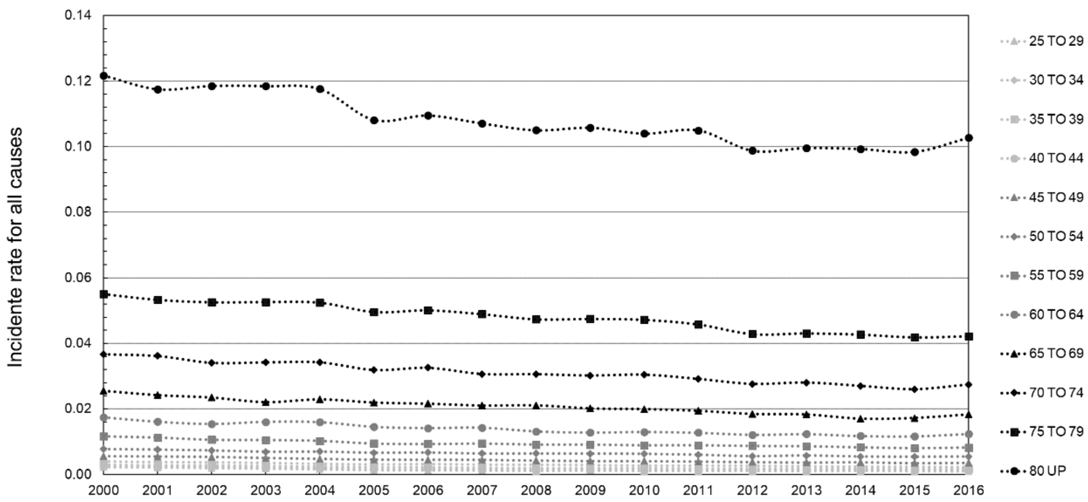

Figure 3.4 - Incident rate for São Paulo city (over 25 years) by age group over the years for all causes.

Figure 3.5 shows the number of deaths for the population (over 25 years) of São Paulo city by all causes, all non-accidental causes, IHD, cardiovascular and lung cancer. In absolute number, there was an increase of deaths for all five causes (22% for all causes, 29% for non-accidental, 12% for cardiovascular, 5% for IHD and 34% for lung cancer, between 2000 and 2016). However, when the incident rate is evaluated, it was observed that just for lung cancer this increase remains the same (i.e., 2.9% of increase from 2000 to 2016). The other causes obtained reductions in incident rate: 6.3% for all causes; 0.8% for non-accidental; 14.2% for cardiovascular and 19.6% for IHD, for the same period.

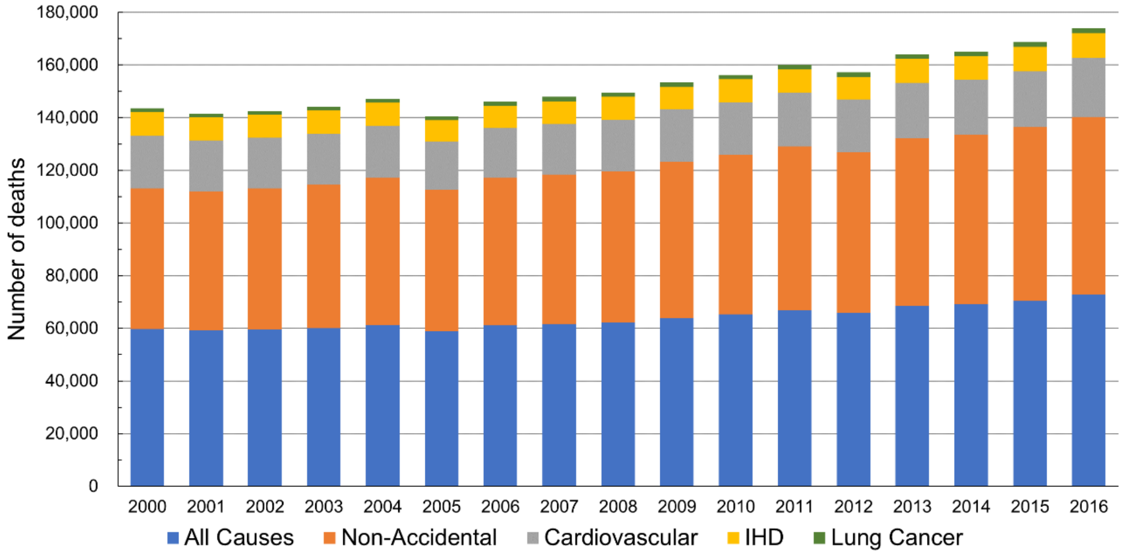

Figure 3.5 - Total number of deaths for the population (over 25 years) of São Paulo.

Rio de Janeiro city presented an increase of 3.9% in the number of inhabitants (over 25 years) from 2011 to 2015, reaching approximately 4.2 million. Although some age groups such as 25 to 29 years and 45 to 49 years showed a decrease of 7.5% and 5%, respectively, over the years. Concerning mortality, the age group with more than 80 years old presented the highest number of deaths, for all causes evaluated. The incident rate increases when the populations get older. There is, for the most of age groups, a decline in incident rate for all five causes over the years evaluated. An exception is, for example, an increase of 8% in the incidence rate for groups up to 44 years for cardiovascular disease. Other increases also found were for the groups of 25 to 29 and 35 to 39 years for non-accidental causes, 24% and 4%, respectively. Compared to São Paulo, Rio de Janeiro had an incident rate higher for all causes and non-accidental causes in all age groups. For cardiovascular diseases, lower values were found. For the other causes, there were higher and lower values depending on the age group.

Belo Horizonte presented a population with more than 25 years of approximately 1.7 million inhabitants in 2015. The incident rates evaluated were lower than those for São Paulo and Rio de Janeiro for all five causes and age groups.

The population over 25 years of Vitória was more than 233 thousand inhabitants in 2015. Compared to Rio de Janeiro, Vitória presented lower values of the incident rate in 2015 for all causes and non-accident causes, for all age groups. Some higher values were found for IHD and lung cancer, especially for the smallest age groups, as for example, for the age group 30 to 34 years, which presented values 40% and 80% higher than Rio de Janeiro for IHD and lung cancer, respectively. Compared to Belo Horizonte, the values for IHD, which are higher, are highlighted. Compared to São Paulo, lower values of incident rate were found for all causes, non-accidental causes, cardiovascular diseases and IHD, for the majority of age groups. On the other hand, the incident rate for lung cancer was higher for most of the age groups.

In São Paulo state, other cities presented incident rate values higher than those found in São Paulo city. For example, in 2015, Guarulhos presented higher values for almost all age group in all the five causes evaluated. Campinas obtained higher values for all causes, non-accidental causes, and lung cancer. Similar situation for Taubaté, which also presented higher values for IHD. In Rio de Janeiro state, compared to the capital, practically all cities presented an incidence rate greater in 2015, with a highlight for Duque de Caxias that presented values superior to the other cities for most causes and age groups. In Espírito Santo, Vila Velha presented incident rates greater than Vitória for the most causes and age groups.

3.3.3 Avoided premature mortality against WHO guideline for annual mean concentrations of $PM_{2.5}$

Table 3.2 summarizes the avoided mortalities for all causes, non-accidental causes, cardiovascular, IHD and lung cancer for the city of São Paulo for some years. A detailed list of annual mortality estimates between 2000 and 2017 can be found in SI Tables S16 to S18. As expected, São Paulo presented the highest values of avoidable deaths among all cities studied. When considering the concentration-response function of Pope *et al*. (2002), the analysis indicated a number of avoidable deaths for all causes ranging from 1,660±620 (in 2009) to 4,280±1,630 (in 2000). Depending on the selected relative risk, the maximum value can reach 7,100±2,140 avoidable deaths. This number represents 18% of the total deaths in 2000 (25 to 74 years old). The results obtained with Krewski *et al*. (2009) relative risk were the lower among all cause category. Adding the avoidable deaths from 2000 to 2017, between 28,880±9,770 and 82,720±24,550 people would not have prematurely died due to $PM_{2.5}$ in São Paulo, depending on the cohort study.

Table 3.2 - Estimate of avoidable deaths for the city of São Paulo, with the standard deviation in parentheses.

Health outcome	**Exposure-response functions**	**2000**	**2009**	**2015**	**2017**
All Causes	Pope III *et al*. (2002)	4,280 (1,630)	1,660 (620)	3,120 (1,170)	2,510 (940)
	Krewski *et al.* (2009)	2,220 (750)	850 (290)	1,600 (540)	1,280 (430)
	Laden *et al.* (2006)	7,100 (2,140)	2,420 (700)	4,180 (1,220)	3,380 (980)
Non-Accidental	Cesaroni *et al.* (2013)	2,676 (342)	1,050 (130)	2,000 (250)	1,620 (200)
	Crouse *et al.* (2012)	9,056 (472)	3,690 (180)	6,890 (350)	5,630 (280)
Cardiovascular	Cesaroni *et al.* (2013)	1,480 (250)	520 (90)	950 (160)	780 (130)
	Crouse *et al.* (2012)	3,580 (290)	1,310 (100)	2,350 (190)	1,930 (150)
	Laden *et al.* (2006)	3,290 (930)	1,100 (290)	1,930 (520)	1,610 (430)
Ischemic Heart Disease	Pope III *et al*. (2004)	1,780 (230)	620 (80)	1,120 (140)	900 (110)
	Krewski *et al*. (2009)	1,530 (240)	520 (80)	950 (140)	770 (110)
	Crouse *et al*. (2012)	2,730 (186)	986 (60)	1,760 (110)	1,440 (90)
	Cesaroni *et al*. (2013)	1,070 (190)	360 (60)	660 (120)	530 (90)
Lung Cancer	Pope III *et al*. (2002)	220 (80)	100 (30)	190 (60)	140 (50)
	Krewski *et al*. (2009)	180 (60)	80 (30)	150 (50)	120 (40)
	Cesaroni *et al*. (2013)	90 (40)	40 (20)	70 (30)	60 (30)

The avoidable deaths for non-accidental cause with the relative risk of Crouse *et al.* (2012) in 2017 were 67% to 339% higher than those found for all-cause, while the relative risk of Cesaroni *et al*. (2013) presented lower values, except when compared with Krewski *et al.* (2009) results. For cardiovascular and IHD, the results obtained with Crouse *et al.* (2012) were very high compared to others (reaching 173% higher), while the relative risk pointed out by Cesaroni *et al*. (2013) results in the lower values.

Considering the relative risk of Pope III *et al.* (2002) for lung cancer, the avoidable deaths were more representative in relation to the total number of deaths by lung cancer for the first eight years (2000 to 2007). Until 2007, the average of representativeness was 13.3%, while from 2008 to 2017 it was 8.9%. Considering the entire period, this average reached 10.9%.

The year 2017 presented the highest number of monitoring sites for $PM_{2.5}$ in São Paulo state (20 stations in 11 cities). Table 3.3 shows the values of avoidable deaths in four of these cities for this year. The complete series of avoidable deaths are presented in Appendix, Tables S9 to S15. Guarulhos city, which is at the border of São Paulo city, showed the highest values of avoidable deaths among the other cities of São Paulo state (524±153, considering the concentration-response function of Laden *et al.* (2006) for all causes). Campinas, further north of São Paulo city, also presented high values (more than double of the average in these cities). It is worth pointing out that these two cities presented annual $PM_{2.5}$ concentration higher than São Paulo (Table S1), what contributed to the higher number of avoidable deaths estimated. São Bernardo do Campo and Santos also presented significant values of avoidable deaths.

Table 3.3 - Estimate of avoidable deaths for four cities of São Paulo state in 2017, with the standard deviation in parentheses.

Health outcome	**Exposure-response functions**	**Campinas**	**Guarulhos**	**São Bernardo do Campo**	**Santos**
All Causes	Pope III *et al.* (2002)	270 (100)	320 (20)	150 (60)	130 (50)
	Krewski *et al.* (2009)	140 (50)	160 (60)	80 (30)	70 (20)
	Laden et al. (2006)	350 (100)	520 (150)	220 (70)	140 (40)
Non-Accidental	Cesaroni *et al.* (2013)	170 (20)	200 (30)	90 (10)	80 (10)
	Crouse *et al.* (2012)	580 (30)	700 (40)	330 (20)	290 (10)
Cardiovascular	Cesaroni *et al.* (2013)	80 (10)	100 (20)	50 (10)	40 (10)
	Crouse *et al.* (2012)	190 (20)	260 (20)	120 (10)	100 (10)
	Laden *et al.* (2006)	150 (40)	270 (70)	110 (30)	60 (20)
Ischemic Heart Disease	Pope III *et al.* (2004)	100 (10)	130 (20)	50 (6)	50 (6)
	Krewski *et al.* (2009)	80 (10)	110 (20)	40 (6)	40 (6)
	Crouse *et al.* (2012)	150 (10)	200 (10)	80 (5)	80 (5)
	Cesaroni *et al.* (2013)	60 (10)	70 (10)	29 (5)	30 (5)
Lung Cancer	Pope III *et al.* (2002)	15 (5)	15 (6)	10 (3)	7 (2)
	Krewski *et al.* (2009)	10 (4)	15 (4)	8 (3)	6 (2)
	Cesaroni *et al.* (2013)	5 (2)	6 (3)	4 (2)	3 (1)

Table 3.4 presents the avoidable deaths for the city of Rio de Janeiro for some years. The complete series of avoidable deaths are presented in Appendix Table S6. The values followed the increase or reduction of the $PM_{2.5}$ concentration over the years (Figure 3.2). Considering Pope III *et al.* (2002) relative risks, the all-cause avoidable deaths represent 3.5% of all deaths in 2011 and lung cancer 7.7% of deaths by this cause. When the results with Krewski *et al.* (2009) relative risks is observed, it is noticed that IHD represents 45.5% of all causes avoidable deaths. The results of 2016 and 2017 were equal due to the same incident rate used and because the annual $PM_{2.5}$ concentration was equal in these two years. The other cities of Rio de Janeiro state with representative $PM_{2.5}$ monitoring presented a number of avoidable deaths of about ten times lower. All the values are in presented in Appendix, Tables S3 to S8.

Table 3.4 - Estimate of avoidable deaths for the city of Rio de Janeiro, with the standard deviation in parentheses.

Health outcome	Exposure-response functions	2011	2013	2015	2017
All Causes	Pope III *et al*. (2002)	1,730 (650)	1,140 (430)	600 (220)	900 (330)
	Krewski *et al*. (2009)	880 (300)	580 (200)	300 (100)	450 (150)
	Laden et al. (2006)	2,340 (680)	1,520 (430)	780 (220)	1,160 (330)
Non-Accidental	Cesaroni *et al.* (2013)	1,100 (140)	730 (90)	380 (50)	560 (70)
	Crouse *et al.* (2012)	3,820 (190)	2,560 (130)	1,340 (70)	2,000 (100)
Cardiovascular	Cesaroni *et al.* (2013)	440 (70)	300 (50)	160 (30)	230 (40)
	Crouse *et al.* (2012)	1,090 (80)	740 (60)	400 (30)	590 (50)
	Laden *et al.* (2006)	930 (250)	610 (160)	330 (90)	500 (130)
Ischemic Heart Disease	Pope III *et al.* (2004)	470 (60)	320 (40)	160 (20)	240 (30)
	Krewski *et al.* (2009)	400 (60)	270 (40)	140 (20)	210 (30)
	Crouse *et al.* (2012)	750 (50)	500 (30)	270 (20)	390 (20)
	Cesaroni *et al.* (2013)	280 (50)	180 (30)	100 (20)	140 (30)
Lung Cancer	Pope III *et al.* (2002)	90 (30)	65 (20)	30 (10)	50 (15)
	Krewski *et al.* (2009)	75 (25)	50 (15)	30 (10)	40 (10)
	Cesaroni *et al.* (2013)	35 (15)	25 (10)	15 (5)	20 (10)

Belo Horizonte presented all causes avoidable deaths between 90±30 and 250±70 in 2013 and between 150±50 and 410±120 in 2014, depending on the cohort study, an increase of 68% on average. Considering the relative risks of Cesaroni *et al*. (2013), the avoidable deaths for cardiovascular, IHD and lung cancer in 2014 represented 37%, 15% and 3% of the non-accidental deaths.

Vitória presented lower values of avoidable deaths. With an improvement of 2 µg m^{-3} on the $PM_{2.5}$ annual average in 2015 and 2016, the values for all five causes investigated were similar in both years. In 2017, due to the lower value of annual $PM_{2.5}$ concentration (10.4 µg m^{-3}), the benefits observed were lower than 10 avoidable deaths. Figure 3.6 shows the avoidable deaths per 100 thousand inhabitants for São Paulo, Rio de Janeiro, Belo Horizonte and Vitória, over the years, for all causes and lung cancer, according to the relative risk of Pope III *et al.* (2002). São Paulo obtained the higher values for all causes, followed by Rio de Janeiro and Belo Horizonte. Vitória presented low values for the three years with monitoring data, with the values for all causes reaching levels of those for lung cancer for the other cities in 2017, due to lower annual $PM_{2.5}$ concentration and lower population.

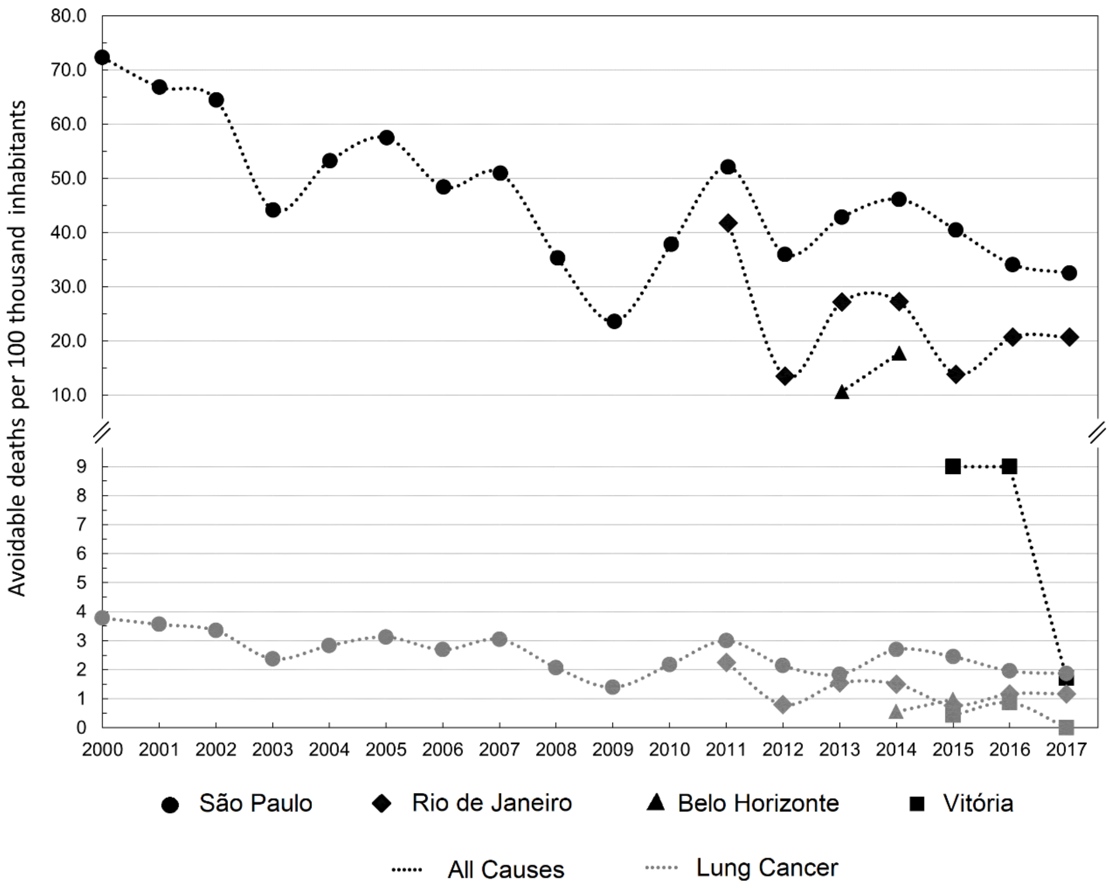

Figure 3.6 - Avoidable deaths per 100 thousand inhabitants for São Paulo, Rio de Janeiro, Belo Horizonte and Vitória, over the years, for all causes and lung cancer.

Figure 3.7 shows the avoidable deaths from 2014 to 2017 by non-accidental causes according to Cesaroni *et al*. (2013). It is noticed that the large urban centers obtained the higher values, mainly due the population size. In some cities, the estimative was not possible because the annual $PM_{2.5}$ concentration was not available, or it was not representative. For the cities with zero avoidable deaths, the $PM_{2.5}$ concentration was below the WHO recommendation.

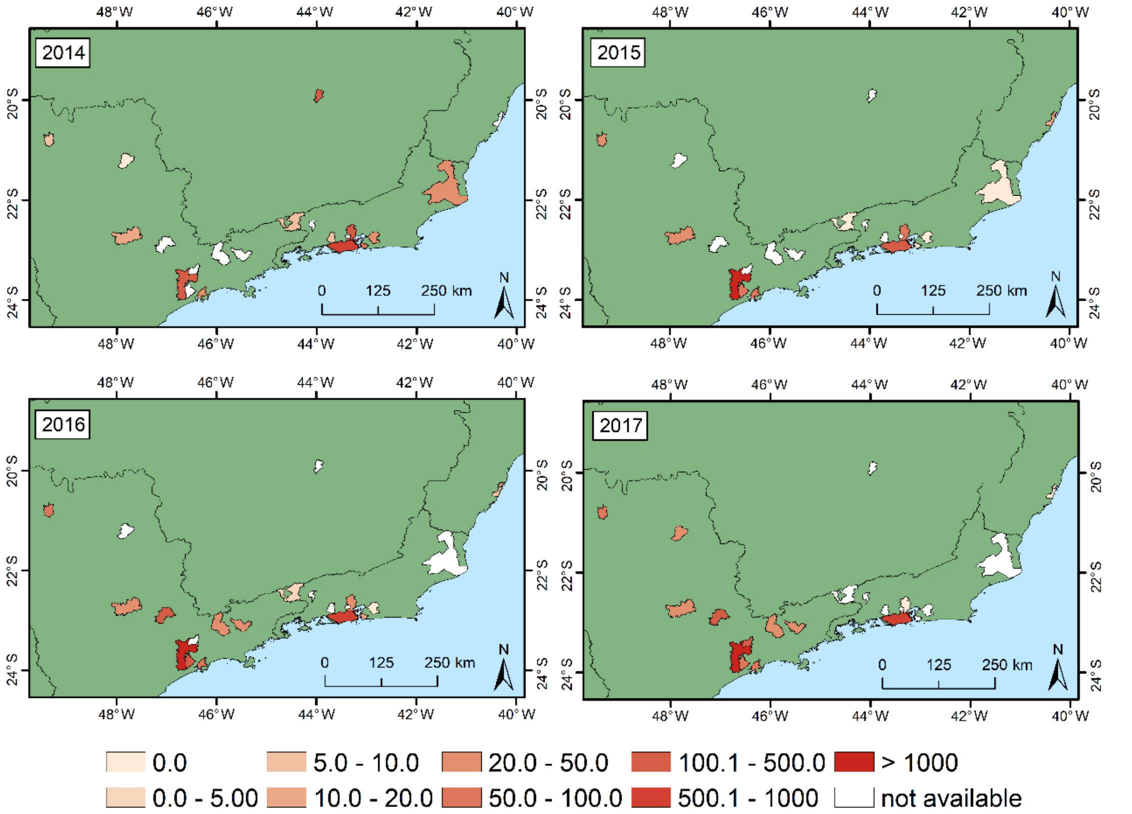

Figure 3.7 - Avoidable deaths for non-accidental causes using Cesaroni *et al*. (2013) relative risk.

3.4 Discussion

The accuracy of the estimated air pollution impact on health in a specific city, region or country depends on air pollution concentrations and exposure, population groups exposed, background incidence of mortality or morbidity, and concentration-response functions. The choice of which health outcomes to include in the assessment may be determined by the strength of available studies, the accessibility of health information, and the importance of the impact from a health and economic perspective (WHO, 2006). However, the decision on which epidemiological studies to use and how to apply them to evaluate the health impact assessment is left to the analyst.

Epidemiological studies allow estimating the effects of a pollutant on human health. The relationship between changes in short-term air pollution levels and changes in various indicators of population health or the health of individuals is studied in time series, panels, and case timeline studies (EFTIM and DOMINICI, 2005; WHO, 2006). These studies provide the basis to examine the short-term benefits of an improvement on air quality (LIN *et al*., 2016a; 2016b; CHEN *et al*., 2017; GOPALAKRISHNAN *et al*., 2018). The estimation of chronic health effects associated with air pollution is carried out through cohort studies, which examine the risk of a health outcome (e.g. death) in relation to long-term environmental exposure to pollution, generally comparing people living in different geographical locations (EFTIM and DOMINICI, 2005; WHO, 2006).

Cohort studies generally provide higher estimates of pollution effects than time-series studies, indicating that long-term exposures have a greater effect than short-term exposures (EFTIM and DOMINICI, 2005). The disadvantages in carrying out this type of study are logistical difficulties, high implementation costs, monitoring of study populations over long periods of time with great potential for losses, and a large number of individuals generally required to carry out the study. In addition, since exposure is generally considered as a city-wide average, different sites need to be assessed to ensure adequate variability of exposure (WHO, 2006).

Most of the cohort studies in the air pollution literature focused primarily on mortality and provided the most comprehensive estimates of the number of deaths attributable to exposure to pollution and the extent of the average reduction in life expectancy. Therefore, they were considered adequate for health impact assessment (KÜNZLI *et al*., 2001; COHEN *et al*., 2004, CHEN *et al*., 2008).

There are some time series studies in Brazil, mainly in São Paulo, relating air pollution and mortality (SALDIVA *et al*., 1994; SALDIVA *et al*., 1995; GOUVEIA and FLETCHER, 2000a; CONCEIÇÃO *et al*., 2001a; GOUVEIA *et al*., 2003; FREITAS *et al*., 2004; MARTINS *et al*., 2004; DAUMAS *et al*., 2004; BRAVO *et al*., 2016; GOUVEIA and JUNGER, 2018), but none of them evaluated the relationship between fine particulate matter and mortality. Therefore, we have estimated the number of avoidable deaths using cohort studies from the USA, Canada and Italy.

The choice of the cohort study that serves as the basis for estimate the health benefits may generate considerable differences. As reported by Boldo *et al*. (2014), the disparities among the cohort studies could be due to chemical composition of $PM_{2.5}$ and its heterogeneous mix of particle sizes, thus encompassing the environmental characteristics of each area of study (geographic location, emission sources and pollutants mixtures), the variability among different populations, social-economic conditions and the exposure assessment methodology. Although they are not studies that represent the local fine particulate matter and the Brazilian population, the cohort studies used in this work were also applied for other countries than those from which they were proposed (BALLESTER *et al*., 2008; HE *et al*., 2010; BOLDO *et al*., 2011; CHAE and PARK, 2011; NAWAHDA, 2013; PASCAL *et al*., 2013; BOLDO *et al*., 2014; ABE and MIRAGLIA, 2016). Voorhees *et al*. (2014) reported that it is a common practice to apply concentration-response functions that are not specific to a particular city or region, but which are recognized as being of high quality and produced from well-conducted epidemiological studies. In the extended follow-up of the Harvard Six Cities study (LADEN *et al*., 2006), the historical annual mean $PM_{2.5}$ concentration was 16.4 µg m^{-3} (range, 11.0 - 29.6 µg m^{-3}); for the ACS study (POPE III *et al*., 2002; POPE III *et al*., 2004) was 20 µg m^{-3} (9.0 - 33.5 µg m^{-3}); for the Rome study (CESARONI *et al*., 2013) was 43.6 µg m^{-3} (13.0 - 75.2 µg m^{-3}); and for the Canadian study was 8.7 µg m^{-3} (1.9 - 19.2 µg m^{-3}). The annual concentrations observed in the present study attend the range of these cohort studies. Furthermore, the 10th revision of the International Statistical Classification of Diseases and Related Health Problems were used according to the health outcome described by the cohort studies, as the population age interval. Therefore, the results presented provide a good picture for environmental authorities develop air quality policies.

The comparison between the results of avoidable deaths obtained in this work with other similar ones in the world should be done carefully, considering the differences related to the concentration-response function used, age groups and the $PM_{2.5}$ concentration difference between the baseline and control scenarios. Boldo *et al*., (2014) showed that an improvement of 4.7 µg m^{-3} in Madrid (Spain) between the years 2007 and 2014 resulted in a total of 30, 8 and 4 annual avoidable deaths per 100,000 inhabitants due to all causes, ischemic heart disease, and lung cancer, respectively. In New York City, 65 premature deaths due all causes per 100,000 inhabitants were estimated, based on the difference relative to nonanthropogenic, policy-relevant background concentrations, which represented approximately 5% of average $PM_{2.5}$ concentrations in New York City (KHEIRBEK *et al*., 2013). The improvements of air quality in Japan by reducing the emissions of $PM_{2.5}$ from 2006 to 2009 could save 28,400 lives (> 65 years) based on a reduction target of 10 µg m^{-3} annual mean concentration (NAWAHDA, 2013). In Shanghai, the estimated impact on all-causes mortality of a year exposure to an annual mean $PM_{2.5}$ concentration was 1,100 deaths from October 2010 to September 2011 and 180 deaths from October 2011 to September 2012 (VOORHEES *et al*., 2014).

In Brazil, the Resolution CONAMA 03/1990 defines the primary and secondary air quality standards[1]. It is nationally legislated the total suspended particles, smoke, inhalable particles (PM_{10}), sulfur dioxide (SO_2), carbon monoxide (CO), ozone (O_3) and nitrogen dioxide (NO_2). As may be noted, they are standards dating back to 1990 and today, because of the whole body of scientific studies related to air pollution and health, they can be considered outdated standards. Moreover, fine particles are not legislated[1]. Therefore, the states of São Paulo and Espírito Santo created they own air quality legislation, with more restrictive standards over time. They also included standards for fine particles, considering the WHO guideline of $PM_{2.5}$ as the final standard.

In São Paulo, although there was an increase in the number of vehicles and in the consumption of fuels, pollutant concentrations have decreased in the last ten years, except for ozone and $PM_{2.5}$ (CARVALHO *et al*., 2015; ANDRADE *et al*., 2017). Vehicular traffic, especially diesel-powered, is a major source of black carbon in urban areas (SANCHÉZ-CCOYLLO *et al*., 2009; WANG *et al*., 2011; ALVES *et al*., 2015). These emissions may come from the exhaust, physical wear of tires, brakes, and roads (PANT and HARRISON, 2013; ANDRADE *et al*., 2017). In São Paulo, Rio de Janeiro and Belo Horizonte, black carbon explained approximately 30% of the $PM_{2.5}$ mass (ANDRADE *et al*., 2012; ANDRADE *et al*., 2014). Black carbon is an important indicator to evaluate the adverse health effects for being one of the main components of the primary combustion particles (JANSSEN *et al*., 2011; WHO, 2012; LI *et al*., 2016). Some cohort studies have identified a positive relationship between mortality (all causes, natural causes, cardiopulmonary, respiratory, lung cancer) and long-term exposure to black carbon (FILLEUL *et al.* 2005; LIPFERT *et al*. 2006; BEELEN *et al*. 2008; SMITH *et al*. 2009). This shows the importance of adoption of control programs which aim to reduce $PM_{2.5}$ emission and concentration in the atmosphere in urban centers.

In this work, we investigated 24 Brazilian municipalities that monitor $PM_{2.5}$. These cities represent 16.7% of the total inhabitants above 25 years old (in 2015) in the country. In relation to the total number of mortalities across Brazil, these municipalities account for 17.5% of all-causes, 18.0% of non-accidental, 18.9% of cardiovascular, 20.0% of IHD, and 19.6% of lung cancer deaths, according DATASUS/SIM system. WHO (2018a)

[1] After the publication of the article, the CONAMA Resolution 03/1990 was updated to CONAMA Resolution 491/2018. See Section 2.1.

estimated 613 deaths per 100,000 inhabitants in Brazil in 2016 attributable to ambient air pollution. This work showed that 16 Brazilian cities faced annual $PM_{2.5}$ concentration above WHO guidelines in 2016 and there were about 28 avoidable deaths per 100,000 inhabitants in these cities.

Although the main centers normally obtain the highest PM concentrations (BOLDO *et al.*, 2014; CHEN *et al.*, 2017), in this study it was verified that cities around capitals presented higher annual $PM_{2.5}$ concentration, as for example, São Caetano do Sul and Guarulhos in São Paulo and Belfort Roxo and Duque de Caxias in Rio de Janeiro. This shows the importance of a monitoring that covers several areas, and, in terms of emission control strategies, the entire metropolitan region should be considered.

The increase in the elderly population observed has also consequences and implications for society and public health. The vulnerable population have higher incidence rate and therefore are the ones benefitting the most from an improved air quality. It is also important to have an air quality database available and up-to-date so that the population can have access to current and past levels of pollutants, both for the conduct of research and to serve as a public policy instrument.

3.5 Conclusion

Adopting the WHO's $PM_{2.5}$ annual air quality guidelines, between 2,380±800 and 6,280±1,820 deaths due to all causes could be avoidable in 2017 in just 15 evaluated cities in Brazil. These numbers show the importance of adoption of a $PM_{2.5}$ guideline in Brazil and improving the monitoring of air quality, expanding throughout the national territory. As $PM_{2.5}$ is also produced via secondary formation in the atmosphere (SEINFELD and PANDIS, 2006), reducing the concentration of other pollutants may result in a decrease of the $PM_{2.5}$ formation. Policies and investments supporting cleaner transport, power generation, industry emissions control and better municipal waste management would reduce key sources of fine particles and reduce the exposure.

The accuracy of results depends on air quality data, exposed population, concentration-response functions and mortality incidence rates. The population and mortality data were obtained from a national database. Therefore, the results are expected to be affected by the air quality data and concentration-response functions. It was used a single annual $PM_{2.5}$ concentration to represent each city. But it is known that there are large variations in concentration across different geographic and meteorological areas, even within a city. For some cities, there was more than one monitoring station, which makes the value obtained more representative, including the local traffic emissions and long-range transport contributions. However, in other, as Belo Horizonte, just one monitoring site was available, sometimes far for the urban center. Therefore, the concentration value obtained may not represent the real mean concentration for the city but is still an indicator of local pollution and may represent a minimum value that would be found in the urban center for that city. As an alternative for monitoring data is the use of chemical transport models, as CMAQ and WRF-Chem, for example. However, an emission inventory with high spatial resolution is required, as a well-described meteorological field. In addition, the modeled results must be validated with monitoring concentrations and the effect of spatial resolution must be evaluated (PUNGER and WEST, 2013; JIANG and YOO, 2018).

The actual impact of air pollution on health presented here shows the importance of adopting more restrictive air quality standards. Such information is essential to implementing, monitoring and evaluating policies that help to tackle air pollution while also protecting health. Therefore, a review of the nation Brazilian air quality standards is necessary, as the inclusion of pollutants not yet legislated, as $PM_{2.5}$. The importance of using local cohort studies to estimate health benefits is also recorded. Unfortunately, in Brazil, long-term cohort studies for $PM_{2.5}$ are non-existent.

4. TOP-DOWN VEHICLE EMISSION INVENTORY FOR SPATIAL DISTRIBUTION AND DISPERSION MODELLING OF PARTICULATE MATTER

4.1 Introduction

Emissions inventories of atmospheric pollutants are strategic environmental management tools. Their development is the starting point for the successful implementation and reorientation of any program that aims to improve air quality (PULLES and HESLINGA, 2010; RUSSO *et al*., 2019 In Brazil, there is a great effort to build emission inventories. However, as the air pollution inventory studies focus on emissions released within an area, they usually do not distribute the emissions spatially and temporally (RÉQUIA Jr. *et al*., 2015). One approach that has been used to disaggregate vehicular emissions over urban areas in Brazil, mainly in São Paulo, is the Night Light Intensity (NLI) satellite imagery, described by Martins *et al*. (2008; 2010). In this case, the map of lights spots is used to estimate the vehicular density in each grid point of the domain (VARA-VELA *et al*., 2016; ALBUQUERQUE *et al*., 2018; GAVIDIA-CALDERÓN *et al*., 2018; ALBUQUERQUE *et al.*, 2019). In Colombia, the use of road density factors to spatial and temporal disaggregation of on-road vehicle emission inventory was investigated by Gómez *et al*. (2018). Alonso *et al*. (2010) extrapolated a few local vehicle emissions inventories to other cities in South America using a correlation between socio-economic data and CO and NOx emissions of mobile sources.

In Brazil, emission inventories are usually provided by environmental agencies or companies, by individual sources, reporting annual emission rate (tons per year), mainly for each pollutant legislated at a given latitude and longitude. However, few environmental agencies report emission data (RÉQUIA Jr. *et al*., 2016). As air quality models require 3D emissions input data, spatialized in an area and time, it is necessary to process the primary data, and transform them into a suitable format to be able to apply them to air quality models. However, when the area of interest is regional or a greater scale domain, local inventories may not cover all necessary areas. Consequently, there is a need for global inventories to perform air quality modeling for those areas.

The Preprocessor of Trace Gas and Aerosol Emission Fields for Regional and Global Atmospheric Chemistry Models (PREP-CHEM-SRC) is a Brazilian software developed by Centro de Previsão de Tempo e Estudos Climáticos (CPTEC/INPE) to provide gridded emissions of trace gases and aerosols, with flexible grid spacing, multiple projections, and for use in regional and global air quality models (FREITAS *et al*., 2011). The emission fields generated by this tool can be used by numerous air quality models, but it was initially developed to be applied in the Brazilian developments on the Regional Atmospheric Modeling System - BRAMS (FREITAS *et al*., 2017), and it was subsequently adapted to be used in the Weather Research and Forecasting (WRF) model coupled with Chemistry - WRF-Chem (GRELL *et al*., 2005). The emissions considered in PREP-CHEM-SRC are from global urban/industrial databases, biomass burning, biogenic, volcanic, biofuel use and burning from agricultural waste sources (FREITAS *et al*., 2011).

Global inventories usually have low spatial resolution and do not represent specific characteristics of urban areas, especially the representation of urban centers (ALONSO *et al*., 2010). In the most urban areas, vehicles are usually responsible for most of the particulate matter (PM) emissions (RÉQUIA Jr. *et al*., 2015; VARA-VELA *et al*., 2016; PACHECO *et al*., 2017; ANDRADE *et al*., 2017; ALBUQUERQUE *et al*., 2018; GAVIDIA-CALDERÓN *et al*., 2018; POLICARPO *et al*., 2018; MIRANDA *et al*., 2019). Because of the limited number of local inventories available in Brazil, and to improve the spatial distribution and PM modelling performance, this work evaluates the use of the PM emissions estimated in the last Brazilian official vehicular emission inventory (VEI) at country level for air quality modeling. The PM emission was spatial distributed based on population and fleet of each Brazilian city.

The particulate matter directly affects health in the short and long-term (FREITAS *et al*., 2013; SOUZA *et al*., 2017; GOUVEIA and JUNGER, 2018). It is of great concern in Brazil (MIRANDA *et al*., 2012; PACHECO *et al*., 2017; ANDREÃO *et al*., 2018), and therefore remains the focus of this work. The finds from this work are expected to help evaluate the PM concentration in the metropolitan areas in the future, along with allowing investigations of the relationship between PM and excess mortality and morbidity.

In this work, the total Brazilian emission of PM from vehicular sources was distributed into the urban areas of the 5557 municipalities, with 1 km grid spacing. The inventory was compiled in PREP-CHEM-SRC, and a month modeling (August 2015, which is representative of a typically dry month) was performed with the WRF-Chem model for the four metropolitan areas in the Brazilian southeast, the most developed region with the most populous cities of the country. Additionally, the results were compared with modeling considering the EDGAR global inventory. All modeling was validated with monitoring data. Therefore, it will be possible to discuss the importance of vehicle emissions in urban areas.

Section 4.2 presents the methodology used to compile the proposed VEI in PREP-CHEM-SRC, as the air quality modeling configuration and modeling validation method. Section 4.3 presents a comparison with local emission inventories, meteorology monitoring data, and PM concentration validation. Section 4.4 discusses the results obtained.

4.2 Methodology

Figure 4.1 shows a flowchart with all the major activities carried out in this study. Here we discuss the left side of the flow chart, which presents the methodology used to distribute the PM emission from the 2nd Top-Down Brazilian National Inventory of Atmospheric Emissions by Road Motor Vehicles, based on population and fleet of each city, besides the compilation method used in PREP-CHEM-SRC. In sequence, the air quality modeling setup and additional emission inventories (biomass burning, for example) are presented. A second air quality modeling was performed considering EDGAR inventory. The methodologies for meteorological conditions and PM concentration validation end this section. The right side of the flowchart represents the results obtained (Section 4.3).

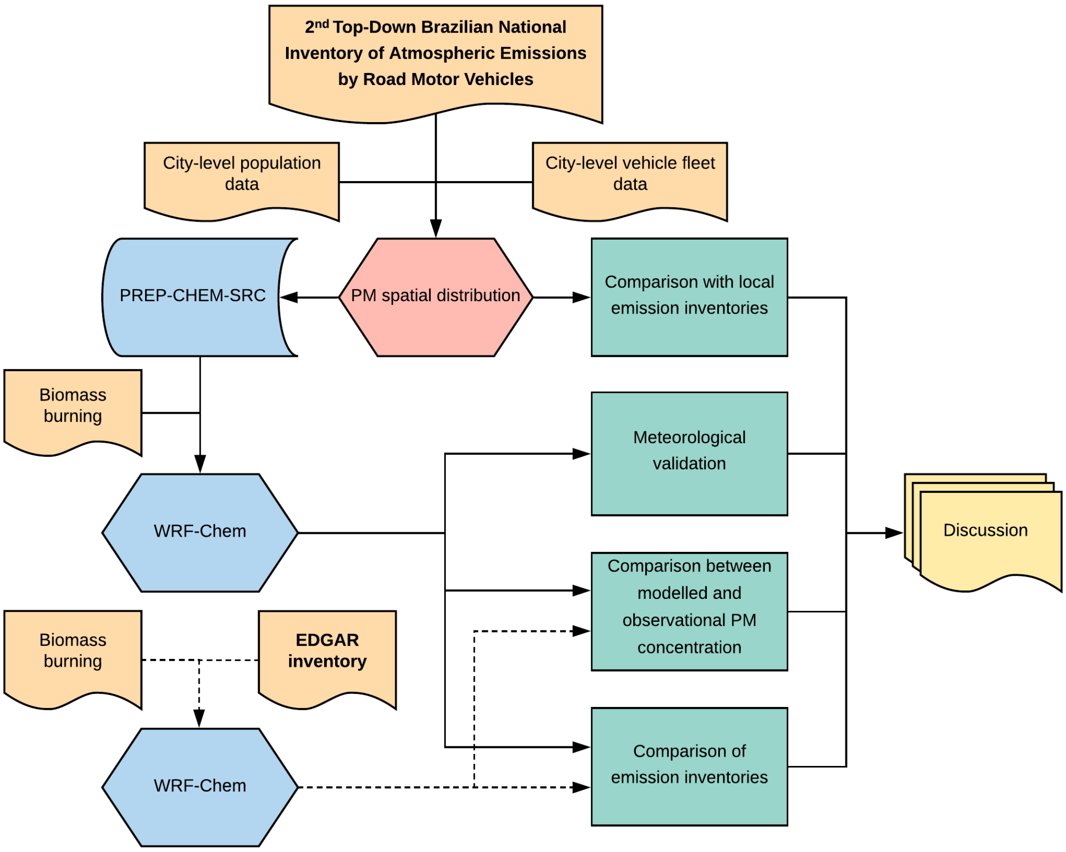

Figure 4.1 - Flow chart, including all major activities carried out in this study.

4.2.1 PM spatial distribution into urban areas

The top-down approach for disaggregate vehicular emission requires less time and resource-consuming, compared to the bottom-up approach, because of the simpler specific statistical data instead of complex traffic models (traffic flow in every segment of the road network). After the top-down inventory distribution, hotspots of emission may also be identified (GÓMEZ *et al.*, 2018).

In this work, the total annual PM (63,000 t) emission estimated for 2012 (last vehicular emission estimation so far) by Brazilian Ministry of the Environment (Brazil 2013) from the 2nd Top-Down Brazilian National Inventory of Atmospheric Emissions by Road Motor Vehicles were distributed among the 5557 Brazilian cities considering two approaches: population and vehicle fleet at city-level. Regarding the proportions of different sources of PMs in the VEI, PM emissions considered tailpipe exhaust emission (59% of total PM), wear from tire and break (26% of total PM), and wear of road surface (15% of total PM) from close to 49 million vehicles. Among the fleet, 57% corresponded to automobiles (57% with a flex-fuel engine, 40% with a gasoline engine and only 3% still dedicated to hydrated ethanol), 28% to motorcycles (84% were gasoline and 16% flex fuel), 11% to light commercial vehicles (38% were flex-fuel vehicles, 36% gasoline, 27% diesel-powered and only 2% dedicated to ethanol), 3% to trucks and 1% to buses (Brazil 2013). The flex-fuel engine is designed to allow the use of gasoline, ethanol or any mixture between these two fuels. In 2015, the Brazilian population was estimated in around 204 million inhabitants (preliminary estimates prepared by General Coordination of Epidemiological Information and Analysis - CGIAE of the Secretariat of Health Surveillance - SVS, Ministry of Health - MS).

After distribution, the emission for each city (in t $year^{-1}$) was recalculated, dividing by its corresponding urban area (in t $year^{-1}$ km^{-2}), and allocated in the urban areas (assuming uniform population and fleet distribution within each city). The vector representation of each urban area (shapefile) used is the result of the work "*Identificação, mapeamento e quantificação das áreas urbanas do Brasil*" developed by *Embrapa Gestão Territorial* in Campinas (São Paulo State). The polygons that represent each urban area (Figure 4.2a) containing the city emissions were converted in raster format with 1 km grid spacing (Figure 4.2b). After, each raster grid cell was converted into points (Figure 4.2c), and PM emission values for each point were summarized in a .txt file and inserted in PREP-CHEM-SRC v.1.5 (FREITAS *et al*., 2011). Based on Santos (2018) and IEMA (2019), it was considered that 63.1% of the total PM is $PM_{2.5}$ and 86.3% of the total PM is PM_{10}.

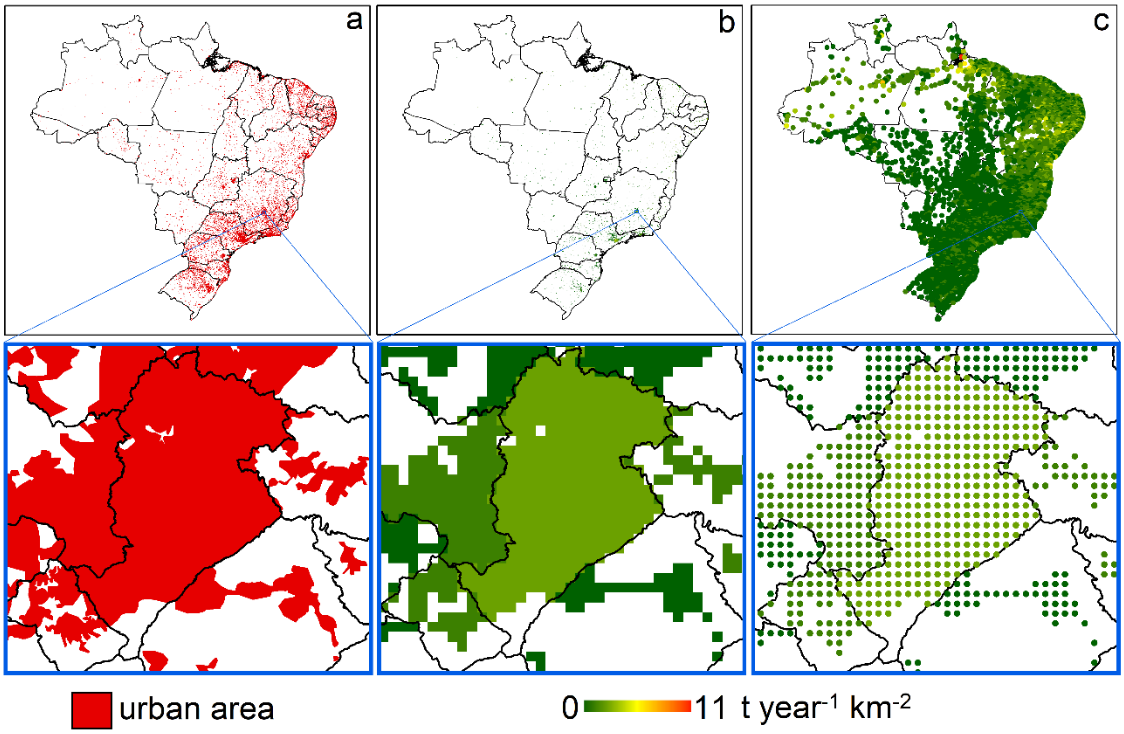

Figure 4.2 - Brazilian urban areas represented by a) polygons; b) raster; c) points; with Belo Horizonte city highlighted.

4.2.2 Air quality modeling setup

To evaluate ambient PM concentration from the developed inventory, one month-long modeling (August 2015) was performed with WRF-Chem (GRELL *et al*. 2005) v.3.9.1.1 for four metropolitan areas (MA): Belo Horizonte (MABH), Great Vitória (MAGV), São Paulo (MASP), and Rio de Janeiro (MARJ), all located in Brazilian Southeast, according to Figure 4.3. August is a winter month, characterized by the worst conditions for pollutants dispersion and the driest month for the region. Table 4.1 brings the number of cities, population and fleet of each metropolitan area for 2015. Together, the four metropolitan areas represented 20.0% and 22.6% of the national population and fleet in 2015, respectively, almost 30% of the Brazilian GDP (2013), with the urban areas occupying almost 25% of the MA territory.

Table 4.1 - Brazilian Southeast metropolitan areas (MA) main characteristics.

MA	Cities	Capital	Urban area/ Total area	Fleet	Population	Fleet/ Population
MABH	34	Belo Horizonte	14.1%	2,830,841	5,239,382	54.0%
MAGV	7	Vitória	17.5%	821,275	1,910,093	43.0%
MARJ	22	Rio de Janeiro	32.2%	4,553,677	12,578,827	36.2%
MASP	39	São Paulo	32.7%	12,256,922	21,090,793	58.1%
Total	102	-	24.9%	20,462,715	40,819,095	50.1%

One larger domain (D01) was created covering the four MA with 25 km grid spacing and 79 × 69 grid-cells in longitudinal and latitudinal directions, respectively, centered in 21.50°S and 43.50°W, forming a domain of 1975 km × 1725 km. The domains covering each MA have a grid spacing of 5 km, with 51 grid-cells in both longitudinal and latitudinal extensions, forming domains of 255 × 255 km. Thirty-two vertical levels represented the vertical structure. The meteorological initial and boundary conditions were obtained from the National Center for Environmental Prediction (NCEP) Global Forecast System (GFS) final (FNL) with six-hour time resolution, 26 vertical levels, and a horizontal resolution of 0.25 × 0.25 degrees (ds083.3). The main physics options, based on Vara-Vela *et al*. (2018), which performed WRF-Chem modeling over MASP to study the impact of biomass burning in the atmospheric aerosol properties, and the chemistry options used are listed in Table 4.2.

Table 4.2 - WRF-Chem configurations considered.

Attributes	WRF-Chem option
Radiation	Longwave and shortwave RRTMG scheme (IACONO *et al*., 2008)
Surface layer	Revised Mesoscale Model version 5 Monin-Obukhov scheme (JIMÉNEZ *et al*., 2012)
Land surface	Unified Noah land surface model (CHEN and DUDHIA, 2001)
Boundary layer	Yonsei University scheme (HONG *et al*., 2006)
Cumulus clouds	Multiscale Kain-Fritsch scheme (ZHENG *et al*., 2016)
Cloud microphysics	Morrison two moment (MORRISON *et al*., 2009)
Gas phase	Regional Acid Deposition Model version 2 (RADM2) (CHANG *et al*., 1989)
Aerosol	Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model (GOCART) (CHIN *et al*., 2000)
Photolysis	Fast Troposphere Ultraviolet Visible (TIE *et al*., 2003)

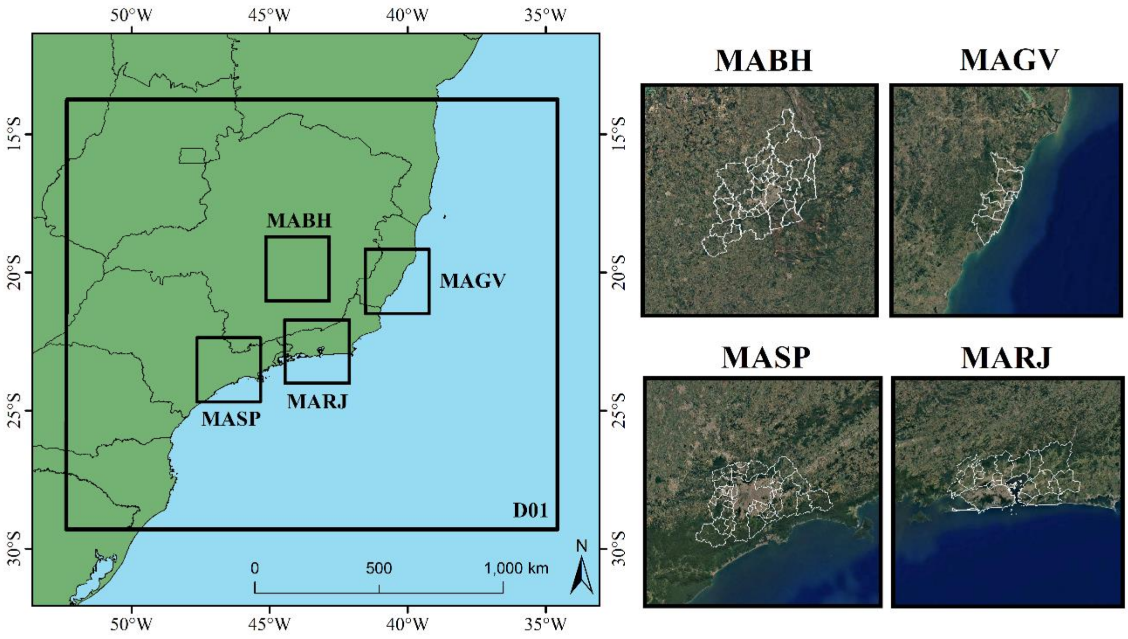

Figure 4.3 - Modeling domains: metropolitan areas of Belo Horizonte (MABH), Great Vitória (MAGV), Rio de Janeiro (MARJ), and São Paulo (MASP).

Mozbc tool, developed by National Center for Atmospheric Research (NCAR)/Atmospheric Chemistry Observations & Modeling (ACOM), was used for creating chemical boundary conditions varying in time, and initial conditions for the larger domain (D01), using the output of the global Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4)/Goddard Earth Observing System Model, version 5 (GEOS-5), being the most used tool in WRF-Chem for this purpose (GAVIDIA-CALDERÓN *et al*., 2018). The initial and lateral boundary conditions for 5-km grid runs were obtained from the 25-km grid run, using a one-way nesting technique with *ndown* tool from WRF-Chem. For all simulations, it was considered 10 days for spin-up (HOGREFE *et al*., 2017).

4.2.3 EDGAR inventory

Additional to the proposed inventories, air quality modeling using the 2010 global inventory Emission Database for Global Atmospheric Research (EDGAR) was performed. EDGAR provides global annual emissions of greenhouse gases, ozone precursors, acidifying gases, and PM. Its database was built considering the location of power and manufacturing facilities, road networks, transportation routes, human and animal population density and agricultural land use, which vary over the years. Country emissions are then compiled considering the International Energy Agency (IEA) energy statistics. The total national emissions are gridded using population, road, power plants, animals, and crop proxy data, for example (JANSSENS-MAENHOUT *et al*., 2013; JANSSENS-MAENHOUT *et al*., 2015). The EDGAR inventory presented in PREP-CHEM-SRC dates back to 2010 and presents agriculture, energy, industries, residences, and transportation emissions, with a spatial allocation of 0.1 degrees × 0.1 degrees.

4.2.4 Additional inventories considered

For the air quality modeling, biogenic, aerosol background, and biomass burning emissions were also considered for all air quality scenarios: fleet and population distribution and EDGAR, according to the respective inventory presented in PREP-CHEM-SRC. The MEGAN global biogenic emission model database available in PREP-CHEM-SRC dates back to the year 2002, with a monthly time resolution, and it has a spatial resolution of 0.5 degrees × 0.5 degrees.

The GOCART model simulates major tropospheric aerosol components, including sulfate, dust, elemental carbon (EC), organic carbon (OC), and marine aerosol. GOCART model uses the assimilated meteorological fields of the Goddard Earth Observing System Data Assimilation System (GEOS DAS). The model has a horizontal resolution of 2.0 degrees × 2.5 degrees or 1.0 degree × 1.0 degree, and 20 to 55 vertical sigma layers, depending on the GEOS DAS version. The emissions of EC, OC, and SO2 have an annual temporal resolution, with 2006 being the most recent year available in PREP-CHEM-SRC, while the emissions of dimethyl sulfide, NO_3, H_2O_2, and OH have monthly temporal resolutions.

In the Brazilian Biomass Burning Emission Model (3BEM) for each fire pixel detected by remote sensing, the pollutant mass emitted is calculated taking into account the estimated values for the available amount of biomass above the ground for burning, combustion factor, the emission factor for a given chemical species and the burning area. The total mass emitted from each chemical species is given per domain cell per day. In PREP-CHEM-SRC, the following combination of 2015 fire database was used to maximize remote sensing observations: 1) Geostationary Operational Environmental Satellite - Wildfire Automated Biomass Burning Algorithm (GOES WF_ABBA), 2) *Instituto Nacional de Pesquisas Espaciais* (INPE), based on the Advanced Very High Resolution Radiometer (AVHRR) aboard the National Oceanic and Atmospheric

Administration (NOAA) series of satellites, in polar orbit, and 3) the Moderate Resolution Imaging Spectroradiometer (MODIS). The double fire counting is avoided due to a filter algorithm in the code, which eliminates fires from different databases within a radius of 1 km (FREITAS *et al*. 2011).

To be used in WRF-Chem, the generated emission files were converted to the appropriate format using the convert_emiss tool of WRF-Chem v. 3.6. Emissions generated by PREP-CHEM-SRC are made for the RADM2 chemical mechanism and GOCART speciation (bulk aerosol scheme) (ARCHER-NICHOLLS *et al.*, 2015). The regional Acid Deposition Model version 2 (RADM2) (CHANG *et al.*, 1989) is widely used in atmospheric models to predict concentrations of oxidants and other atmospheric pollutants and includes 59 chemical species and 157 reactions. For the aerosol module, the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model (GOCART) (CHIN *et al*., 2000) is a bulk aerosol scheme for species with non-reactive species (dust and sea salt). Only total mass of aerosol compounds is known and no secondary organic aerosol is considered in this approach. GOCART scheme includes 14 defined aerosol species and a 15th variable representing unspecified aerosol contributions (P25). The 14 species of aerosols defined are: sulfate; hydrophobic (OC1) and hydrophilic (OC2) organic carbon; hydrophobic (BC1) and hydrophilic (BC2) elemental carbon; dust in five particle sizes (effective rays of 0.5, 1.4, 2.4, 4.5 and 8.0 μm, referred to as D1, D2, D3, D4 and D5, respectively); and sea salt in four particle size distributions (effective radiations of 0.3, 1.0, 3.25 and 7.5 μm for dry air, referred to as S1, S2, S3 and S4 in the code, respectively) (PENG *et al*., 2017). Due to its simplicity compared to other aerosol schemes, GOCART is numerically efficient.

4.2.5 Meteorological validation

The meteorological modeling validation was performed by comparing hourly modeled results with surface data observed by 32 meteorological stations of the *Instituto Nacional de Meteorologia* in Brazil. The following statistical indices suggested by Emery et al. (2001) was investigated: Mean Bias (MB); Mean Error (ME); Root Mean Square Error (RMSE); and Index of Agreement (IOA), according to Equations 4.1 to 4.4, respectively, for wind speed and direction, temperature and specific humidity. These indices have been widely used (GAO *et al*., 2015; WANG *et al*., 2016a; MUGHAL *et al*., 2017; DANDOU *et al*., 2017; PERMADI *et al*., 2018; MUES *et al*., 2018; PEDRUZZI *et al*., 2019).

Benchmarks for complex conditions suggested by Ramboll (2018) and LADCO and WDNR (2018) was used. For those indices that did not present criteria for complex conditions, the criteria for simple conditions suggested by Emery *et al*. (2001) was used.

$$MB = \frac{1}{n}\sum_{i=1}^{n}(\varphi_{mod} - \varphi_{obs}) \tag{4.1}$$

$$ME = \frac{1}{n}\sum_{i=1}^{n}|\varphi_{mod} - \varphi_{obs}| \tag{4.2}$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(\varphi_{mod} - \varphi_{obs})^2} \tag{4.3}$$

$$IOA = 1 - \left[\frac{\sum_{i=1}^{n}(\varphi_{mod} - \varphi_{obs})^2}{\sum_{i=1}^{n}\left(\left|\varphi_{mod} - \overline{\varphi}_{obs}\right| + \left|\varphi_{obs} - \overline{\varphi}_{obs}\right|\right)^2}\right] \tag{4.4}$$

where φ_{mod} is the modeled parameter and φ_{obs} is the observed parameter. Overbars signify means over the site.

For wind direction, the validation must be treated carefully. Since the wind direction is a circular variable, Equations 4.1 and 4.2 must be adapted to consider the fact that the absolute deviation of wind direction cannot exceed 180 degrees in modulus. Therefore, if $|\varphi_{mod} - \varphi_{obs}| > 180°$, the MB is calculated by Equation 4.5 and ME by Equation 4.6 (MUGHAL *et al*., 2017).

$$MB = \frac{1}{n}\sum_{i=1}^{n}(\varphi_{mod} - \varphi_{obs})\left|\left[1 - \left(\frac{360}{|\varphi_{mod} - \varphi_{obs}|}\right)\right]\right| \tag{4.5}$$

$$ME = \frac{1}{n}\sum_{i=1}^{n}|\varphi_{mod} - \varphi_{obs}|\left|\left[1 - \left(\frac{360}{|\varphi_{mod} - \varphi_{obs}|}\right)\right]\right| \tag{4.6}$$

Although Emery *et al*. (2001) proposed benchmarks for wind direction, the use of the circular correlation coefficient (CCC) is also recommended (FISHER and LEE, 1983; CAIN, 1989; JAMMALAMADAKA and LUND, 2006; PAPANASTASIOU *et al*., 2010;

MA *et al*., 2016), according to the Equation 4.7, where a and b are two angular variables, with mean directions $\bar{a}$ and $\bar{b}$, respectively.

$$CCC = \frac{\sum_{i=1}^{n}\left[sin(a_i - \bar{a}) \times sin\left(b_i - \bar{b}\right)\right]}{\left\{\left[\sum_{i=1}^{n} sin^2(a_i - \bar{a})\right] \times \left[\sum_{i=1}^{n} sin^2\left(b_i - \bar{b}\right)\right]\right\}^{0.5}} \quad (74.)$$

4.2.6 Air quality modeling evaluation

The air quality modeling results were evaluated by comparing the PM_{10} and $PM_{2.5}$ modelled concentrations with environmental from 61 air quality monitoring stations available in each metropolitan region, applying the following statistical indices: Normalized Mean Bias – NMB (Equation 4.8), Normalized Mean Error – NME (Equation 4.9), and correlation coefficient – *r* (Equation 4.10), according to Simon *et al*. (2012) and Emery *et al*. (2017).

$$NMB = \frac{\sum(\varphi_{mod} - \varphi_{obs})}{\sum \varphi_{obs}} \times 100 \quad (4.8)$$

$$NME = \frac{\sum|\varphi_{mod} - \varphi_{obs}|}{\sum \varphi_{obs}} \times 100 \quad (4.9)$$

$$r = \frac{\sum_{n=1}^{N}\left[\left(\varphi_{mod} - \overline{\varphi}_{mod}\right) \times \left(\varphi_{obs} - \overline{\varphi}_{obs}\right)\right]}{\sqrt{\sum_{n=1}^{N}\left(\varphi_{mod} - \overline{\varphi}_{mod}\right)^2 \times \sum_{n=1}^{N}\left(\varphi_{obs} - \overline{\varphi}_{obs}\right)^2}} \quad (4.10)$$

4.3 Results

4.3.1 Comparison with local emission inventories

Vehicular emission inventories can be compiled using top-down and bottom-up approaches. The first one is based on vehicle composition, traffic speeds, and country balances, with the result being the total emission for a region. The second one is more refined, using detailed data on each vehicular emission sources, such as road vehicle, and give the emission per segment of road. As a consequence, this approach is expensive and time-demanding (GÓMEZ *et al*., 2018; PINTO *et al*., 2020a).

In this context, a direct comparison between emission inventories should be made carefully, considering the differences related to the methodologies used to estimate the emissions in each inventory, such as emission factors, vehicle category, vehicle age, and fuel considered, for example. Table 4.3 summarizes PM emissions for Belo Horizonte, Federal District, and Metropolitan Areas of Rio de Janeiro (MARJ), São Paulo (MASP), Vitória (MAGV), and Fortaleza (MAF), and compare the presently estimated emission based on population and fleet with a local reference.

Table 4.3 - Comparison of PM [t year^{-1}] emission with inventories.

	PM [t year^{-1}]		
Local	Reference[a-f]	Population	Fleet
Belo Horizonte	1,220[a]	772	1,192
Federal District	5,107[b]	899	1,147
MARJ	351[c#*]	2,230*	1,807*
MARJ	889[c*]	2,230*	1,807*
MASP	1,529[d]	6,508	8,523
MAGV	490[e]	546	523
MAF	604[f#*]	621*	498*

References.: [a]Santos (2018); [b]Réquia Jr. *et al*. (2015); [c]INEA (2016b); [d]CETESB (2016); [e]IEMA (2019); [f]Policarpo *et al*. (2018); [#]Bottom-up methodology; *only PM exhaust emission.

A better agreement of the estimated emission was obtained for Belo Horizonte, MAGV, and MAF when comparing to the local emission inventories. In MAGV, it was observed the smaller difference in the estimated emission between population and fleet segregation, both with a good concordance with IEMA (2019). In MASP, otherwise, the fleet segregation resulted in higher PM emissions than the population segregation, the opposite of MAF.

We highlight the different methodologies in which each inventory was constructed. The fleet segregation presented a close emission value for Belo Horizonte, with 2.3% of the difference. The methodology carried out by Santos (2018) was the same as the VEI used in this work, which explains the small difference found. In MARJ, otherwise, INEA (2016) considered only the exhaust emissions. Therefore, in MARJ, exhaust emission for the VEI accounted for 2,230 and 1,807 t year^{-1} for population and fleet segregation, respectively. Moreover, INEA (2016) used the bottom-up methodology for the estimation of line sources on some stretches of public roads. Different databases for emission factors were also used. Also, INEA (2016) also presented the total PM emission (exhaust plus resuspension), 1,266.86 t year^{-1}, which was still below the values obtained in this work.

The top-down estimation for MARJ of INEA (2016) (emissions as area source; diffuse sources) also resulted in lower emissions than those found in the present study, which may be attributed to different emission factors and database sources for the total number of vehicles per type, and fuel, for example. As the resulted emission is directly proportional to the emission factor, circulating vehicle fleet, and intensity of vehicle use, any difference in these three variables will generate different results.

For MASP, the PM for automobiles estimated by CETESB (2016) considered just flex-fuel vehicles using Gasoline C. As discussed in Pacheco *et al*. (2017), after 2005, CETESB has modified the methodology to estimate PM emissions in São Paulo, which may contribute to underestimating them, compared to the segregated national VEI. In the Federal District, 97.4% of PM emissions estimated by Réquia Jr. *et al*. (2015) were from heavy vehicles. The average total PM emission is five times higher than the estimated in the present work. In Réquia Jr. *et al*. (2015), it was not considered the age of the vehicles and the type of fuel used, which may have contributed to the higher emission observed. In MAF, Policarpo *et al*. (2018) used a macrosimulation, bottom-up method, not considering PM emission from brakes, tires and pavement wear.

Table 3 showed a diverse picture of emission inventories. As there is no standard to construct vehicle emission inventories, the evaluation and comparison among them must be treated individually. The main differences identified are related to different emission factors, databases, fleet characteristics considered, including fuels, vehicle scrap curves, and the methodology itself (top-down or bottom-up).

4.3.2 Meteorological validation

The indices obtained from the comparison between hourly observed and modeled meteorological parameters are presented in Figure 4.4, where the grey area represents the benchmarks suggested by Ramboll (2018) and LADCO and WDNR (2018) for complex situations, except for IOA and wind direction MB, which considered Emery *et al.* (2001) benchmarks for simple situations, and CCC, where we have assumed 0.5 as a reasonable benchmark for this index. Overall, the meteorological parameters were better represented in MABH, which reached all benchmarks for specific humidity and temperature and a good representation for wind direction. The other metropolitan areas also presented good indices for specific humidity.

For temperature, the WRF tends to underestimate it in average 1.0 K in MAGV, while the other metropolitan areas presented, in most of the meteorological stations, positive MB values. RMRJ obtained the highest ME (median equal to 2.3 K). Good IOA values were obtained in the four domains.

All metropolitan areas tend to overestimate wind speed. Shimada *et al*. (2011) found that the annual mean wind speed simulated by the WRF had a “remarkable positive bias” near the surface. Moreover, using different PBL schemes did not reduce the positive bias. Avolio *et al*. (2017), Albuquerque *et al*. (2018), and Pedruzzi *et al*. (2019) also found an overestimation of the wind speed. Nevertheless, the values of RMSE and IOA were closer to the benchmarks suggested by Emery *et al*. (2001).

Regarding wind direction, greater variability in MB was observed in MARJ, which presents the higher ME (median equal to 67 degrees). Pedruzzi *et al*. (2019) stand that wind direction is “*one of the most complex indicators to collect*”. Jiménez and Dudhia (2013) showed that differences between observed and modeled wind direction depend on wind speed, with higher velocities leading to better representation of the direction. Additionally, simulations in complex terrain domains present “*larger systematic differences between model and observations*”. Previous works (e.g., Zhang *et al*., 2013 and Mughal *et al*., 2017) have also concluded that complex terrain affects wind speed and wind direction predictions.

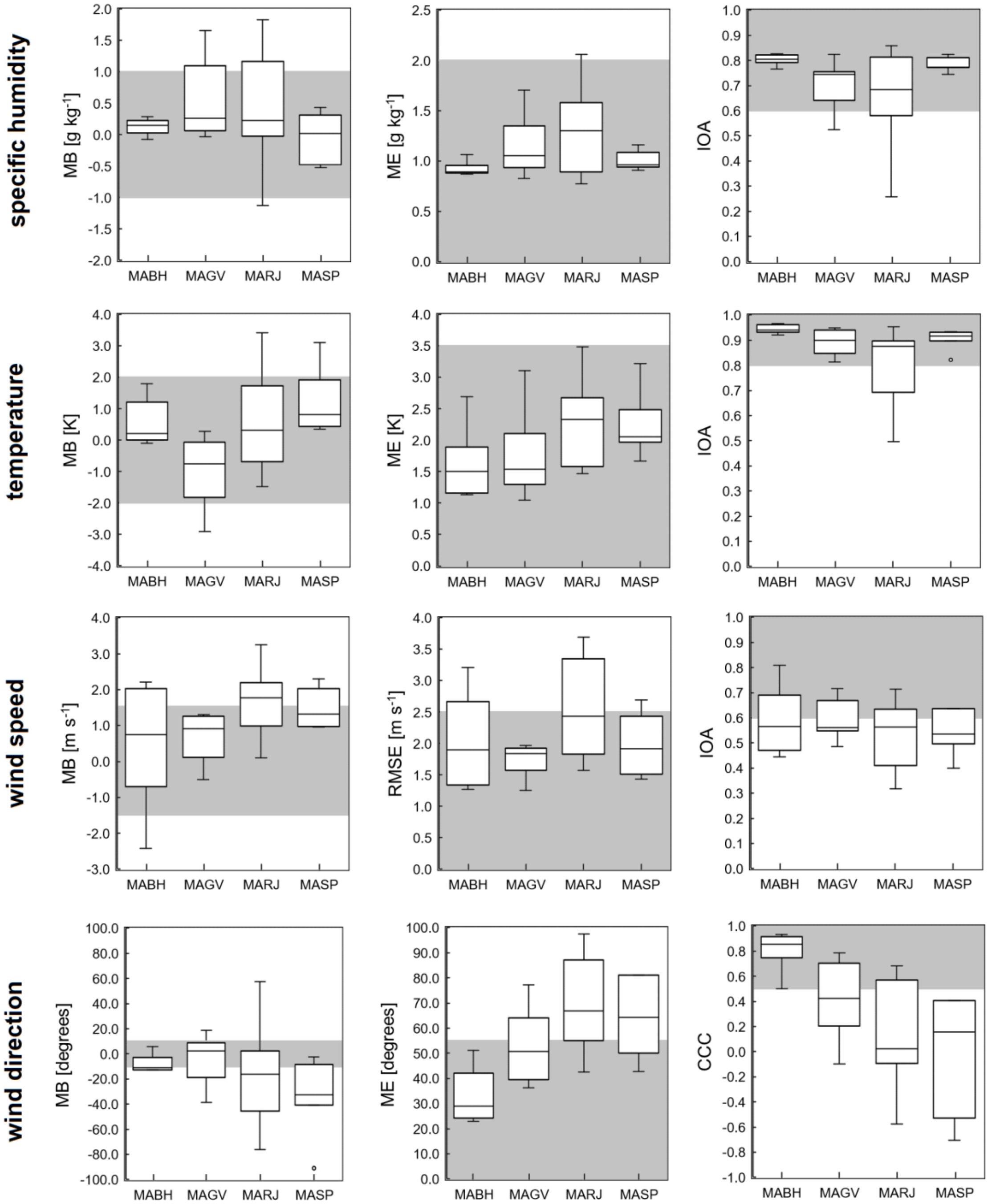

Figure 4.4 - Mean bias (MB), mean error (ME), index of agreement (IOA), root mean square error (RMSE) and circular correlation coefficient (CCC), for specific humidity, temperature, wind speed and wind direction. Grey areas represent the benchmark interval considered.

4.3.3 Comparison of emission inventories

Figure 4.5 shows PM_{10} emission after processed by PREP-CHEM-SRC, with 5-km of grid spacing, for the four metropolitan areas modelled and three inventories, with the maximum PM_{10} emission highlighted for each case. In MABH, emissions are concentrated in the capital Belo Horizonte, and near cities such as Contagem and Betim. These differences between the VEI and EDGAR for PM_{10} is maintained in the other metropolitan areas. In MAGV, the maximum PM_{10} emission with EDGAR was 1.8×10^{-4} kg m^{-2} day^{-1}, higher than Belo Horizonte. In MASP, the EDGAR inventory presented the highest PM_{10} emission among the four metropolitan areas investigated, 2.6×10^{-4} kg m^{-2} day^{-1}. The VEI with population and fleet as variables to distribute the emission has maximums of 1.1×10^{-5} kg m^{-2} day^{-1} and 1.6×10^{-5} kg m^{-2} day^{-1}.

Table 4.4 summarizes the maximum emission rate found in each domain for PM. The maximum emissions were found for PM_{10} and $PM_{2.5}$ with EDGAR inventory for the four metropolitan areas. In MABH and MASP, the fleet distribution presented higher maximum emissions than population distribution, while in MAGV and MARJ, the population distribution presented the maximums.

Table 4.4 - Maximum PM_{10} and $PM_{2.5}$ emission [kg m^{-2} day^{-1}] found in each domain for each inventory investigated.

	MABH			MAGV		
	population	fleet	EDGAR	population	fleet	EDGAR
$PM_{2.5}$	4.98×10^{-6}	7.68×10^{-6}	1.07×10^{-4}	3.21×10^{-6}	3.11×10^{-6}	8.35×10^{-5}
PM_{10}	6.81×10^{-6}	1.05×10^{-5}	1.69×10^{-4}	4.39×10^{-6}	4.25×10^{-6}	1.82×10^{-4}
	MARJ			MASP		
	population	fleet	EDGAR	population	fleet	EDGAR
$PM_{2.5}$	5.23×10^{-6}	4.06×10^{-6}	1.00×10^{-4}	8.37×10^{-6}	1.18×10^{-5}	1.62×10^{-4}
PM_{10}	7.16×10^{-6}	5.55×10^{-6}	2.05×10^{-4}	1.15×10^{-5}	1.61×10^{-5}	2.55×10^{-4}

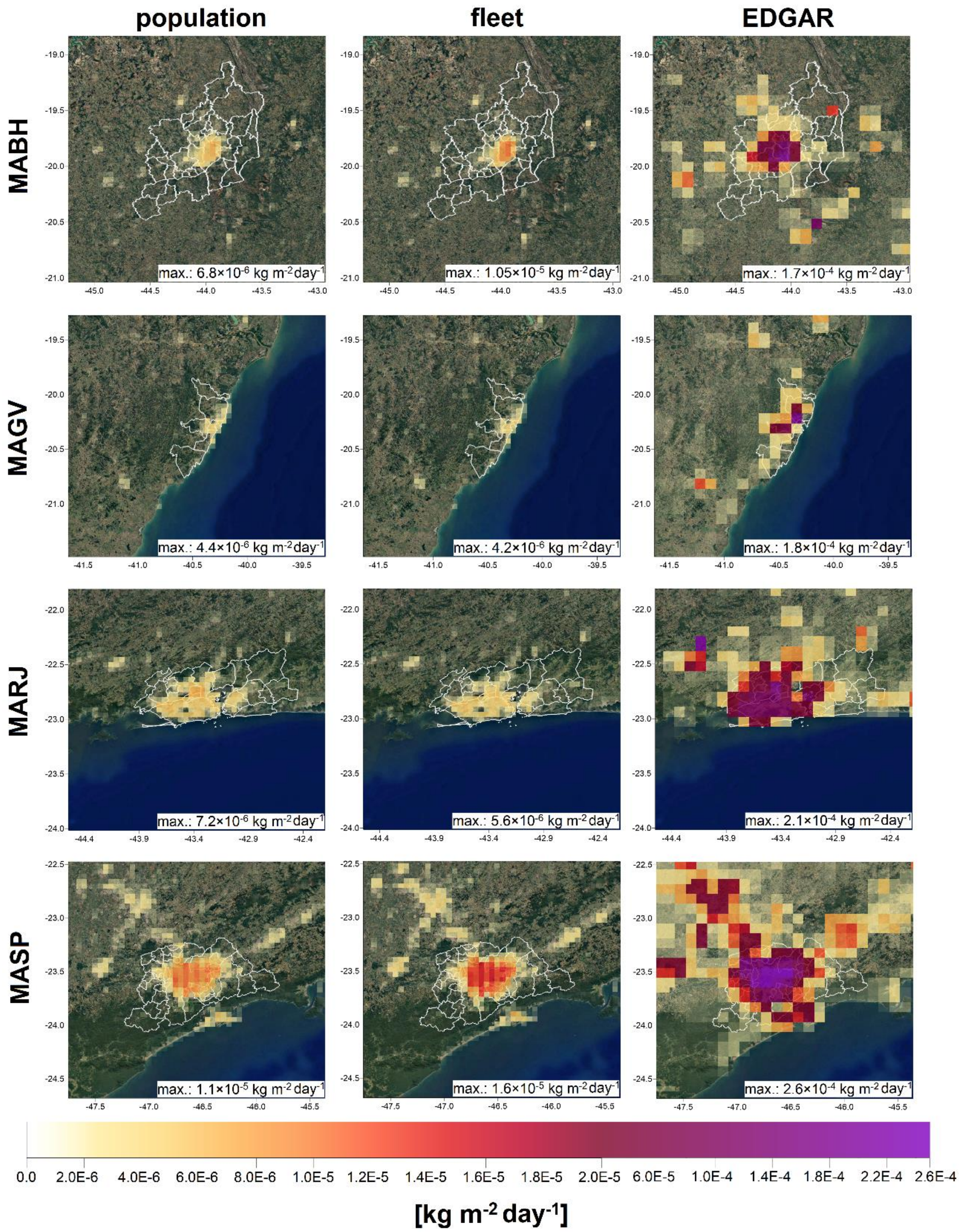

Figure 4.5 - Anthropogenic PM_{10} emission [kg m^{-2} day^{-1}] for metropolitan areas of Belo Horizonte (MABH), Great Vitória (MAGV), Rio de Janeiro (MARJ) and São Paulo (MASP) considering the three inventories investigated.

Table 4.5 summarizes the total anthropogenic PM_{10} and $PM_{2.5}$ emission (kg day^{-1}) found in each metropolitan area, for each inventory processed in PREP-CHEM-SRC. Once again, it is notable the larger differences between the VEI and EDGAR database. It is again highlighted the diverse sources that EDGAR considers: agriculture; energy;

industries; residences; and transportation emissions, while the VEI considered just vehicle sources.

Table 4.5 - Total PM_{10} and $PM_{2.5}$ emission [kg day^{-1}] found in each metropolitan area.

Domain	Pollutant	Inventory		
		population	fleet	EDGAR
MABH	PM_{10}	4,044	4,963	93,712
	$PM_{2.5}$	2,957	3,629	54,715
MAGV	PM_{10}	608	562	16,000
	$PM_{2.5}$	445	411	8,956
MARJ	PM_{10}	3,685	3,435	92,512
	$PM_{2.5}$	2,694	2,511	57,613
MASP	PM_{10}	16,801	22,093	385,181
	$PM_{2.5}$	12,284	16,154	244,581

4.3.4 Air quality modeling

Figure 4.6 shows the monthly-averaged concentration of PM_{10} [µg m^{-3}] for the four MA and three inventories. The highest monthly averages were observed with EDGAR inventory: 68.5 µg m^{-3} in MABH, 147 µg m^{-3} in MAGV, 115 µg m^{-3} in MARJ, and 190.5 µg m^{-3} in MASP. The peak observed in MAGV, also presented in the other two simulations for the region, is due biomass burning close to the coast of MAGV for many days in August 2015, which resulted in PM_{10} high concentration. The monthly average concentration from the VEI segregated by population and fleet presented a similar spatial distribution, which shows that, besides there is a difference between PM emission, it was not sufficient to have a great impact on PM_{10} modeled concentrations, behavior which is also observed with the comparison with monitoring stations measurements.

Figure 4.7 brings the NMB and NME for PM_{10} 24-hr averages for the four metropolitan areas, and Figure 4.8 shows the correlation coefficient found. For MABH, the VEI segregated by population obtained a mean NMB of -46.1%, while the VEI segregated by the fleet, -45.3%, both outside the criteria zone (-30% to 30%). The EDGAR inventory presented a mean NMB of 22.8%, with four of the seven monitoring stations reaching the criteria zone. The mean NME was 47.0%, 46.4%, and 41.4%, for the three inventories, respectively. The r values for MABH were similar for all inventories, all reaching the criteria (> 0.4), with the median close to the goal value (0.7).

For MAGV, the results with the proposed inventories were slightly better, presenting small values of NMB (-9.1%, -9.9%, and 47.7%, on average, for the VEI segregated by population and fleet, and EDGAR inventory, respectively) and NME (50.0%, 48.7%, and 80.1%, on average, respectively). MAGV was the only metropolitan region with r values below the benchmark.

In MARJ, from the 23 monitoring stations with PM_{10} available data, 21% of them were within the criterion limits, while with the EDGAR, 46% of the stations fit into the criterion limits, with 5 stations also reaching the objective limits (-10% to 10%). For NME, 58% of stations met the criteria target with the proposed inventories and 67% with EDGAR. The three inventories obtained a similar variation of r.

In contrast with the other metropolitan regions, in MASP, the proposed inventories obtained better NMB and NME values, compared to EDGAR inventory, which tended to overestimate PM_{10} concentrations. For the VEI segregated by population, 16 of the 24 monitoring stations presented NMB values within the criterion (6 within the goal zone). With the VEI segregated by the fleet, 15 monitoring stations are within the criterion (5 within the goal zone), while with EDGAR inventory, only one station is within the criterion (none within the goal zone). Except for one station, all the other 23 obtained NME values with the criterion for the proposed inventories, with 14 (population) and 12 (fleet) monitoring stations also reaching the goal benchmark. Otherwise, the EDGAR inventory presented two stations reaching the criterion and one the goal benchmark. Even presenting the worst NMB and NME, the modeling with EDGAR inventory presented the higher *r* median (0.7) at the goal limit.

From the comparison with monitoring data, it can be inferred that in MASP vehicular emissions are the main responsible for PM_{10}. In the other metropolitan areas, industrial sources may play an important role in air quality, since with EDGAR inventory, PM_{10} concentrations were better represented. For Brazilian southeast region, industrial (68%) and residential (24%) sectors represents most of the PM emission in the EDGAR inventory used, with transportation accounting for 3.5% of $PM_{2.5}$ and 6.7% of PM_{10}.

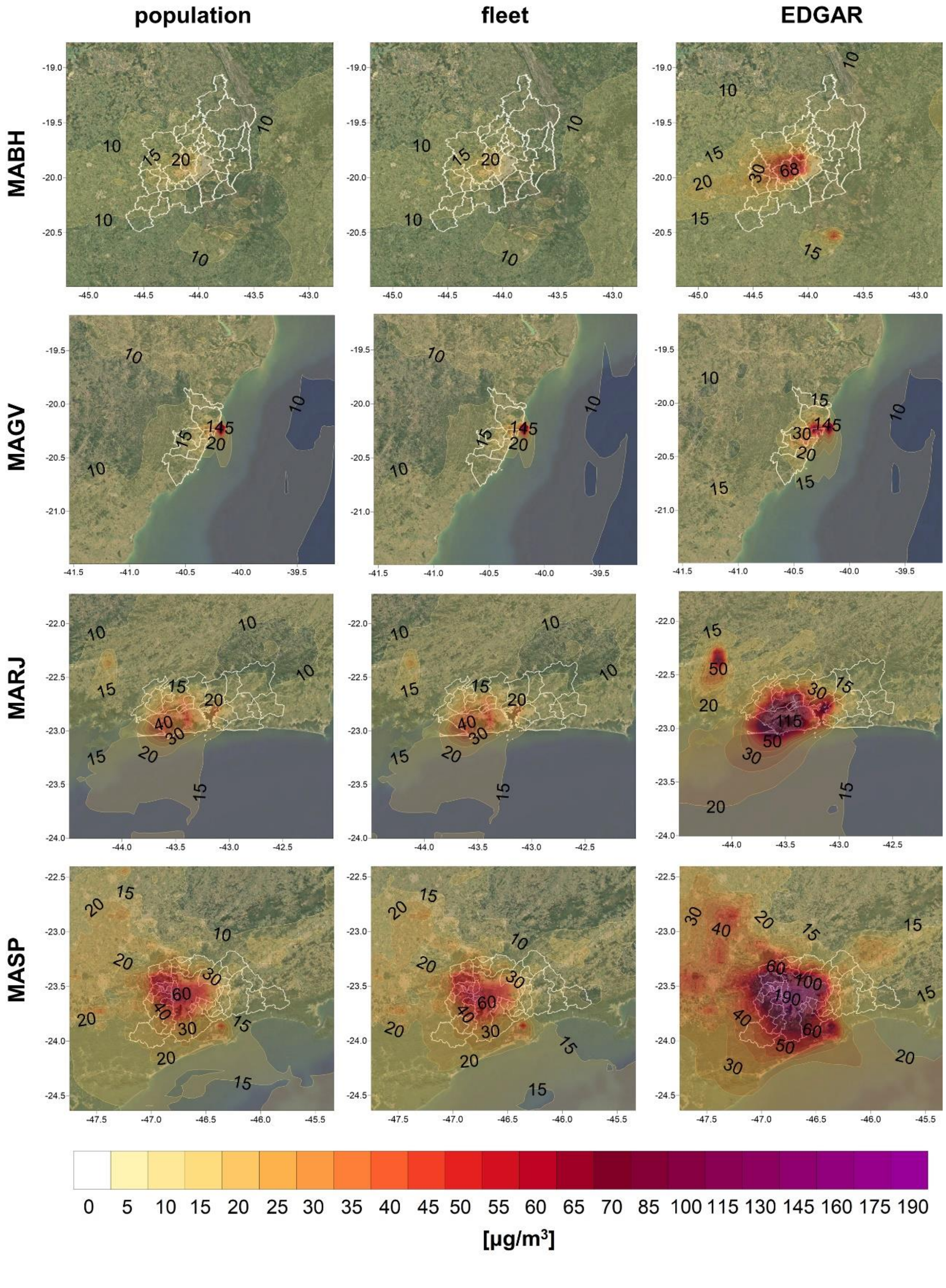

Figure 4.6 - Month-average PM_{10} concentration for metropolitan areas of Belo Horizonte (MABH), Great Vitória (MAGV), Rio de Janeiro (MARJ) and São Paulo (MASP) considering the three inventories investigated.

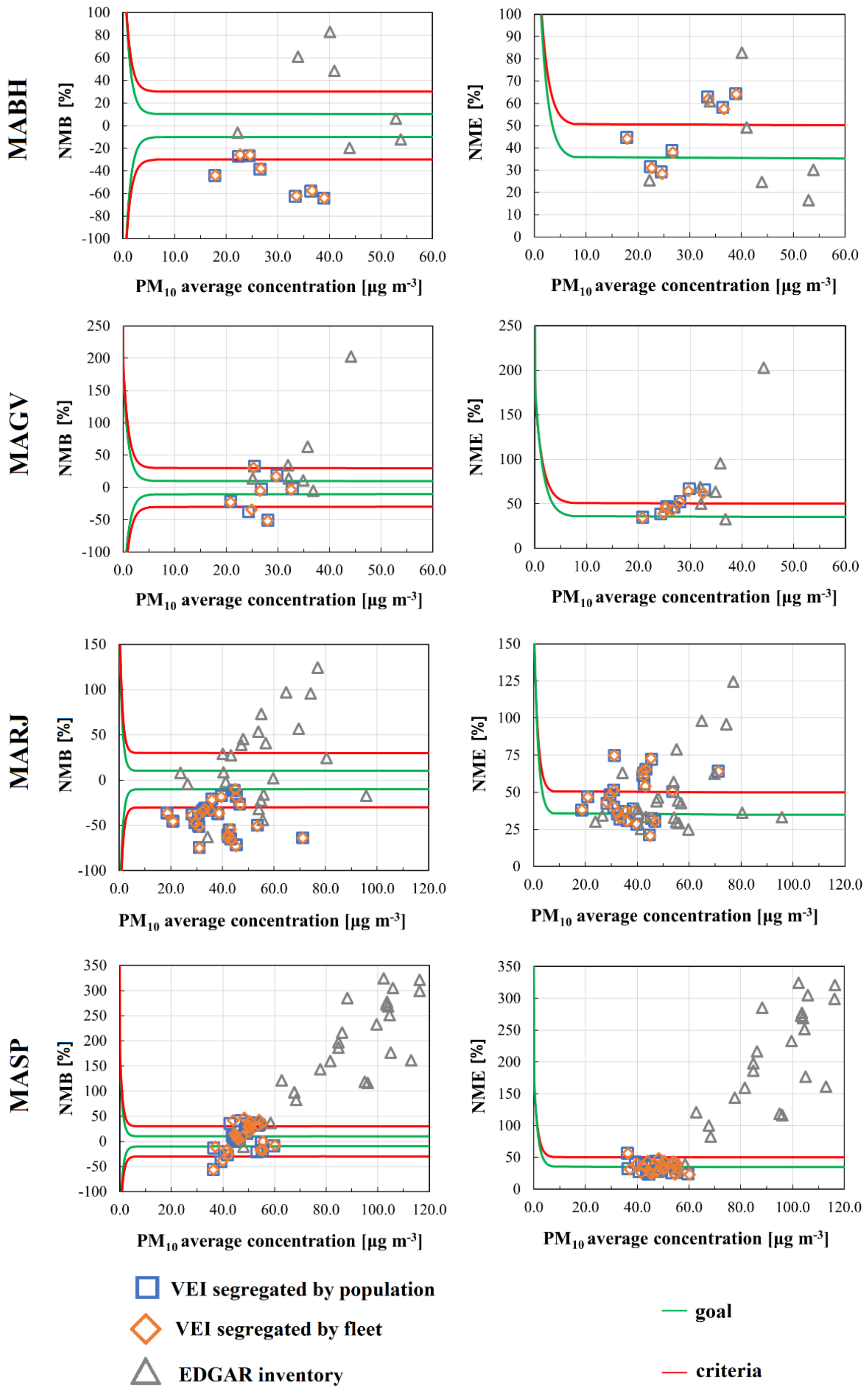

Figure 4.7 - NMB and NME for PM_{10} 24-hr averages for MABH, MAGV, MARJ and MASP, considering the three inventories investigated. The NMB goal and criteria considered < ±10% and < ±30%, respectively, while the NME goal and criteria considered < 35% and < 50%, respectively.

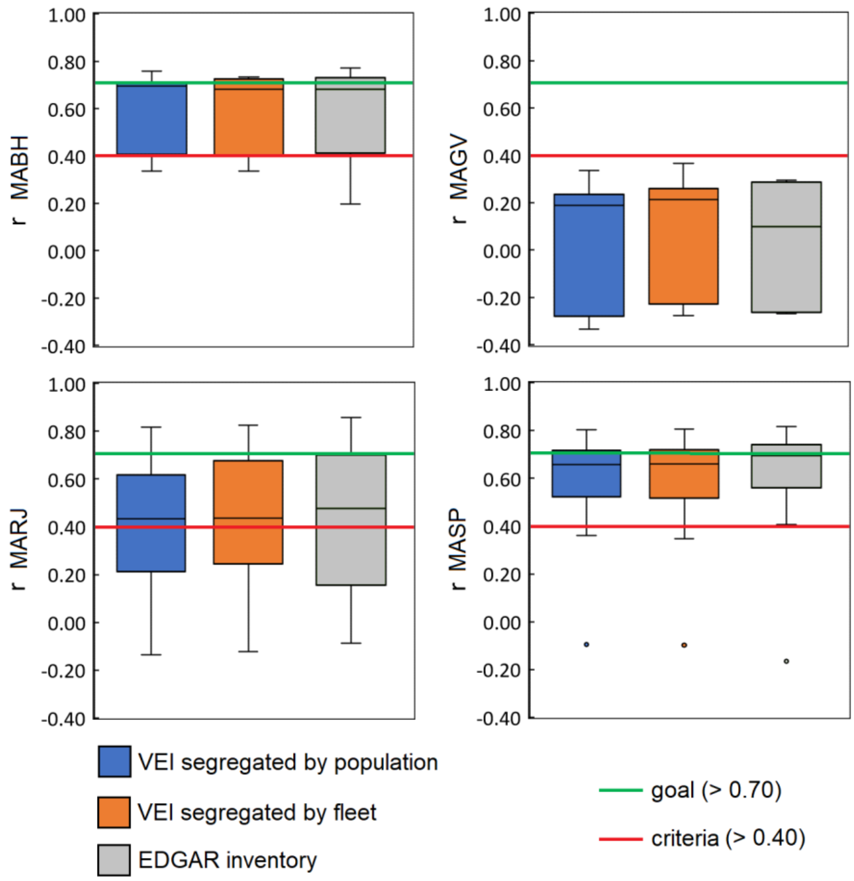

Figure 4.8 - Correlation coefficient (*r*) for 24-hr PM_{10} averages.

Table 4.6 presents the statistic indices for 24-hr $PM_{2.5}$ averages. The indices that obtained the values within the criteria benchmarks are in bold. Only 14 air quality monitoring stations monitored $PM_{2.5}$ in 2015 in the metropolitan areas investigated, most of them in MASP. In MARJ, just one automatic station of the local ones monitors this pollutant continuously. In MABH and MAGV there are two stations each. Overall, the EDGAR inventory overestimates $PM_{2.5}$ in all metropolitan areas, with MASP presenting NMB and NME values much higher than when the VEI was used. Except for MAGV, all r values attended the benchmark (> 0.4), with 50% of the monitoring stations also reaching the goal (> 0.7).

Table 4.6 - Comparison of modelled 24-h $PM_{2.5}$ concentration with monitoring stations data. In bold, the stations that reached the criteria benchmarks suggested by Emery *et al*. (2017): NMB < ± 30%; NME < 50%; and r > 0.40.

MA	Monitoring station	number of 24-h $PM_{2.5}$ averages	VEI by Population			VEI by Fleet			EDGAR		
			NMB	NME	r	NMB	NME	r	NMB	NME	r
MABH	Cidade Industrial	31	**-18.40**	**23.70**	**0.49**	**-17.96**	**23.03**	**0.48**	**29.00**	**32.64**	**0.47**
	Delegacia Amazonas	30	71.94	72.13	**0.69**	73.86	73.90	**0.69**	173.99	173.99	**0.67**
MAGV	RAMQAr 4	30	38.58	77.49	-0.07	37.22	76.19	-0.04	83.81	119.36	-0.05
	RAMQAr 6	31	**-4.42**	**37.23**	0.28	**-3.76**	**38.58**	0.31	**23.07**	53.53	0.34
MARJ	Irajá	31	**3.30**	**27.0**	**0.84**	**5.60**	**26.40**	**0.85**	64.60	64.60	**0.85**
MASP	CID universitária USP IPEN	31	144.93	144.93	**0.72**	149.63	149.63	**0.71**	362.86	362.86	**0.71**
	Congonhas	31	69.70	70.80	**0.69**	73.77	74.42	**0.69**	220.75	220.75	**0.67**
	Guarulhos Pimentas	31	-38.45	**40.75**	**0.50**	-38.09	**40.37**	**0.49**	**-7.70**	**26.50**	**0.48**
	Ibirapuera	31	98.98	98.98	**0.82**	103.75	103.75	**0.82**	276.10	276.10	**0.81**
	Itaim Paulista	31	**-27.8**	**31.20**	**0.74**	**-26.00**	**30.08**	**0.74**	**19.20**	**29.40**	**0.75**
	Marginal Tietê Pte Remédios	31	38.70	**45.10**	**0.52**	40.79	**46.75**	**0.51**	156.60	156.60	**0.52**
	Parelheiros	31	49.84	56.53	**0.76**	49.43	55.95	**0.76**	149.74	150.07	**0.76**
	Pinheiros	31	93.67	93.67	**0.72**	97.86	97.86	**0.71**	267.96	267.96	**0.73**
	São Bernardo do Campo - Centro	31	**25.75**	**38.54**	**0.75**	**29.53**	**40.94**	**0.76**	117.42	120.92	**0.75**

Figure 4.9 brings the time series for six monitoring stations, comparing the $PM_{2.5}$ 24-hr averages modeled with observational. Overall, the modeling was capable of following the increases and decreases of $PM_{2.5}$ concentrations, as indicated by the r values in Table 6. As observed in Irajá (MARJ), for example, where the $PM_{2.5}$ 24-hr averages decreased from August 1st to 10th and increased from August 28th to 31st, and the modeling well-reproduced this variation. For most of the stations, the results with the proposed inventory were superior to EDGAR, presenting a smaller deviation from the observed concentration.

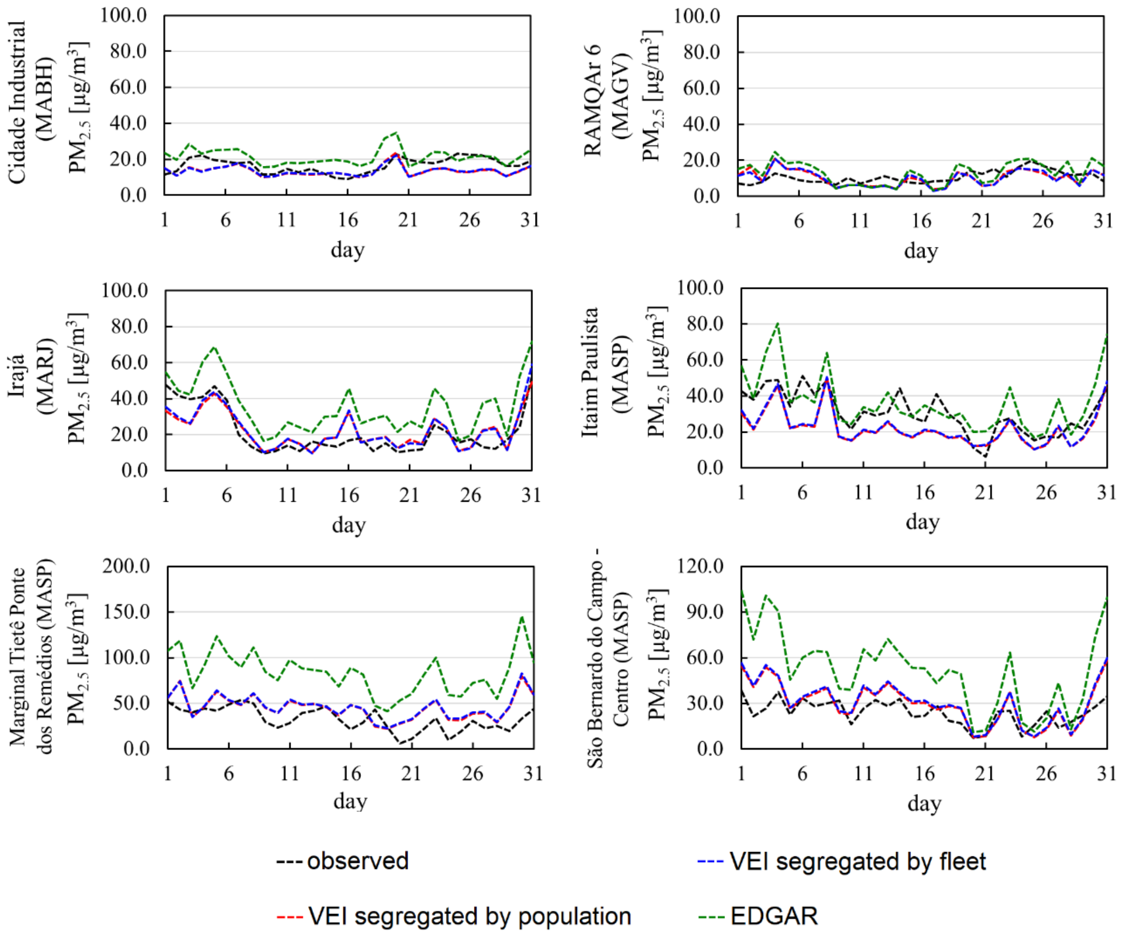

Figure 4.9 - $PM_{2.5}$ 24-hr averages during August 2015, comparing the three inventories with observational data.

4.4 Discussion

The availability of adequate emission inventories is the most critical input for the chemical transport models to predict air quality. These models use detailed emission data, including the quantification of emissions and their chemical characterization. Therefore, the quality of results obtained is directly related to the quality and detail of the atmospheric emissions inventory of the region under study. Regarding Brazil, only a few cities have detailed inventories, and the use of global inventories is often a necessity. Therefore, although the total PM from the VEI was estimated for 2012, its use for more recent years in air quality modeling is still necessary as an alternative to the global inventories. Thus, the uncertainties must be considered in the analyses. Although the number of vehicles has increased over the years in studied regions, the concentrations of primary pollutants have been reduced due to national control programs, improved fuel, and vehicle technology (ANDRADE *et al*., 2017). The period chosen for modeling (August 2015) is related with available observational data in all four metropolitan areas, and allows to verify the fact that if a past inventory continues to be representative for subsequent years.

The percentages used to divide total PM into PM_{10} and $PM_{2.5}$ were 86.3% and 63.1%, respectively, which were based on inventories that considered PM emission from the exhaust, wear from the tire, brake, and road surface. The results obtained with this division showed to be adequate to represent the two PM fractions, with PM_{10} in MASP obtaining better results. Pacheco *et al*. (2017) showed that the difference between PM_{10} and $PM_{2.5}$ in MASP are decreasing over the years, denoting the dominance of combustion emission. These results confirm the fact that, in MASP, the majority of emissions are vehicular, and most of the industrial sources are placed out of the urban center. On the other hand, for MAGV, MABH, and MARJ, relevant industrial sources are still located in the urban centers that may contribute to increasing ambient PM concentration. Other PM sources include unpaved road and windblown dust emissions. Thus the differences found between modeling and observational PM may be related to sources that are not considered in the inventories, but they also be linked with an underestimation of vehicle PM emissions, which may be related with low emission factors, or fleet characteristics (e.g., vehicle age, fuel consumption, and scrappage curve) that are not adequately considered.

In this work, the aerosol scheme used (GOCART) has no size information for sulfate, BC, and OC, and secondary organic aerosol is not considered. Besides this limitation, this mechanism has been used successfully in PM studies (SCHWARTZ *et al*., 2012; GUERRETTE and HENZE, 2015; PENG *et al*., 2017; WERNER *et al*., 2019), and in the present study, PM concentrations obtained were reasonable.

The spatial and temporal concentrations of pollutants around buildings, in hotspots, and on roads can vary by orders of magnitude. The spatial disaggregation of mobile sources using methods such as interpolation techniques, regression analysis, cluster analysis in modeling steps is essential to improve the accuracy of air quality modeling when a bottom-up inventory is available, or traffic models and vehicle emissions models are used (PINTO *et al*., 2020b). The solutions needed for more accurate modeling are not simple, and include tools for the pre-treatment of input data, detecting outliers, correcting missing values, and estimating uncertainties. Accurate traffic data generates better results in vehicle emissions models and air quality models as a consequence.

Alonso *et al*. (2010) developed an inventory of vehicular urban emissions for South America based on analyzes and aggregations of local inventories of the largest cities in the region and correlations with socioeconomic indices. Urban emissions were also extrapolated to other cities that had no emissions inventory based on city vehicle density and mobile CO and NOx emission sources. Ibarra-Espinosa *et al*. (2020) applied 120 million real-time GPS recordings and travel demand models for South-East Brazil to estimate vehicular emissions. In the present work, the use of population and fleet indices to desegregate PM emission was found to be adequate, and the spatial distribution based on urban areas resulted in PM concentrations with reasonable statistic indices, especially when it is considered that the comparison is made between a PM concentration found in an area of 25 km^2 (5-km grid spacing) and a point (monitoring station).

Moreover, the different small characteristics of the local monitoring stations, which may contribute to altering the air quality, are not possible to represent in the modeling (e.g., Ibirapuera station in MASP is located in a park). Therefore, discrepancies in model-observation are not only attributed to inaccuracies in the emissions, but also reflect divergences in land use and land cover, and meteorological fields, especially wind direction. However, the model represented the PM variability well along the month simulated, denoted by *r* coefficients. Therefore, the present study becomes an important environmental management tool for the strategic planning of cities in the studied region since it presents regions that can be impacted by vehicular PM emissions. It also shows the necessity to expand air quality monitoring, particularly for fine particulate matter.

4.5 Conclusion

The VEI proposed in this work was found to be a satisfactory tool for air quality modeling, especially for a region that does not have relevant information on pollutant emissions from road vehicles. Additionally, other emission inventories or databases may be merged in models such as PREP-CHEM-SRC, EDGAR (other pollutants), MEGAN (biogenic), and 3BEM (biomass burning). The PM emission distribution into urban areas is simple and proved to be a practical methodology for air quality modeling in 5×5 km grid spacing. A more accurate, highly resolved, and updated vehicle emission inventories are essential to improve the accuracy of the prediction of air quality models.

Regarding the comparison with local emission inventories, the main differences between our estimates and other local inventories (e.g., Réquia Jr. *et al.*, 2015, and CETESB, 2016) may be attributed to different methodologies and input data used (e.g., type of fleet and fuel, and emissions factors) for making the estimates.

An important factor in WRF-Chem modeling is the parameterization choices to represent the physical processes that occur in the atmosphere, and that can be combined in different ways. The choice of a group of parametrizations that are most appropriate to represent the meteorological conditions such as ambient temperature, relative humidity, and wind speed/direction of a region is a primordial step in the simulation with WRF. The meteorological fields produced with the parametrizations used in this work could satisfactorily represent specific relative humidity and ambient temperature. For wind speed, the model tends to overestimate them, mainly because the grid spacing used in the model does not allow the consideration of urban morphological interferences such as buildings and trees. Therefore, the roughness represented in the model is smaller than in urban areas. Therefore, wind speeds are overestimated due to the lack of obstacles that are not considered. For wind direction, a much better agreement with observational data was found for MABH.

Comparing the emission inventories, EDGAR showed higher PM emissions in all four metropolitan areas. As expected, the use of these emissions in the air quality modeling produced higher PM concentrations than those found in monitoring stations in MASP. Our emission inventory produced relatively better statistical indices. For MABH, MAGV, and MARJ, the VEI segregated by population and fleet and EDGAR obtained NMB and

NME indices for PM concentrations in an acceptable range. It is highlighted that the air quality modeling was performed for a dry month during winter when PM concentrations are usually higher than those during summer. Therefore, a year-long simulation is recommended to evaluate the VEI also during summer. As population and fleet segregation resulted in similar PM concentrations, both segregations may be recommended for top-down emission inventories in Brazil. The proposed inventory can also be used to generate emissions for a larger domain (when nesting technique is applied) if a refined inventory for the area of interest is available.

5. QUANTIFYING THE IMPACT OF PARTICLE MATTER ON MORTALITY AND HOSPITALIZATIONS IN FOUR BRAZILIAN METROPOLITAN AREAS

5.1 Introduction

Air quality control programs are basic instruments of environmental management to protect the health and well-being of the population and improve the quality of life (SILVEIRA *et al*., 2016; SLOVIC and RIBEIRO, 2018). Considering the need of continuous air pollution control programs evaluation, it is strategic to adopt air quality standards as a complementary action and referential to the established maximum pollutants emission limits (GULIA *et al*., 2015; KUMAR *et al*., 2016; HOWARD *et al*., 2019).

In Brazil, the air quality standards were updated in 2018, by Resolution CONAMA 491/2018, considering three intermediate phases and a final standard that considers WHO guidelines (WHO, 2006) to be reached in the future. Each phase will be implemented following air emission control plans and air quality assessment reports prepared by state and district environmental agencies, but intermediate and final standards have no deadline for their implementation (FERNANDES *et al*., 2020; SICILIANO *et al*., 2020). For the first time, air quality standards for fine particle matter ($PM_{2.5}$) were established at the national level. However, the air quality monitoring in Brazil is still limited, restricted and unsatisfactory in terms of sample history, territorial coverage, several parameters monitored, and representativeness in measurements, due primarily to managerial difficulties and the low number of technicians involved, as well as the lack of resources for installing and maintaining monitoring equipment and networks (IEMA, 2014). For instance, in 2017, only 24 Brazilian cities (0.43% of all cities) monitored fine particles with 50 monitoring stations, all located in the southeastern region (ANDREÃO *et al*., 2018).

Alongside to monitoring, air quality modeling is widely used to estimate the impacts on the atmosphere caused by emissions, and also used for projects to install air quality monitoring network, being an economically viable tool. An atmospheric model is a representation of the dynamic, physical, chemical, and radiation processes in the atmosphere, described by partial differential equations. To obtain an approximate numerical solution, the equations are discretized by finite differences or finite volumes, for example, generating a system of algebraic equations, which can then be solved. In this sense, air quality models help to understand how air pollutants behave in the environment (JACOBSON, 2005; OKE *et al*., 2017), from the local to the global scale.

By modeling the weather and air quality of a region, it is possible to assess the current level of pollution (ALONSO *et al*., 2010; VARA-VELA *et al*., 2016; 2018; PEDRUZZI *et al*., 2019), track trends (ANDRADE *et al*., 2015; ZHANG *et al*., 2018), define responsibilities for air pollution levels (RING *et al*., 2018; SONG *et al*., 2019), assess the potential impact of future emission sources (COLLET *et al*., 2018; CAMPBELL *et al*., 2018), study emission reduction scenarios (WANG *et al*., 2016b; ALBUQUERQUE *et al*., 2019) and estimate the health impacts (BOLDO *et al*., 2014; DING *et al*., 2016).

Especially for estimating health impacts, some software tools for quantifying the impact of air pollution are available to the community, as BenMAP-CE (SACKS *et al*., 2018), AirQ+ (WHO, 2018b) and Aphekom (Improving Knowledge and Communication for Decision Making on Air Pollution and Health in Europe) (APHEKOM, 2011). The concept is to use epidemiological studies as the base to investigate the number of avoidable hospital admissions and deaths related to a reduction in air pollutants concentrations. This information is important, especially for decision-makers, to construct and investigate public health policies for air quality management.

Among pollutants that can cause harm to human health and loss of quality of life, particulate matter, especially fine particles, stands out as one of the major pollutants associated with all-cause, non-accidental cause, lung cancer, cardiopulmonary, and cerebrovascular mortality (POPE *et al*., 2002; KREWSKI *et al*. 2009; CROUSE *et al*. 2012; CESARONI *et al*., 2013; THURSTON *et al*., 2016; DOWNWARD *et al*., 2018; POPE *et al*., 2019a), besides to be an important contributor to the global burden of disease (COHEN *et al*., 2017) and responsible to reduced life expectancy (APTE *et al*., 2018). Additionally, short-term air pollution levels are also responsible for an increase in the number of hospitalizations (RODRIGUES *et al*., 2015; FREITAS *et al*., 2016; GOUVEIA *et al*., 2019) and they can also be correlated to daily mortality (BRAVO *et al*., 2016; GOUVEIA and JUNGER, 2018).

A previous work (ANDREÃO *et al*., 2018) estimated the excess mortality associated with fine particles for a few Brazilian cities using monitoring data. This current study aims to quantify the avoidable deaths and hospitalizations associated with a reduction in particulate matter (PM_{10} and $PM_{2.5}$) concentrations, considering the new final standards proposed in Brazilian legislation (based on WHO guidelines) for 102 cities that form the four metropolitan areas of Brazilian southeastern, encompassing important cities as São Paulo, Rio de Janeiro, Belo Horizonte, and Vitória. The four metropolitan areas correspond for 20.0% of the national population (2015), 22.6% of the fleet (2015), and almost 30% of the Brazilian GDP (2013). One-year modelling with WRF-Chem model was performed to obtain daily city levels $PM_{2.5}$ and PM_{10} concentrations for 2015 (base year). The results are important in helping to assess the spatial distribution of the air quality monitoring stations in the region, to estimate PM levels in unmonitored cities, to support policymakers to project the population health improvements, and to confirm the importance of adopting more restrictive air quality standards.

5.2 Materials and methods

5.2.1 Study area

This work focuses on metropolitan areas of the four states that compounds Brazilian southeastern: metropolitan area of São Paulo (MASP), Rio de Janeiro (MARJ), Belo Horizonte (MABH), and Great Vitória (MAGV), as shown in Figure 5.1. Table 5.1 summarizes the basic information of each metropolitan area.

Pacheco *et al*. (2017) reviewed the emissions and concentrations of particulate matter in MASP, MARJ ad MABH, discussing the representativeness of the fleet and fuel type on emissions, while Santos *et al*. (2017) and Galvão *et al.* (2019) present the main PM sources in MAGV. In common, the authors highlighted the significance of vehicular emissions in all four MA.

Biomass burning from wild and deforestation fires is another critical source that contributes to deteriorating air quality in the region (MIRANDA *et al*., 2017; ANDRADE *et al*., 2017), especially from July to October (dry season), when the urban Southeast atmosphere receives smoke plume pollution transported from the central region of Brazil and the south of the Amazon basin (FREITAS *et al*., 2005; MIRANDA *et al*., 2017). Outside of the burning season, a smaller impact from biomass burning on air quality is expected. Vara-Vela *et al*. (2018) showed that biomass burning accounted for between 8 and 24% (5 to 15 μg m^{-3}) of maximum $PM_{2.5}$ concentrations in MASP from August 19th to September 3rd 2014.

5.2.2 Health effects

Epidemiological studies usually report the concentration-response function (C-R) (the relationship between the concentration of a pollutant and the population response) or, most common, the estimation of the relative risk (RR) (a measure of the change in risk of an adverse health effect associated with an increase in a particular pollutant concentration). Therefore, it is necessary to estimate the effect to develop a functional relationship between a change in PM concentration and the number of cases avoided, since we are not interested in the C-R function itself, but in the relationship between the change in PM concentration and the corresponding change in the population health response. In this case, the log-normal formulation (Equation 2.1) (page 35) is the most used health impact function to estimate the health effects in short (daily) and long-term (annual) (SACKS *et al*., 2018), and it was used in this research.

The indicated epidemiological studies to investigate the avoidable hospitalizations are those carried out in time series, panels, and case timeline studies (EFTIM and DOMINICI, 2005; WHO, 2006). In this work, Brazilian short-term studies were used to estimate the total hospitalizations attributable to PM_{10} (FREITAS *et al*., 2016; SOUZA *et al*., 2018) and $PM_{2.5}$ (CESAR *et al*., 2013; NASCIMENTO *et al*., 2017) for respiratory diseases (ICD-10: chapter X). The few epidemiological research studies in Brazil relating $PM_{2.5}$ concentration and hospitalizations focused on children, most of them up to 10 years. This age group studied involves children less than one-year-old. However, in the present study, they were not considered, due to the uncertainties and confounding factors regarding breathing problems in newborns. Therefore, for $PM_{2.5}$, the age group evaluated ranged from one to nine years old, and for PM_{10}, above one-year-old. Regarding cardiovascular diseases, a few Brazilian studies investigated their relationship with PM. For PM_{10}, RR's given by Gouveia *et al*. (2006) for circulatory system diseases (ICD-10: chapter IX) for the elderly group were selected. For $PM_{2.5}$, Ferreira *et al*. (2016) also reported RR for circulatory system diseases for the elderly.

Table 5.1 - Main characteristics of the four metropolitan areas.

Metropolitan Area	Cities	Capital	Population (2015) [million]	Fleet (2015) [million]	Demographic density [inhab./km^2]	Geography	Main emission sources
MASP	39	São Paulo	21.1	12.3	2,714	Moderate-high plains	Transportation, industries
MARJ	22	Rio de Janeiro	12.6	4.6	1,726	Costal; Low plains	Transportation, petrochemical industries, refinery
MABH	34	Belo Horizonte	5.2	2.8	625	High plains	Transportation, mining industries, refinery
MAGV	7	Vitória	1.9	0.8	837	Costal; Low plains	Transportation, iron ore pelletizing, steel and iron industries

Cohort studies are used to study long-term environmental exposure to pollution (EFTIM and DOMINICI, 2005; WHO, 2006). Since no cohort study relating air pollution and health is available in Brazil (ANDREÃO *et al*., 2018), the number of avoidable deaths for all causes (ICD-10: A00-Y98) and lung cancer (ICD-10: C33-C34) were estimated using RR's based on the meta-analysis of 75 cohort studies linking $PM_{2.5}$ with excess mortality risk carried out by Pope *et al*. (2019b). For non-accidental causes (ICD-10: A00-R99), cardiovascular (ICD-10: I20- I79), and ischemic heart diseases (ICD-10: I20-I25), the number of avoidable deaths due to $PM_{2.5}$ was estimated using concentration-response functions based on cohort studies conducted in Europe (CESARONI *et al*., 2013) and North America (POPE *et al*., 2004; LADEN *et al*., 2006; KREWSKI *et al*., 2009; CROUSE *et al*., 2012). Table 5.2 summarizes all RR's selected.

The meta-analysis carried out by Pope *et al*. (2019b) estimated the mean of the distribution of effects of cohort studies from North America, Europe, and Asia, indicating robust $PM_{2.5}$-mortality associations, with heterogeneity in estimates between different cohorts and different analyses of the same or similar cohorts (RR's varying widely). Therefore, the RR's presented for all causes, and lung cancer, was considered appropriate to be used in Brazil. There is no single most suitable RR function that can suit our study areas. Moreover, there is a scarcity for such functions that are developed for Brazilian pollution and socioeconomic conditions. Therefore, we have used different exposure-response functions for non-accidental, cardiovascular, and ischemic heart diseases to allow a more representative mortality estimate along with a mean and a standard deviation range. The findings should, therefore, be interpreted individually by each function due to the different characteristics and methods in each cohort study, such as population size, confounder variables used, the geographic area covered, and $PM_{2.5}$ chemical compositions.

Voorhees *et al*. (2014) discuss the use of RR's from other cities or regions, considering the population, environmental, and $PM_{2.5}$ characteristics, which is a common practice when they are recognized as being of high quality and produced from well-conducted epidemiological cohort studies. Andreão *et al*. (2018) highlighted this necessity practice for Brazilian case estimation.

Table 5.2 - Selected epidemiological studies characteristics.

Health outcome	Exposure	Reference	Age range	Hazard ratio (95% CI)
Respiratory diseases	Short-term (PM_{10})	Freitas *et al*. (2016)	> 1	1.0967 (1.0764 - 1.11)
		Souza *et al*. (2017)	1 - 6	1.075 (1.001 - 1.092) [a]
Circulatory System Diseases		Gouveia *et al.* (2006)	≥ 65	1.010 (1.005 - 1.015)
Respiratory diseases	Short-term ($PM_{2.5}$)	Nascimento *et al*. (2017)	1 - 9	1.0382 (0.99 - 1.089) [b]
		César *et al*. (2013)	1 - 9	1.008 (1.001 - 1.016)
Circulatory System Diseases		Ferreira *et al*. (2016)	≥ 65	1.196 (1.064 - 1.346)
All Causes	Long-term ($PM_{2.5}$)	Pope *et al*. (2019b)	≥ 30	1.08 (1.06 - 1.11)
Non-Accidental		Cesaroni *et al.* (2013)	≥ 30	1.04 (1.03 - 1.05)
		Crouse *et al.* (2012)	≥ 25	1.15 (1.13 - 1.16)
Cardiovascular		Laden *et al.* (2006)	25 - 74	1.28 (1.13 - 1.44)
		Cesaroni *et al.* (2013)	≥ 30	1.06 (1.04 - 1.08)
		Crouse *et al.* (2012)	≥ 25	1.16 (1.13 - 1.18)
Ischemic Heart Disease		Pope *et al.* (2004)	30 -99	1.18 (1.14 - 1.23)
		Krewski *et al.* (2009)	30 - 99	1.15 (1.11 - 1.20)
		Cesaroni *et al.* (2013)	≥ 30	1.10 (1.06 - 1.13)
		Crouse *et al.* (2012)	≥ 25	1.31 (1.27 - 1.35)
Lung Cancer		Pope *et al.* (2019b)	≥ 30	1.13 (1.07 - 1.20)

[a] For 10.49 $\mu g\ m^{-3}$ of increment in PM_{10} concentration; [b] For 4.25 $\mu g\ m^{-3}$ of increment in $PM_{2.5}$ concentration. The other hazard ratios consider 10 $\mu g\ m^{-3}$ of increment in PM concentrations.

The baseline incidence rate (hospitalizations and deaths), and the population data were obtained from *Departamento de Informática do Sistema Único de Saúde* (DATASUS) for each city for 2015, and they are presented in the Appendix II. The number of hospitalizations is summarized by month in DATASUS. Therefore, for each month, it was considered a constant daily number of hospitalizations. The age structure follows the epidemiological studies selected (Table 5.2).

5.2.3 Air quality modeling

One-year modeling for the year 2015 was performed with WRF-Chem (GRELL *et al.*, 2005) version 3.9.1.1 for each metropolitan area to obtain daily PM concentrations. As shown in Figure 5.1, one larger domain (D01) covered all four MA, with 25 km grid spacing and 79 × 69 grid-cells in longitudinal and latitudinal directions, respectively, resulting in a domain of 1,975 km × 1,725 km. For each metropolitan area, a second domain was created considering a grid spacing of 5 km, and 51 grid-cells in both longitudinal and latitudinal extensions, resulting in domains with 255 × 255 km. The vertical structure was represented with 32 levels, refined closer to the ground. Meteorological initial and boundary conditions were obtained from the National Center for Environmental Prediction (NCEP) Global Forecast System (GFS) final (FNL) with six-hour time resolution, 26 vertical levels, and a horizontal resolution of 1.00 × 1.00 degrees (January to July) and 0.25 × 0.25 degrees (August to December, when a higher horizontal resolution was available).The main physics options used (Table 5.3) were based on Vara-Vela *et al*. (2018) and Andreão *et al*. (2020a), studies carried out for the region. No meteorological nudging was used, and land use/cover was based on MODIS 20 classes 2001-2010.

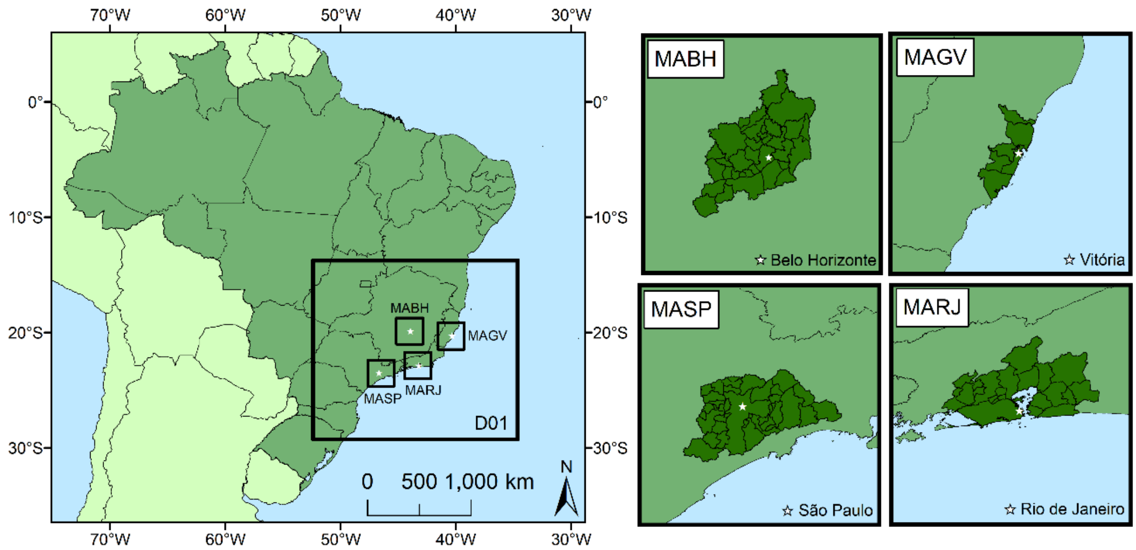

Figure 5.1 - Modeling domains, highlighting the four metropolitan areas studied.

Table 5.3 - WRF-Chem configurations.

Attributes	WRF-Chem option
Radiation	Longwave and shortwave RRTMG scheme (IACONO *et al*., 2008)
Surface layer	Revised Mesoscale Model version 5 Monin-Obukhov scheme (JIMÉNEZ *et al*., 2012)
Land surface	Unified Noah land surface model (CHEN and DUDHIA, 2001)
Boundary layer	Yonsei University scheme (HONG *et al*., 2006)
Cumulus clouds	Multiscale Kain-Fritsch scheme (ZHENG *et al*., 2016)
Cloud microphysics	Morrison two-moment (MORRISON *et al*., 2009)
Gas phase	Regional Acid Deposition Model version 2 (RADM2) (CHANG *et al*., 1989)
Aerosol	Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport model (GOCART) (CHIN *et al*., 2000)
Photolysis	Fast Troposphere Ultraviolet-Visible (TIE *et al*., 2003)

The emission inventory was prepared in PREP-CHEM-SRC (FREITAS *et al*., 2011) version 1.5. Particulate emissions are based on vehicular emission available in the 2nd Top-Down Brazilian National Inventory of Atmospheric Emissions by Road Motor Vehicles of 2012 (MMA, 2013). The 63,000 t/year of PM from close to 49 million vehicles was spatially distributed in the urban areas of each Brazilian city considering the fleet of each city. The emissions were then summarized in a .txt file at a 1 km grid resolution and compiled in PREP-CHEM-SRC. A complete description of the applicability of this inventory and its validation is described by Andreão *et al*. (2020a).

Global inventories available for PREP-CHEM-SRC were used for other anthropogenic chemical species (EDGAR 2010), biogenic (MEGAN 2002), aerosol background (GOCART 2006), and biomass burning emissions (3BEM 2015), which considers each fire pixel from wild and deforestation fires detected by remote sensing. All inventories used in PREP-CHEM-SRC are described by Freitas et al. (2011). To be used in WRF-Chem, the emission files prepared with PREP-CHEM-SRC in binary format were converted to the required format using the *convert_emiss* tool of WRF-Chem v. 3.6. Emissions generated by PREP-CHEM-SRC are made for RADM2 chemical mechanism and GOCART speciation (ARCHER-NICHOLLS *et al*., 2015).

The GOCART scheme includes 14 defined aerosol species (sulfate; hydrophobic and hydrophilic organic carbon; hydrophobic and hydrophilic elemental carbon; dust in five particle sizes: 0.5, 1.4, 2.4, 4.5, and 8.0 μm, and sea salt in four particle size distributions: 0.3, 1.0, 3.25, and 7.5 μm), and an unspecified aerosol contributions variable (PENG *et al*., 2017; ANDREÃO *et al*., 2020).

Output from the global Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4)/Goddard Earth Observing System Model, version 5 (GEOS-5) (EMMONS *et al*. 2010), was used to create initial and lateral-boundary chemical conditions, according to Gavidia-Calderón *et al*. (2018). The initial and lateral boundary conditions for 5-km grid runs were created applying one-way nesting technique from the 25-km grid run, using *ndown* tool from WRF-Chem. The simulations were performed monthly, considering +10 days of spin-up for each month (Hogrefe *et al*., 2017).

For modeling validation, hourly meteorological parameters (temperature, specific humidity, wind speed, and wind direction) were compared to 32 automatic meteorological monitoring stations of the National Institute of Meteorology in Brazil (INMET) (Appendix II - Figure SII8), using the statistical indices and benchmarks suggested by Emery *et al*. (2001): Mean Bias (MB); Mean Error (ME); Root Mean Square Error (RMSE); and Index of Agreement (IOA). The circular correlation coefficient (CCC) was also calculated for wind direction. To evaluate the performance of the meteorological model simulations, the resulting criteria for complex conditions suggested by Ramboll (2018) and LADCO and WDNR (2018) were used. For those benchmarks that did not present criteria for complex conditions, the criteria for simple conditions suggested by Emery *et al*. (2001) were used instead.

Daily mean PM_{10} and $PM_{2.5}$ concentrations were compared to environmental measures from 61 (PM_{10}) and 18 ($PM_{2.5}$) urban air quality monitoring stations, respectively, of the four states environmental agencies: *Companhia Ambiental do Estado de São Paulo* (CESTEB-SP), *Fundação Estadual do Meio Ambiente* (FEAM-MG), *Instituto Estadual do Ambiente* (INEA-RJ), and *Instituto Estadual de Meio Ambiente e Recursos Hídricos* (IEMA-ES). The statistical indices suggested by Simon et al. (2012) and Emery et al. (2017) were used: Normalized Mean Bias (NMB), Normalized Mean Error (NME), and correlation coefficient (*r*). Each state uses different measurement techniques to quantify the PM: gravimetric in São Paulo (virtual impaction – dichotomous; or impaction and cyclone), Large Volume Samplers in Rio de Janeiro, Tapered Element Oscillating Microbalance measurement methodology in Espírito Santo, and monitor with Beta radiation in Minas Gerais (ANDREÃO *et al*., 2018).

For health impact assessment, 24-h modelled PM_{10} and $PM_{2.5}$ concentrations fields were averaged for each city area, with Visual Environment for Rich Data Interpretation (VERDI) version 1.5. Therefore, a single daily average was obtained to represent each city, since the total number of hospitalizations and deaths are in city level in DATASUS, resulting, therefore, in the baseline scenario. The control scenarios considered the standards of Resolution CONAMA 491/2018, which final standard is based on WHO guidelines (WHO, 2006): a maximum daily concentration of 50 $\mu g\ m^{-3}$ for PM_{10} and 25 $\mu g\ m^{-3}$ for $PM_{2.5}$, and a maximum annual concentration of 10 $\mu g\ m^{-3}m^{3}$ for $PM_{2.5}$. Therefore, the health gains will only be observed and calculated for the cities with daily PM_{10} and $PM_{2.5}$ concentrations and annual $PM_{2.5}$ concentrations higher than the standards. Results for intermediate standards (temporary values to be reached in stages) are also presented.

5.3 Results

The statistical indices and benchmarks obtained from the comparison between modelled and monitored meteorological parameters, and comparison for precipitation is presented in the Appendix II.

5.3.1 PM modeling evaluation against monitoring data

Figure 5.2 shows the NMB and NME values obtained by comparing modelled PM_{10} concentrations (24-h averaging) with monitored data. Figure 5.3 presents the PM_{10} correlation coefficient found. Criteria and goals benchmarks found in literature represent a direct comparison with statistic indices obtained by past U.S. modeling studies. For example, criteria values indicate statistical values that about two-thirds of past studies in the U.S. have achieved. Goal benchmarks indicate those that the third of top-performing past modeling studies have met. Therefore, these two benchmarks should be viewed as the best a model can be expected to achieve (EMERY *et al*., 2017).

For MABH, the mean NMB value was –37.9 (quite below the criteria value of –30.0), and the NME was 42.2 (within the criteria value of 50.0). The *r* values attained the criteria benchmark (>0.4). For MAGV, similar results were obtained. The mean NMB was –37.9, and the mean NME was 44.5. The *r* values varied from 0.04 to 0.37. MARJ presented better benchmarks for NMB (mean value: –32.9). The mean NME was 43.6, with only seven of the 23 monitoring stations presenting NME values above the criteria. Almost half of the monitoring stations presented *r* values within the criteria benchmark. For MASP, a larger variability of NMB and NME values was obtained, with stations reaching the goal benchmarks (±10.0 for NMB and <30.0 for NME), and others with values much above the criteria. The mean NMB and NME values were 36.3 and 54.7, respectively. About 75% of the stations presented *r* values within the criteria benchmark, with a median value equal to 0.53.

Represent all monitoring sites concentrations is a difficult task, especially considering the existence of local interferences that the modeling cannot represent or capture, and lack of observed monitoring data for some periods. Therefore, the macro view representation of the concentration fields is more appropriate. Hence, as some of the monitoring stations reached the benchmarks, including the goal, it can be intended a suitable representation of PM concentrations for the region.

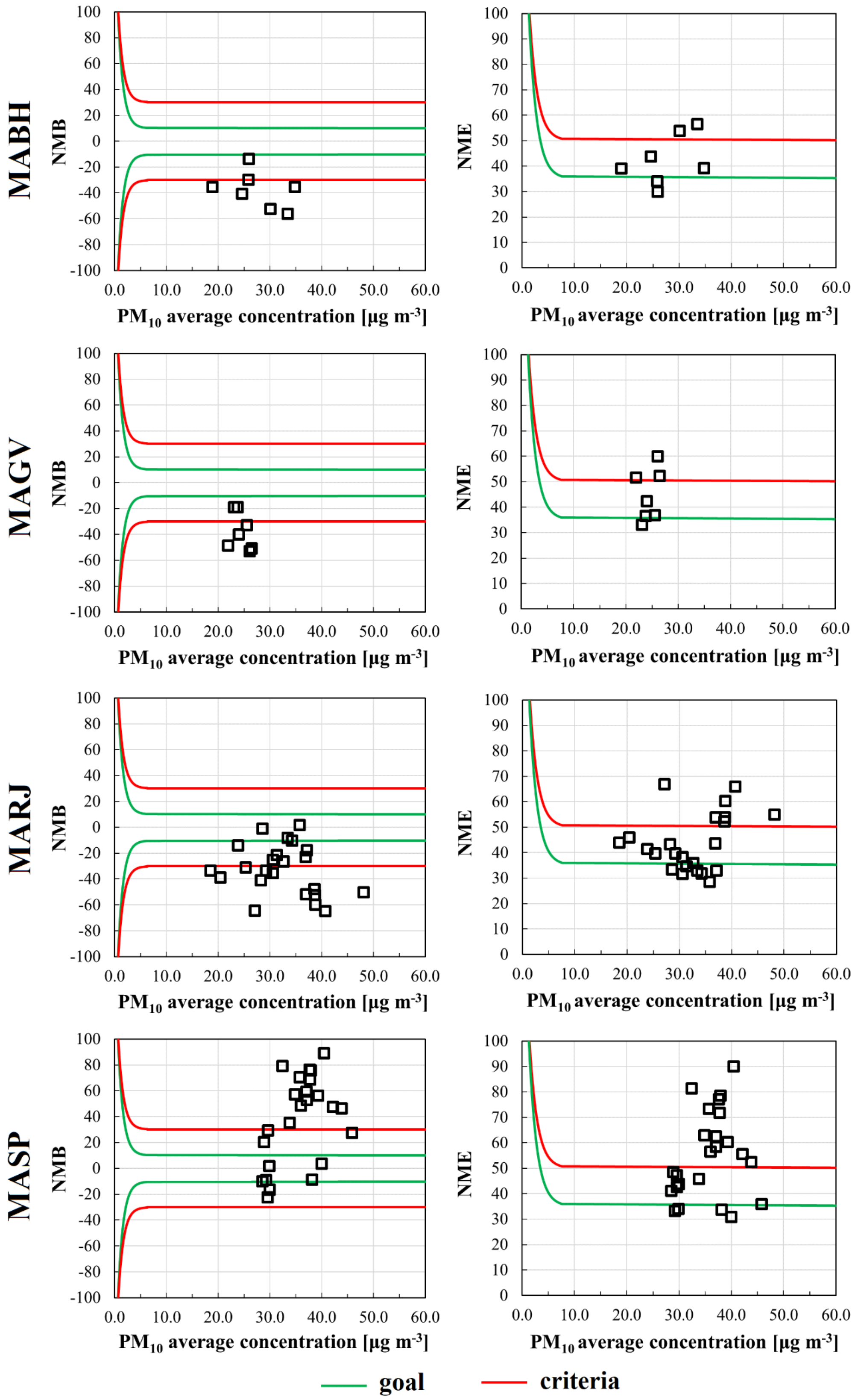

Figure 5.2 - NMB and NME for PM_{10} 24-hr averages for MABH, MAGV, MARJ and MASP, The NMB goal and criteria considered < ±10% and < ±30%, respectively, while the NME goal and criteria considered < 35% and < 50%, respectively.

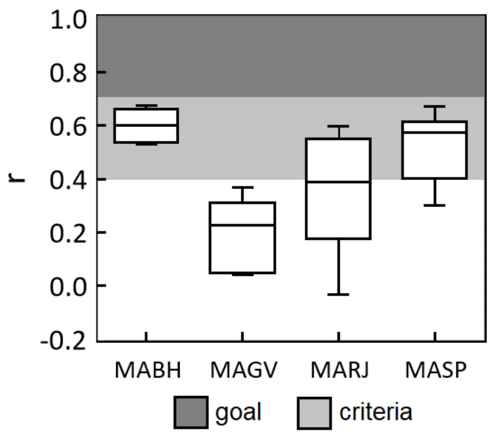

Figure 5.3 - Correlation coefficient for PM_{10}.

Figure 5.4 shows the annual modelled $PM_{2.5}$ obtained in each grid cell and the measurements at each monitoring station, while Table 5.4 presents the benchmarks reached for daily $PM_{2.5}$ comparisons with monitoring data. Here is highlighted the quantitative lack of data for most of the year for some stations in MABH. There are two stations in MAGV that monitor $PM_{2.5}$ while only one in MARJ. In most of the stations, the modelled daily $PM_{2.5}$ concentrations were higher than the monitored value. MASP presented a large variability for NMB and NME values. The *r* coefficient was satisfactory for most stations, with 78% reaching the criteria benchmark (>0.4). The higher PM concentrations found to west and northwest of São Paulo may be related to: (*i*) higher emission in this route, which includes significant traffic and encompasses middle cities as Campinas and Sorocaba; (*ii*) the southeast wind direction modelled (Appendix II - Figure SII13); and (*iii*) local characteristics not represented by the modeling, which may influence the monitoring stations, such as urban morphological interferences (buildings and trees).

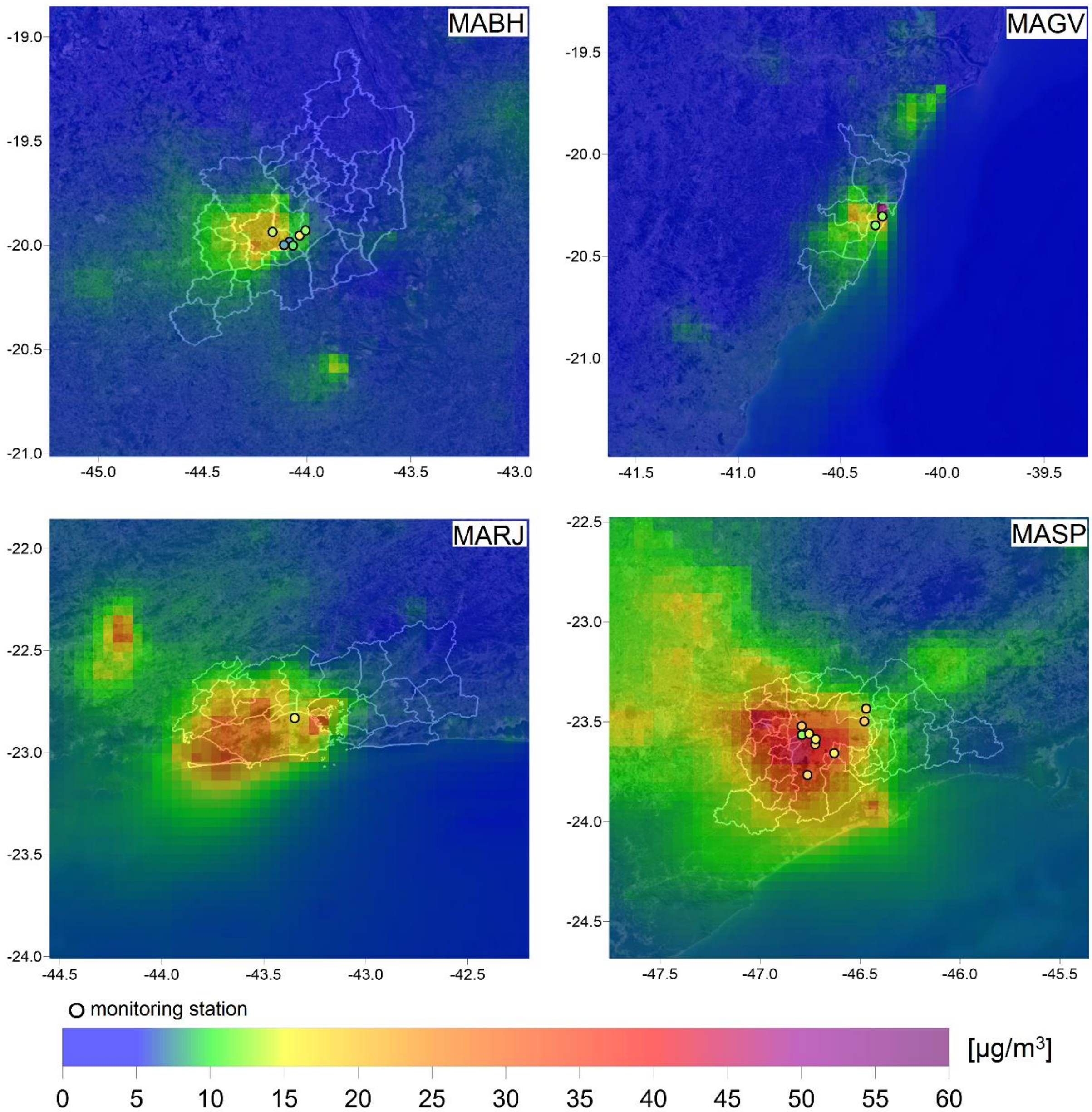

Figure 5.4 - Modelled and monitored $PM_{2.5}$ annual concentrations for MABH, MAGV, MASP, and MARJ.

Table 5.4 - Comparison between modeled and monitored daily $PM_{2.5}$ concentration. Stations that reached the criteria benchmarks suggested by Emery *et al*. (2017) (NMB < ± 30%; NME < 50%; and r > 0.40) are highlighted in bold.

MA	Monitoring station	number of 24-h $PM_{2.5}$ averages (maximum of 365)	NMB	NME	r
MABH	Alterosa	34	47.1	**47.4**	**0.47**
	Cascata	50	137.7	137.7	**0.61**
	Cidade Industrial	249	**-12.3**	**27.3**	**0.56**
	Delegacia Amazonas	208	41.9	53.5	**0.65**
	Petrovale	55	82.1	89.5	0.17
	Piratininga	56	**2.3**	**25.5**	0.32
MAGV	RAMQAr 4	347	**1.8**	**37.0**	0.23
	RAMQAr 6	361	**7.7**	**37.2**	0.30
MARJ	Irajá	363	**29.2**	**47.0**	**0.62**
MASP	CID universitária USP IPEN	364	266.4	266.4	**0.64**
	Congonhas	352	101.2	103.5	**0.56**
	Guarulhos Pimentas	187	**-2.5**	**38.3**	**0.47**
	Ibirapuera	320	131.4	132.4	**0.57**
	Itaim Paulista	172	**-2.0**	**36.6**	**0.60**
	Marginal Tietê Pte Remédios	356	99.5	100.7	**0.62**
	Parelheiros	336	52.1	59.8	**0.72**
	Pinheiros	278	162.7	163.2	**0.65**
	São Bernardo do Campo - Centro	328	**28.5**	50.0	**0.49**

Figure 5.5 shows time series plots for some stations, where it can be seen that the model was able to represent $PM_{2.5}$ concentrations increases and decreases throughout the year. Higher $PM_{2.5}$ concentrations were observed in cities located in São Paulo and Rio de Janeiro state. In Belo Horizonte, a 24-h $PM_{2.5}$ average exceeds 50 µg m^{-3} in October 2015, probably due to biomass burning. In MAGV, the seasonality observed in the other cities was not registered. Once again, the lack of monitoring data in MABH is remarkable.

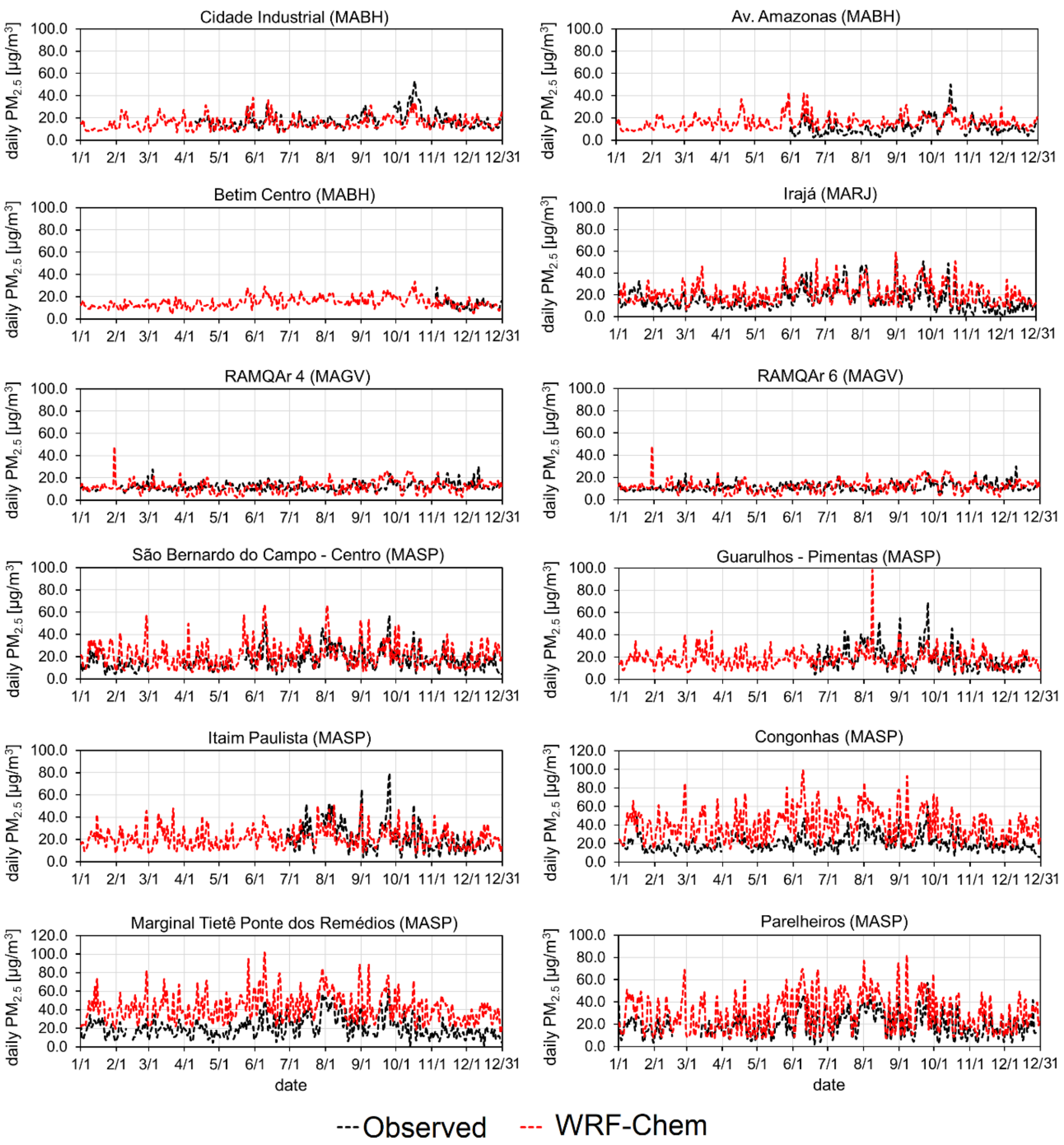

Figure 5.5 - Time series for daily $PM_{2.5}$ concentration in 2015.

5.3.2 Avoided premature mortality for annual $PM_{2.5}$ mean concentrations

Among the 102 cities analyzed, 67 presented annual $PM_{2.5}$ mean concentrations above WHO guideline (10 μg m^{-3}): 13 in MABH (38%), 4 in MAGV (57%), 15 in MARJ (68%), and 35 in MASP (90%). The total number of all-cause avoidable premature deaths in these cities was 32,000±5,300 (using Pope *et al*. (2019b) meta-analysis RR), 6.5% of all-cause deaths. For non-accidental causes, the maximum avoidable mortality is higher, due to the higher RR presented in the cohort studies of Crouse *et al*. (2012), compared to the RR of all-causes.

For cardiovascular diseases, a small variability was found for the avoidable deaths calculated based on Crouse *et al*. (2012) and Laden *et al*. (2006), 16,700±1,500 and 13,600±4,300, respectively. Using the RR provided by Cesaroni *et al*. (2013), the avoidable deaths are smaller, 7,300±1,300, which represents around 46% of the non-accidental avoidable deaths estimated using the same cohort study. For IHD, a subgroup of cardiovascular diseases, the higher number of avoidable deaths was given using Crouse *et al*. (2012) RR, 10,900±900. This number represented 65.3% of the avoidable cardiovascular deaths. A similar estimation is possible with Cesaroni *et al*. (2013) RR, where 64.8% of avoidable cardiovascular deaths may be attributed to IHD, which shows a good agreement between the studies. Finally, for lung cancer, the avoidable deaths were 1,200±300 (using Pope *et al*. (2019b) meta-analysis RR), which correspond to 4% of all-causes deaths.

As expected, the capitals with a larger population presented higher numbers of avoidable deaths. Table 5.5 summarizes the avoidable mortalities for all causes, non-accidental causes, cardiovascular, IHD, and lung cancer for the four-state capitals of Brazilian Southeast states per 100,000 inhabitants. On this basis, São Paulo presented the major avoidable deaths, but with results close to Rio de Janeiro. Although Vitória has 14% of the population of Belo Horizonte (above 25 years old), the avoidable deaths by 100,000 inhabitants were almost three times higher than Belo Horizonte, which may be linked to a higher incident rate (ANDREÃO *et al*., 2018).

Table 5.5 - Estimation of avoidable deaths for Southeast state capitals in 2015 by 100,000 inhabitants, with the standard deviation in parentheses.

Health outcome	**Reference (RR used)**	**Belo Horizonte**	**Vitória**	**Rio de Janeiro**	**São Paulo**
All Causes	Pope *et al*. (2019b)	17 (3)	46 (7)	177 (29)	181 (30)
Non-Accidental	Cesaroni *et al*. (2013)	8 (1)	22 (3)	88 (11)	92 (12)
	Crouse *et al*. (2012)	29 (1)	74 (4)	286 (16)	286 (17)
Cardiovascular	Cesaroni *et al*. (2013)	3 (<1)	9 (1)	36 (6)	43 (8)
	Crouse *et al*. (2012)	7 (1)	21 (2)	84 (7)	97 (9)
	Laden *et al*. (2006)	5 (1)	13 (4)	63 (19)	68 (22)
Ischemic Heart Disease	Pope *et al*. (2004)	2 (<1)	9 (1)	34 (5)	45 (7)
	Krewski *et al*. (2009)	4 (<1)	8 (1)	30 (5)	40 (7)
	Crouse *et al*. (2012)	1 (<1)	15 (1)	51 (4)	65 (5)
	Cesaroni *et al*. (2013)	1 (<1)	5 (1)	21 (4)	29 (6)
Lung Cancer	Pope *et al*. (2019b)	1 (<1)	2 (1)	7 (2)	7 (2)

Table 5.6 shows the total avoidable deaths for all causes and lung cancer for each metropolitan area according to their $PM_{2.5}$ annual intermediate standard (IS) and final standard (FS) of CONAMA Resolution 491/2018: IS-1: 20 μg m^{-3}; IS-2: 17 μg m^{-3}; IS-3: 15 μg m^{-3}; FS: 10 μg m^{-3}. In MAGV, the health benefits, in terms of avoidable mortality, were observed after IS-2. Reducing the standard from IS-1 to FS, the avoidable mortality was more than eight times higher in MAGV, the double in MARJ, and 1.5 times higher in MASP, which was associated with more cities presenting concentrations above FS and a higher PM concentration change from baseline to control scenario (ΔPM).

Table 5.6 - Estimation of total all causes and lung cancer avoidable deaths for Southeast metropolitan areas in 2015, with the standard deviation in parentheses.

Standard	**Health outcome**	**Reference (RR used)**	**MABH**	**MAGV**	**MARJ**	**MASP**
IS-1 (20 μg m^{-3})	All Causes	Pope et al. (2019b)	100 (20)	0 (0)	5,100 (800)	13,500 (2,200)
	Lung Cancer		3 (1)	0 (0)	190 (50)	550 (150)
IS-2 (17 μg m^{-3})	All Causes		200 (30)	15 (2)	6,550 (1,050)	15,700 (2,600)
	Lung Cancer		6 (1)	1 (0)	240 (60)	635 (170)
IS-3 (15 μg m^{-3})	All Causes		290 (45)	40 (5)	7,500 (1,200)	17,100 (2,800)
	Lung Cancer		8 (2)	2 (1)	270 (70)	690 (185)
FS (10 μg m^{-3})	All Causes		840 (130)	170 (30)	10,300 (1,700)	20,700 (3,500)
	Lung Cancer		27 (7)	8 (2)	360 (100)	820 (230)

5.3.3 Avoided premature morbidity for daily $PM_{2.5}$ and PM_{10} mean concentrations

Table 5.7 shows the total avoidable hospital admissions estimated for respiratory diseases, considering the RR's of the four Brazilian epidemiological studies, and according to daily standards from CONAMA Resolution 491/2018. For PM_{10}: IS-1: 120 μg m^{-3}; IS-2: 100 μg m^{-3}; IS-3: 75 μg m^{-3}; FS: 50 μg m^{-3}. For $PM_{2.5}$: IS-1: 60 μg m^{-3}; IS-2: 60 μg m^{-3}; IS-3: 37 μg m^{-3}; FS: 25 μg m^{-3} A total of 66 cities (65%) exceed the FS for daily $PM_{2.5}$ concentrations. For PM_{10}, 46 cities (45%) exceed the FS, 31 of them in MASP.

For MABH, most of the avoidable admissions were obtained between October 12th and 18th. Daily PM_{10} concentrations were above 50 μg m^{-3} in most of the cities. Considering the maximum $PM_{2.5}$ daily concentration of 25 μg m^{-3} (WHO guideline), the avoidable hospitalizations of children may reach 70±45. The capital (Belo Horizonte) was responsible for 48% of total $PM_{2.5}$ hospital admissions in this age group. In MAGV, only two avoidable hospitalizations were attributed to PM_{10}, with SD <1. For $PM_{2.5}$, 6±4 avoidable hospitalizations were estimated, being 3±2 for Vitória, most of them during one week in October.

In MARJ, 220±20 hospital admissions were estimated for PM_{10}, with the capital, Rio de Janeiro, contributing for the major part, 75%, or 160±15 hospitalizations. For $PM_{2.5}$, 330±220 avoidable hospitalizations were estimated for the children group, with 200±130 in Rio de Janeiro. In MASP, 4,340±430 avoidable hospitalizations were associated with PM_{10}, with São Paulo presenting around 56% of them (2,250±230). For $PM_{2.5}$, 3,020±2,070 children hospital admissions were estimated, with São Paulo accounting for 60% of them (1,810±1,230). Considering the age group from 1 to 5 years old, 18,500 hospitalization occurred in MASP in 2015 due to respiratory diseases. Therefore, the number estimated here showed that around 4.7% of them would be avoided if the maximum daily PM_{10} concentration was 50 μg m^{-3}. The lower avoidable hospital admissions are associated with lower PM 24-h concentrations in some cities, lower incident rate, and small RR's.

The avoidable hospitalizations related to $PM_{2.5}$, considering the RR from Cesar *et al.* (2013), were lower than those reported by Nascimento *et al.* (2017). The ecological time-series study of Cesar *et al*. (2013) was conducted in Piracicaba (São Paulo state), for a period between August 1st, 2011 and July 31st, 2012. Daily levels of $PM_{2.5}$ were obtained from air quality modeling with the CATT-BRAMS model. On the other hand, the study of Nascimento *et al*. (2017) was conducted in MAGV during winter (June 21st to September 21st, 2013) and summer period (December 21st, 2013 to March 19th, 2014), using monitoring data from a field campaign (portable particle samplers).

Although Souza *et al*. (2017) and Freitas *et al*. (2016) have different RR's, most of the avoidable hospitalizations may be attributed to the children's group. In this case, less than five years old.

For circulatory system diseases, the avoidable hospitalizations in the elderly, presented in Table 5.8, show that a decrease in $PM_{2.5}$ concentrations could avoid 9,850±3,950 hospitalizations in the four MA. PM_{10} caused in total 330±85 avoidable hospitalizations, showing that, as expected, $PM_{2.5}$ has a stronger effect in the circulatory system than PM_{10}. As for respiratory diseases, the capitals obtained the highest percentage of these numbers.

5.4 Discussion

The main uncertainties in air quality modeling are brought by three factors: the adequate meteorological representation, and emission inventory used, and the chemistry. Besides the difficulties of representing complex terrains in mesoscale meteorological modeling, the results are also a function of the grid space, long-time simulation, land use, and land cover, and physical parameterizations used. Another important reason for poor representation is just the limitation of mesoscale models in representing the 3-D urban structure. In this work, the results obtained from the meteorological modeling showed to be adequate to represent the metropolitan areas, although some meteorological surface stations presented indices out of the benchmarks. The simulation was performed monthly to diminish the inherent errors when a long time is simulated (loss of initial meteorological conditions dependence with time). Moreover, the parametrizations used were already tested for the region in other researches.

One limitation found is the chemistry aerosol module (GOCART), which does not support in WRF-Chem secondary organic aerosol (SOA) formation, wet scavenging, and specific ammonium-nitrate speciation.

Table 5.7 - Estimation of total hospital admissions due to respiratory diseases in the Brazilian Southeast metropolitan areas in 2015, with the standard deviation in parentheses.

Pollutant	Standard	Reference (RR used)	Age group	MABH	MAGV	MARJ	MASP
PM_{10}	IS-1 (120 μg m^{-3})	Freitas et al. (2016)	>1	0 (0)	0 (0)	0 (0)	160 (15)
		Souza et al. (2017)	1-5	0 (0)	0 (0)	0 (0)	35 (10)
	IS-2 (100 μg m^{-3})	Freitas et al. (2016)	>1	0 (0)	0 (0)	< 1 (< 1)	400 (40)
		Souza et al. (2017)	1-5	0 (0)	0 (0)	0 (0)	85 (30)
	IS-3 (75 μg m^{-3})	Freitas et al. (2016)	>1	< 1 (< 1)	0 (0)	15 (1)	1,230 (125)
		Souza et al. (2017)	1-5	0 (0)	0 (0)	3 (1)	255 (85)
	FS (50 μg m^{-3})	Freitas et al. (2016)	>1	13 (1)	2 (<1)	220 (20)	4,340 (430)
		Souza et al. (2017)	1-5	2 (1)	<1 (<1)	40 (15)	870 (290)
$PM_{2.5}$	IS-1 (60 μg m^{-3})	Nascimento et al. (2017)	1-9	1 (1)	0 (0)	7 (4)	535 (365)
		Cesar et al. (2013)		0 (0)	0 (0)	< 1 (< 1)	55 (25)
	IS-2 (50 μg m^{-3})	Nascimento et al. (2017)		4 (3)	<1 (<1)	25 (15)	950 (650)
		Cesar et al. (2013)		<1 (<1)	0 (0)	2 (1)	100 (50)
	IS-3 (37 μg m^{-3})	Nascimento et al. (2017)		20 (10)	2 (1)	100 (70)	1,825 (1,245)
		Cesar et al. (2013)		2 (1)	<1 (<1)	10 (5)	190 (90)
	FS (25 μg m^{-3})	Nascimento et al. (2017)		80 (50)	6 (4)	330 (220)	3,020 (2,070)
		Cesar et al. (2013)		10 (5)	1 (<1)	30 (15)	320 (150)

Table 5.8 - Estimation of total hospital admissions in elderly due circulatory system diseases in the Brazilian Southeast metropolitan areas in 2015, with the standard deviation in parentheses.

Pollutant	Standard	Reference (RR used)	Age group	MABH	MAGV	MARJ	MASP
PM_{10}	IS-1 (120 μg m^{-3})	Gouveia et al. (2016)	≥ 65	0 (0)	0 (0)	0 (0)	9 (2)
	IS-2 (100 μg m^{-3})			0 (0)	0 (0)	0 (0)	25 (5)
	IS-3 (75 μg m^{-3})			0 (0)	0 (0)	1 (<1)	80 (20)
	FS (50 μg m^{-3})			1 (<1)	<1 (<1)	16 (4)	310 (80)
$PM_{2.5}$	IS-1 (60 μg m^{-3})	Ferreira et al. (2016)		5 (2)	1 (0)	15 (5)	1,270 (50)
	IS-2 (50 μg m^{-3})			15 (5)	3 (1)	70 (25)	2,475 (970)
	IS-3 (37 μg m^{-3})			55 (20)	10 (5)	375 (135)	4,975 (1,980)
	FS (25 μg m^{-3})			240 (90)	35 (15)	1,180 (440)	8,390 (3,400)

The comparison against PM monitoring data showed that, for some sites, the modeling underestimated PM_{10} concentrations, mainly in MABH and MAGV, which may be an indication that local industrial sources may also contribute to air quality (SANTOS *et al*., 2017; KAWASHIMA *et al*., 2020), or SOA formation plays an important role (GALVÃO *et al*., 2019). In these cases, the avoidable deaths and hospitalizations may be higher than those presented. In MARJ, PM concentrations were better represented, with some station still presenting lower modelled PM_{10} concentrations than the observed. In MASP, part of the PM concentrations was overestimated. The annual $PM_{2.5}$ mean modelled in this work in São Paulo (city average) was 39 μg m^{-3}. Considering the average among all monitoring stations, Andreão et al. (2018) estimated an annual $PM_{2.5}$ concentration of 18 μg m^{-3} for 2015. Here three hypotheses are raised: (*i*) the modeling is overestimating $PM_{2.5}$ concentrations; (*ii*) the monitoring stations are not placed in the neighborhoods with higher PM concentrations (low PM exposure), which may contribute to underestimating the city-level concentration; or (*iii*) a combination of both (*i*) and (*ii*). In Rio de Janeiro, this comparison is more uncertain, since just one site monitors $PM_{2.5}$ automatically. The estimated annual $PM_{2.5}$ concentration in this work was about 31 μg m^{-3}, while Andreão *et al*. (2018), using data from the single monitoring station, found 12 μg m^{-3}. A well-distributed air quality network in a city involves planning, design, and establishment of objectives. In this sense, the air quality modeling may help to identify the areas most affected by air pollution, which will also contribute to the urban zoning.

Regarding the aerosol size distributions, as the distribution is skewed towards the smaller particles, for some monitoring stations grid cell the biases in the modelled $PM_{2.5}$ fields were greater than those in the modelled PM_{10} values, which may have resulted in an overestimation in the avoidable mortality and morbidity regarding the fine particles, especially in MASP (Figures 5.4 and 5.5), where the population is also relatively large. This overestimation may also has contributed to the higher avoidable hospitalizations due circulatory system diseases in the elderly.

Alonso *et al*. (2010) found large differences between EDGAR data and local inventories, especially in Brazilian cities, and concluded that global inventories have flaws in the detailed information of emissions at a local scale, mainly in South America cities. Therefore, PM overestimation caused by SO_2 reactions may also be possible (ALBUQUERQUE *et al*., 2019). In Brazilian metropolitan areas, especially in MASP, MARJ, and MABH, vehicular emission is the main contributor to air pollution (PACHECO *et al*., 2017; ANDRADE *et al*. 2017;

ALBUQUERQUE *et al*. 2018; GAVIDIA-CALDERÓN *et al*. 2018; POLICARPO *et al*. 2018; MIRANDA *et al*. 2018; 2019; LEIRIÃO and MIRAGLIA, 2019). The PM emission inventory used was based on the last national vehicle emission inventory, spatially distributed into the urban areas, the methodology described and validated by Andreão *et al*. (2020a). Additionally, biomass burning was included, which is known to influence air quality significantly in Brazilian regions, not only in the Amazon region (VARA-VELA *et al*., 2018). An update of the national VEI used, the consideration of CO, SO_2, and NOx from the VEI, and the inclusion of local sources may improve the results obtained here. However, local emission data is challenging to obtain for the region (PINTO *et al*., 2020a) due to its inexistence, restriction in data access, and without spatial details (KAWASHIMA *et al*., 2020).

This work identifies cities with potentially high $PM_{2.5}$ concentrations that do not have PM monitoring stations, as shown in Figure 5.4. Although São Paulo city is located around 760 m above mean sea level, the sea breeze affects the local pollutant dispersion (FREITAS *et al*., 2007; VEMADO and FILHO, 2016; BOURSCHEIDT *et al*., 2016; BENDER *et al*., 2019). Therefore, cities located west and northwest of MASP are influenced not only by their emission but also by São Paulo city emissions. A similar approach is applied in MABH, where the east wind, characteristic of the region for the most of the year (SANTOS *et al*., 2019), transports pollutants from Belo Horizonte to Contagem and Betim. Those cities in MASP and MABH may present higher PM concentrations than the capitals, as shown in Figure 5.4, which will also reflect in the health estimates. In this sense, we recommend that PM monitoring, especially fine particle monitoring, should expand to the west of São Paulo and Belo Horizonte, as in northwest of Rio de Janeiro. In MAGV, the results showed that Cariacica and Serra might exceed WHO guidelines, and therefore, $PM_{2.5}$ monitoring is also recommended in these cities.

Other uncertainties in the present work refer to the temporal (monthly) and spatial (city-level) resolution of morbidity data. The number of hospitalizations was equally distributed in days from the available monthly data, and the PM concentration fields needed to be averaged in city-level. The same approach was performed by Fernandes *et al*. (2020). Therefore, daily data could improve the estimation. Since it is expected that in days with higher pollutant concentrations, the number of hospitalizations increase (or in subsequent days), the avoidable hospitalizations would probably be higher in those days because of the higher daily incident rate. Spatial refined morbidity and mortality data within each city could also improve the estimations, where the grid cell with the highest number of avoidable hospitalizations and mortality could be identified.

Table 5.9 presents some results found in different studies for different locations. However, a direct comparison must be treated carefully since different methodologies, and initial parameters were used in each study ($PM_{2.5}$ change concentration, C-R function, incident ratio). Howard *et al*. (2019) showed the benefits in terms of health effects in adopting emission control strategies in power plants for the northeast of Brazil. Reducing PM_{10} emission from 28.15 g/kWh to 0.69 g/kWh, around 168 premature deaths and 16,257 hospital admissions would be avoided per year.

Table 5.9 - Avoidable deaths per 100,000 population per year.

Reference	Location	Outcome	Avoidable deaths
Present study	Brazilian Southeast MA (108 cities)	All-Causes	124
		Non-accidental causes	62-197
		IHD	18-42
		Lung Cancer	5
Ghude *et al*. (2016)	India	IHD	21
		Lung Cancer	1
Boldo *et al*. (2014)	Spain	All-Causes	9-16
		Non-accidental causes	10-33
		Lung Cancer	1-5
		IHD	3-6
Kihal-Talantikite *et al*. (2019)	Paris	All-Causes	22
Han *et al*. (2018)	Republic of Korea	IHD	5
		Lung Cancer	5
Maji *et al.* (2018)	338 Chinese cities	IHD	23
		Lung Cancer	7
Kheirbek *et al*. (2013)	New York City	All Causes	65

The present study reinforces the importance of a well-established goal in air quality management. However, any control strategy in a city must consider the role of its surroundings. In the studied metropolitan areas, management policies should be constructed considering all the regions, especially when considering urban and populational growth, changes in land use and land cover, different urban zones, and integrated transportation routes, for example.

Regarding the transport sector, public policies should integrate three axes to reduce pollutant emission: user, vehicle, and road. Traffic Management Strategies (TMS) involves new transportations services and sub-systems (e.g., bike-share and car-share programs) (BRAND *et al*., 2019). Accessibility, attendance frequency, reliability, security, information system to the traveler, operator behavior, vehicle operations restrictions, developing new technologies to improve fuel economy, programs to renew and improve the vehicle fleet and engine, emission regulations and infrastructure such as truck stop electrification (NOGUEIRA *et al*., 2019; VENTURA *et al*., 2019). Operating restrictions and pricing (e.g., low emission zones, parking management), truck and buses lanes, land use planning, speed management (e.g., eco-driving, traffic calming), traffic flow control (roundabouts, signal coordination, intersection design) and road pricing (PÉREZ-MARTÍNEZ *et al*., 2017). The integration of actions and public policies implemented in the transport sector contributes to vehicular emissions reduction and, consequently, improves air quality in urban centres (YU *et al*., 2019; ALBUQUERQUE *et al*., 2019). However, the evidence of the real impact of TMS implementation is still necessary to assess the effectiveness of public actions and policies in reducing vehicle emissions in urban areas (BIGAZZI and ROULEAU, 2017; PISONI *et al*., 2019), which is directly linked to human exposure and health.

Ongoing air quality management evaluation involves, but is not limited to, updating emission inventories, evaluation of emission reductions and programs for air pollution control strategies, studies of interactions among pollutants, especially when the interest is reducing concentrations of ozone and secondary particles, impacts on human health, and environmental and economic assessments. As discussed by Franco *et al.* (2019b), urban air quality management involves good practices, from the recognition of the importance of air pollution to the monitoring, planning, involvement of the community, and well-defined targets, for example. The present work endorses two other essential factors for urban air quality management: human exposure and health effects.

5.5 Conclusion

Although the Brazilian air quality standard has been updated after almost three decades, the first intermediate target established (current standard) is still permissive against WHO guidelines. For annual $PM_{2.5}$, for example, the first intermediate standard (20 μg m^{-3}) is double the WHO recommendations. The present study estimated the health gains related to $PM_{2.5}$ mortality and PM_{10} and $PM_{2.5}$ hospital admissions for 2015, showing that an improvement in PM concentrations, derived from an implementation of more restrictive air quality standards, could reduce 32,000±5,300 deaths due to all causes in the four metropolitan areas of Brazilian southeast.

Because of the largest emission and population, MASP presented the majority of the mortality estimated (68.3%), with MARJ presenting 29.2% of avoidable deaths. MABH was responsible for 2.0% and MAGV for 0.5% of the total avoidable deaths estimated (the percentages vary according to the health outcome). However, when a base of 100,000 is used, MASP and MARJ presented similar results, while in MAGV avoidable deaths are higher than MABH. Another important finding is that ischemic heart diseases were primarily responsible for the estimated avoidable deaths due to cardiovascular diseases.

Regarding the avoidable hospitalizations, the effect of PM_{10} was smaller than $PM_{2.5}$ concentrations, with the avoidable number of hospitalizations in children for $PM_{2.5}$ being higher than PM_{10} in MABH, MAGV, and MARJ, which is related to the ΔPM in Equation 1 (the WHO 24-h guideline for $PM_{2.5}$ concentration is half the PM_{10} concentration), the RR's used, and the $PM_{2.5}$ overestimation. This result highlights the importance of spread $PM_{2.5}$ monitoring to other cities, especially in those identified as possible to have higher $PM_{2.5}$ concentration.

The integration of air quality modeling with emission control strategies, or with the direct evaluation of air quality standards, as applied in this work, represents a handy approach to air quality management and assessment, to evaluate the areas where pollutant concentrations may exceed air quality standards and to estimate possible health gains. Epidemiological evidence on airborne PM and public health is substantial, comprising hundreds of reports. It is worth noting that the WHO points out that it is not possible to establish a minimum limit of PM concentration, below which no harmful effects to health would occur (WHO, 2006). Therefore, a standard should be the lowest concentration possible in the context of local and region constraints, capabilities, and public health priorities.

6. FINE PARTICLES AS A PUBLIC HEALTH INDICATOR IN BRAZIL: FROM MONITORING TO MODELING

6.1 Introduction

Several epidemiological studies have been investigating the impacts of air pollution on health. The estimate of chronic health effects associated with air pollution is carried out through cohort studies, in which the risk of a health outcome (for example, death) is examined concerning medium and long-term exposure to pollution, usually comparing people living in different locations. The relationship between changes in short-term air pollution levels and changes in various indicators of population health is studied in time series and panels (EFTIM and DOMINICI, 2005; WHO, 2006).

The statistical techniques involved also differ between the temporal outcome. In the case of short-term effects, the main techniques used are the Generalized Linear Models (GLM) and the Generalized Additive Models (GAM) (CONCEIÇÃO *et al*., 2001b; RAVINDRA *et al*., 2019; YAN *et al.*, 2019), usually employing Poisson regression (TADANO *et al*., 2009; SOLEIMANI *et al*., 2019), often associated with Principal Component Analysis (SOUZA *et al*., 2014), and more recently, with Auto Regressive Vector (SOUZA *et al*., 2018) to eliminate the problem associated with the presence of automatic correlation in the main components when applying GAM. For long-term effects, it is used the Cox Regression Model to analyze survival data (CROUSE *et al.*, 2012; CESARONI *et al*., 2013; CROUSE *et al*., 2015; POPE III *et al*., 2019a), since death is the outcome of interest.

Cohort studies generally provide higher estimates of the pollution effects than time series studies, indicating that long-term exposures present more significant effect than short-term exposures (EFTIM and DOMINICI, 2005). However, the disadvantages of carrying out this type of study are related to logistical difficulties, high cost of implementation, monitoring of populations over long periods with great potential for losses. Also, a large number of individuals are generally needed to carry out the study, in addition to air pollutants data and confounding factors. Moreover, as exposure is generally considered to be an average for the entire city, it is necessary to evaluate different locations to ensure adequate exposure variability (WHO, 2006). Most cohort studies in the air pollution literature focused mainly on mortality and provided the complete estimates of the deaths attributable to exposure to air pollutants and the extent of the average reduction in life expectancy. Therefore, they are considered more suitable for long-term health impact assessment (COHEN *et al.*, 2004; CHEN *et al.*, 2008).

Such studies served as a basis for the World Health Organization (WHO) revise its air quality guidelines in 2005 (WHO, 2006). The review process is based on scientific studies on the effect of air pollutants on health, and it takes into account the opinion of air quality managers and those responsible for public policies, regarding the logic and format of the guidelines, to improve their applicability worldwide. For particulate matter, for example, the guideline value is based on studies that showed, with 95% confidence, an increase in total mortality, cardiopulmonary and lung cancer in response to long-term exposure.

Air quality standards, on the other hand, are set by each country (and states) to protect public health, and they are an essential component of national risk management and environmental policies. In 2018, CONAMA Resolution 491 was published in Brazil, revoking CONAMA Resolution 03/1990, which had been in force for almost three decades without changes, with very permissive air quality standards. The new standards defined by the new legislation must be adopted by the Brazilian states sequentially in four stages (intermediate standard IS-1 to IS-3 and final standard - FS). The last stage (FS) is based on WHO guidelines, considering the implementation of plans to control atmospheric emissions (which must be prepared by state and district environmental agencies) and reports on air quality assessment. Furthermore, it was only in this update in 2018 that the fine fraction of particulate material ($PM_{2.5}$) started to be legislated at the national level. However, the lack of establishing deadlines for the implementation of each intermediate goal and the final standard is still a moot point in CONAMA Resolution 491/2018. Also, IS-1 is still entirely permissible, and does not advocate WHO principles for protecting the health of the population.

Among air pollutants, particulate matter is one of those that most affect people's health. While particles with an aerodynamic diameter less than or equal to 10 μm (PM_{10}) can penetrate and lodge in the lungs, particles with an aerodynamic diameter equal to or less than 2.5 μm ($PM_{2.5}$) are even more harmful to health. $PM_{2.5}$ can cross the pulmonary barrier and enter the blood system. Chronic exposure to particles contributes to the risk of developing cardiovascular and respiratory diseases, as well as lung cancer (WHO, 2006; GAUTAM and BOLIA, 2020).

For particulate matter, Table 6.1 shows the reference values of CONAMA Resolution 491/2018. As the PM thresholds below which no adverse effects have not yet been identified, the values of WHO (FS in bold) guidelines may not fully protect human health, which highlights the importance of restrictive standards.

Table 6.1 - National standards for particulate material according to CONAMA Resolution 491/2018. The final standard of PM_{10} and $PM_{2.5}$ coincides with WHO guidelines.

Pollutant	**Sampling Time**	**IS-1 (μg/m³)**	**IS-2 (μg/m³)**	**IS-3 (μg/m³)**	**FS (μg/m³)**
Total Suspended Particles (TSP)	24 hours	-	-	-	240
	AGM[1]	-	-	-	80
Particulate matter (PM_{10})	24 hours	120	100	75	50
	AAM[2]	40	35	30	20
Particulate matter ($PM_{2.5}$)	24 hours	60	50	37	25
	AAM[2]	20	17	15	10

Notes: [1]annual geometric mean; [2] annual arithmetic mean.

A review regarding epidemiological studies carried out in Brazil and PM_{10}, mainly concerning respiratory diseases, was recently published (FERNANDES *et al*., 2020). The present work aims to review the epidemiological studies for $PM_{2.5}$. The current situation of the Brazilian monitoring of $PM_{2.5}$ was also assessed. As an alternative to the use of monitored data, the use of air quality models for epidemiological studies were evaluated. Finally, the importance of PM emission control strategies is shown and how a well-designed public emission control policy can bring health benefits.

6.2 Lack of Brazilian studies relating fine particles and their effects on health

Several epidemiological studies show and search for quantify the relationship between particulate matter and its adverse health effects in short-term (NASCIMENTO *et al.*, 2017; MACHIN *et al.*, 2018; GOUVEIA *et al.*, 2019), medium-term (AGUDELO-CASTAÑEDA *et al.*, 2019) e long-term (CROUSE *et al.*, 2015; DOWNWARD *et al.*, 2018; POPE III *et al.*, 2019a).

In Brazil, epidemiological studies have focused, so far, on the effects of short-term pollutants, with PM_{10}, O_3, NO_2, SO_2, and CO being the most studied pollutants. Fernandes et al. (2020) reviewed 36 scientific articles for 36 Brazilian cities since 1999, with PM_{10} being the focus of the majority.

Online database searches were conducted by using Web of Science, Scopus, and Google Scholar, combining the following keywords: epidemiological studies, time series, morbidity, hospital admission, hospitalization, health impact, air pollution, air quality, hazard ratio, relative risk, Brazil, Brazilian cities.

Only 13 Brazilian epidemiological time series studies in the literature relating to $PM_{2.5}$ and respiratory diseases were identified, and most studies focused on the age group below 10 years old. Many authors investigated economic activities relationships, such as the burning of sugarcane (CANÇADO *et al.*, 2006; CESAR *et al.*, 2013), and forest burning and the Amazon deforestation (IGNOTTI *et al.*, 2010; CARMO *et al.*, 2010; SILVA *et al.*, 2013). Another three studies evaluated other diseases: Montavani *et al*. (2016) investigated the relationship between $PM_{2.5}$ and cardiovascular diseases; Ferreira *et al*. (2016) with diseases of the circulatory system; and Ribeiro *et al*. (2019) with ischemic heart diseases. Such studies obtained relative risks higher than most studies related to respiratory diseases.

Table 6.2 summarizes the relative risks found in the epidemiological studies researched. The most common increase in $PM_{2.5}$ concentrations related to RR was 10.0 $\mu g\ m^{-3}$. The interval between the observed daily concentration and the outcome (lag) varied between 0 and 7 days in the studies surveyed, with the majority of studies bringing RR for each lag. Table 2 presents the significant RR found in each study (one RR per study).

Meteorological variables also play an important role in estimating regression coefficients. For example, applying GAM, Pearce *et al*. (2011) showed that, for Melbourne (Australia), the aggregate impact of meteorological variables explained 26.3% of the ozone variation (related to temperature, boundary layer height, and radiation), 21.1% in PM_{10} (temperature, wind, water vapor pressure, and boundary layer height) and 26.7% in NO_2 (temperature, wind, and water vapor pressure). Other meteorological variables obtained less pronounced responses. He *et al*. (2017) demonstrated that the weather conditions were the main factor that determined the daily variations in pollutant concentrations, explaining more than 70% of the variation of the daily average pollutant concentrations in China between 2014 and 2015. Andreão *et al*. (2019a) evaluated the influence of mesoscale meteorological phenomena on $PM_{2.5}$ in the Metropolitan Area of Greater Vitória, showing that precipitation, associated with cold fronts, was responsible for the decrease in fine particles concentration. In contrast, high pressure systems in the region led to an increase in particle concentration due to atmospheric stagnation conditions.

In the statistical analyses carried out by the researched epidemiological studies, temperature and relative humidity were the two most used meteorological variables as a confounding factor in the regressions. Carmo *et al*. (2010) also considered precipitation, and Ferreira *et al*. (2016) wind speed and wind direction.

Another critical issue is the removal of seasonal patterns in the data before assessing the short-term effects. The number of hospital admissions generally differs between weekdays and weekends, and the concentration of air pollutants varies throughout the year (TADANO *et al.*, 2009; WANG *et al.*, 2019). Therefore, confounding factors such as the days of the week and seasonality itself (variation over the year) were usually adopted in the researched studies.

Regarding the study region, the state of São Paulo was the one with the highest number of works, followed by Mato Grosso. Vitória and Volta Redonda were two cities outside these two states that presented studies.

Table 6.2 - Epidemiological studies of time series in Brazilian cities focusing on $PM_{2.5}$.

Reference	Study area	Age range	Studied disease	RR (CI 95%)	Increment	Lag
Menezes *et al.* (2019)	Cuiabá (MT)	girls < 10 years old	Respiratory	1.22 (1.05-1.42)*	5.0 μg m^{-3}	Lag-6
Machin *et al.* (2019)	Cuiabá (MT)	> 60 years old	Respiratory	1.467 (1.118-1.882)*	3.5 μg m^{-3}	Lag-4
Machin and Nascimento (2018)	Cuiabá (MT)	< 10 years old	Tracheitis, laryngitis, pneumonia, bronchitis, bronchitis, asthma	1.035 (1.006-1.065)	1.0 μg m^{-3}	Lag-2
Nascimento *et al.* (2017)	Vitória (ES)	< 12 years old	Respiratory	1.06 (1.00-1.11)	4.2 μg m^{-3}	Lag-6
Ferreira *et al.* (2016)	São José dos Campos (SP)	> 60 years old	Respiratory	1.085 (0.932-1.263)	10.0 μg m^{-3}	Lag 0 a 5
César *et al.* (2016)	Taubaté (SP)	< 10 years old	Pneumonia and asthma	1.051 (1.016-1.088)	5.0 μg m^{-3}	Lag-0
Nascimento *et al.* (2016)	Volta Redonda (RJ)	All ages	Pneumonia, acute bronchitis, bronchiolitis and asthma	1.022 (1.005-1.038)	5.0 μg m^{-3}	Lag-5
Patto *et al.* (2016)	São José do Rio Preto (SP)	< 10 years old	Pneumonia	1.28 (1.05-1.56)*	10.0 μg m^{-3}	Lag-5
Cesar *et al.* (2013)	Piracicaba (SP)	< 10 years old	Respiratory	1.079 (1.001-1.016)	10.0 μg m^{-3}	Lag-1
Silva *et al.* (2013)	Cuiabá (MT)	< 5 years old	Respiratory	1.091 (1.018-1.181)	10.0 μg m^{-3}	Lag-0
Carmo *et al.* (2010)	Alta Floresta (MT)	< 5 years old	Respiratory	1.029 (1.003-1.055)	10.0 μg m^{-3}	Lag-6
Ignotti *et al.* (2010)	Alta Floresta (MT)	< 5 years old	Respiratory	1.042 (1.001-1.085)	10.0 μg m^{-3}	Lag-4
Cançado *et al.* (2006)	Piracicaba (SP)	< 13 years old	Respiratory	1.214 (1.043-1.385)	10.2 μg m^{-3}	Lag-0
Ribeiro *et al.* (2019)	Taubaté (SP)	> 40 years old	Ischemic heart disease	1.19 (1.05-1.34)	5.0 μg m^{-3}	Lag-4
Montavani *et al.* (2016)	São José do Rio Preto (SP)	All ages	Cardiovascular	1.17 (1.01-1.34)*	10.0 μg m^{-3}	Lag-5
Ferreira *et al.* (2016)	São José dos Campos (SP)	> 60 years old	Circulatory system	1.196 (1.064-1.346)	10.0 μg m^{-3}	Lag 0 a 5

* Estimated from figures and tabulated regression coefficients.

Regarding cohort studies referring to $PM_{2.5}$, Vodonos *et al*. (2018) conducted a meta-regression study in which 53 studies were selected from 29 cohort studies that provided 135 estimates of the quantitative association between mortality risk and exposure to $PM_{2.5}$. Of the total studies evaluated, 39 studies (eighteen cohorts) were from North America, eight studies (six cohorts) from Europe and six studies (five cohorts) from Asia. These numbers underscore the lack of such study for South America. Andreão *et al*. (2018) also highlight the non-existence of cohort studies for Brazil, which shows that there is a gap in the research that still needs to be investigated. Pope III *et al*. (2019b) performed a meta-analysis with 75 cohort studies from North America, Europe, and Asia. The RR estimated by 10 μg m^{-3} of long-term exposure to $PM_{2.5}$ was 1.08 (95% CI, 1.06-1.11) for all-cause mortality, 1.11 (1.08-1.14) for cardiopulmonary mortality and 1.13 (1.07-1.20) for lung cancer mortality.

6.3 $PM_{2.5}$ monitoring in Brazil

Despite several studies found for Brazil relating particulate matter and health[38], few have investigated $PM_{2.5}$, mainly due to the lack of monitoring data. The start of continuous $PM_{2.5}$ monitoring is recorded in São Paulo city in 1999 with a single manual station, but with a valid annual average only in 2000 (23 μg m^{-3}) (ANDREÃO *et al.*, 2018). In 2018, 81 stations (between manual and automatic) were identified in Brazil, as depicted in Figure 6.1, all stations located in the Southeast.

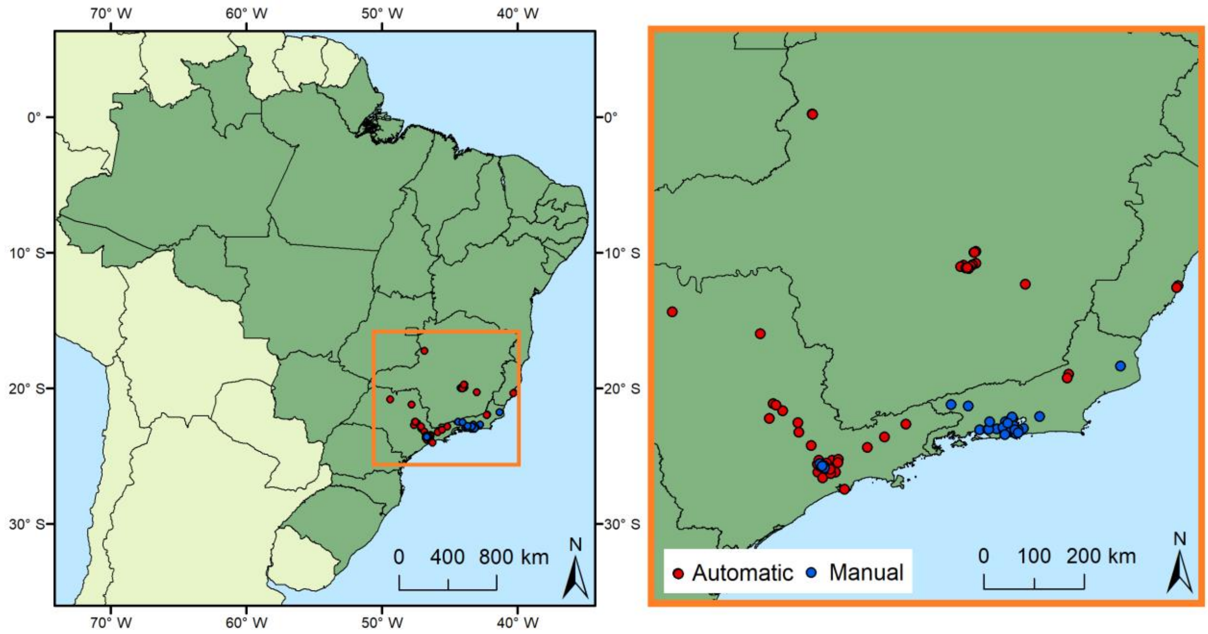

Figure 6.1 - $PM_{2.5}$ monitoring stations in Brazil (2018/2019).

Figure 6.2 shows the evolution of $PM_{2.5}$ annual averages for São Paulo city, from 2000 to 2019, for both automatic and manual monitoring stations. In 2019, among the fifteen monitoring stations, eleven were automatic and four manuals. Until 2010, the stations were only manual. It is observed that in all years, the WHO annual guideline (10 μg m^{-3}) is exceeded.

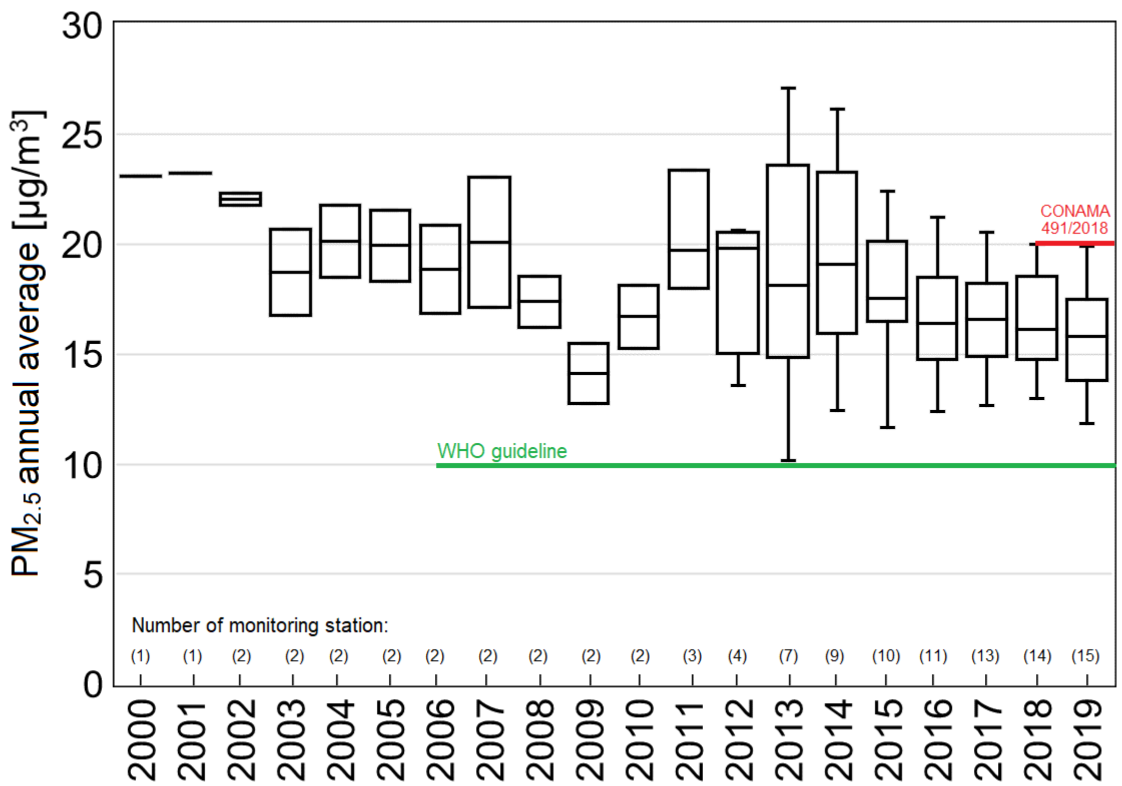

Figure 6.2 - Annual $PM_{2.5}$ averages of the monitoring stations (between manual and automatic) in São Paulo.

The annual averages, showed in Figure 6.3, include the cities of Belo Horizonte, Vitória, and Rio de Janeiro, with the values presented being the average value of all stations in each city. It is noticed that throughout the period, the $PM_{2.5}$ annual concentrations were above the WHO annual guideline (10 μg m^{-3}), except for Rio de Janeiro in 2018. In 2019, Vitória presented 60% of hourly $PM_{2.5}$ data, which probably contributed to the increased observed. Belo Horizonte started monitoring $PM_{2.5}$ recently, and the stations are slightly consolidating over time, in terms of missing and valid data.

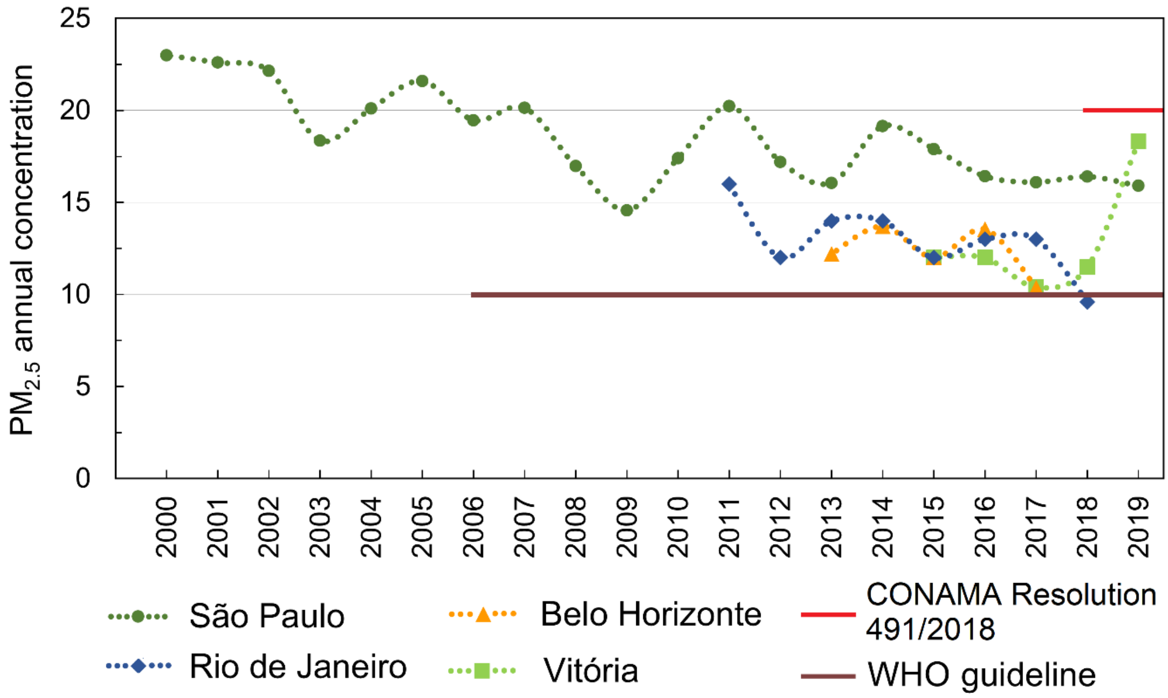

Figure 6.3 - Annual $PM_{2.5}$ concentration [µg m^{-3}] in the four Brazilian Southeast capitals.

The scarce distribution and lack of monitoring stations for particulate matter in Brazil means that the environmental concentrations of $PM_{2.5}$ are unknown in most Brazilian cities, which makes it challenging to carry out epidemiological studies in these cities. However, an increasingly used alternative (or even an extension to monitoring to fill in missing data) is air quality modeling.

6.4 Modeling of particulate matter for use in epidemiological studies

Chemical transport models have become widely recognized and are used as tools to follow trends (ANDRADE *et al.*, 2015; ZHANG *et al.*, 2018), define responsibilities for air pollution levels in regulatory analysis (RING *et al.*, 2018; SONG *et al.*, 2019), and evaluation of control strategies (WANG *et al.*, 2016; ALBUQUERQUE *et al.*, 2019), for example. They are models that simulate changes in pollutant concentrations in the atmosphere using a set of mathematical equations that characterize the chemical and physical processes in the atmosphere. They are applied at multiple spatial scales, from local to global (VALLERO, 2008). The use of gridded spatialized data facilitates the association of air pollution on health impacts, for example.

The particles can be emitted directly into the air (primary particles) or be formed in the atmosphere (secondary particles) from gaseous precursors, such as sulfur dioxide, nitrogen oxides, ammonia and non-methane volatile organic compounds (ALBUQUERQUE *et al.*, 2019). Despite varying from region to region, the main components of PM are sulfate, nitrates, ammonia, sodium chloride, organic and elemental carbon, mineral dust and water (MIRANDA *et al.*, 2018). It consists of a complex mixture of solid and liquid particles of organic and inorganic substances suspended in the air. Therefore, when the focus of epidemiological studies is fine particles, and air quality modeling will be employed, it is crucial to consider the formation of secondary aerosols in the model, employing an appropriate chemical mechanism and a correctly speciated inventory of chemical emissions.

Twelve of the sixteen Brazilian epidemiological studies used modeling to generate daily $PM_{2.5}$ concentration (IGNOTTI *et al.*, 2010; CARMO *et al.*, 2010; CESAR *et al.*, 2013; SILVA *et al.*, 2013; SOUZA *et al.*, 2014; MONTAVANI *et al.*, 2016; CÉSAR *et al.*, 2016; NASCIMENTO *et al.*, 2016; PATTO *et al.*, 2016; RIBEIRO *et al.*, 2019; MENEZES *et al.*, 2019; MACHIN *et al.*, 2019), using the Brazilian air quality model, the Chemical Coupled Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System (CCATT-BRAMS), developed by the *Centro de Previsão de Tempo e Estudos Climáticos/Instituto Nacional de Pesquisas Espaciais* (CPTEC/INPE).

However, in CCATT-BRAMS, currently only BRAMS (FREITAS *et al.*, 2017), PM chemical reactions are not validated, and therefore, it is recommended its application only for PM dispersion (FREITAS *et al.*, 2017). In this case, other chemical transport models would be more suitable for PM studies, such as the *Weather Research and Forecasting* (WRF) *model coupled with Chemistry* (WRF-Chem) and the *Community Multi-Scale Air Quality* (CMAQ). Both models have been used in Brazil (ANDRADE *et al.*, 2015; ARCHER-NICHOLLS *et al.*, 2015; VARA-VELA *et al.*, 2018; ALBUQUERQUE *et al.*, 2019; PEDRUZZI *et al.*, 2019; ANDREÃO *et al*., 2020b) and may serve for future investigations of PM effects on health.

It is emphasized the need for meteorological and pollutant validation, given by comparing the modeled data with observed data. In the case of meteorological variables, the most common ones for validation are temperature, specific humidity, and wind speed and direction (EMERY *et al.*, 2001), using statistical indicators such as mean bias, mean error, root of mean square error and index of agreement. Benchmark values for each meteorological variable and statistical indicator are found in the literature (EMERY *et al.*, 2001; LADCO and WDNR, 2016; RAMBOLL, 2018). Such modeled variables can also be used as confounding factors in epidemiological studies.

For the validation of the air quality model, Simon *et al.* (2012) reviewed of the main statistical indicators used in the literature, and Emery *et al.* (2017) bring benchmark values for particulate matter for the indicators that they considered to be the most appropriate: mean normalized bias; mean normalized error; and correlation coefficient.

Therefore, even with the application of air quality models, there is a need to use measured environmental data to validate the model, which reinforces the need to expand the fine particle monitoring network in Brazil.

Other important parameters to be considered in the modeling are the parameterization used to represent microphysics of clouds, radiation, planetary boundary layer, cumulus, surface layer, and photolysis rate, for example. Good practices also include evaluating grid spacing (PUNGER and WEST, 2013; KORHONEN *et al.*, 2019), initial (HOGREFE *et al.*, 2017) and boundary conditions (BORGE *et al.*, 2010; GAVIDIA-CALDERÓN *et al.*, 2018; PEDRUZZI *et al.*, 2019), segmentation or not of simulation (ANDREÃO *et al.*, 2019b), use of nudging (SILVA *et al.*, 2018; TRAN *et al.*, 2018) and what many consider the most important: emission inventories (SANTOS *et al.*, 2019; PINTO *et al.*, 2020a; IBARRA-ESPINOSA *et al.*, 2019).

An emission inventory, including knowledge of daily, weekly and monthly emissions variations, is essential for successful air quality modeling (ARYA, 1999; PINTO *et al.*, 2020a; IBARRA-ESPINOSA *et al.*, 2019). Emission inventories are usually supplied by environmental agencies or companies, from single sources, reporting only the annual emission rate (tons per year), mainly for each pollutant legislated in a given latitude and longitude. As air quality models require, as input data, emissions in three dimensions (or even only for the first layer, when simplified), spatialized in an area and varying in time, it is necessary to process the primary data and transform them in a suitable format to be able to apply them in air quality models. However, when working in Brazil on a regional scale, or even higher, it is still necessary to resort to global inventories, such as, for example, the *Emission Database for Global Atmospheric Research* (EDGAR), *REanalysis of the TROpospheric chemical composition over the past 40 yr.* (RETRO), among others, or build their inventory.

Modeling also assists in identifying the contribution of local and regional sources to air quality. Chen *et al*. (2014) used CMAQ to simulate the effects of East Asian emissions on $PM_{2.5}$ levels in Taiwan, assessing the effects of direct (precursors directly forming $PM_{2.5}$ in local areas) and indirect (precursors transported that interact with local precursors in the formation of $PM_{2.5}$) long-range transport. The results indicated that the contributions to the annual average of $PM_{2.5}$

of 30 μg m^{-3} found in Taiwan are 60, 27, 9 and 3%, respectively, of Taiwan's contribution, direct long-range transport, indirect long-range transport, and background. Shimadera *et al.* (2016) evaluated the performance of the CMAQ model for simulating long-range transport and local $PM_{2.5}$ pollution in Japan between April 2010 and March 2011. The contribution of long-range transport was 50% on average and generally higher in western Japan (closer to the mainland). Guo *et al*. (2019), for example, identified that 24% of premature deaths due to $PM_{2.5}$ in Delhi (India) are from non-local sources. Therefore, the impact of non-local emissions may be significant when evaluating health impacts.

6.5 Excess hospitalizations, deaths and their costs

Estimates of excess mortality and morbidity can be a direct measure of the use of the relative risks of epidemiological studies and give a clearer view to the population and decision makers about the effect of air pollutants on health. They can also be a critical component of assessments of the air quality policy. After estimating the reductions in the incidence of adverse health effects, it can be calculated the monetary benefits associated with these reductions. For this, some tools were developed to quantify the impact of air pollutants on health, such as BenMAP-CE (SACKS *et al.*, 2018), AirQ+ (WHO, 2018b) and Aphekom (APHEKOM, 2011), for example.

Health effects can be estimated using scenarios of air pollutant concentrations, usually one representing current conditions and a future one, with reduced pollutants concentrations. These air quality data are specified by the user (modeling or monitoring). The log-normal formulation (Equation 6.1) is the most used health impact function to estimate short (daily) and long-term (annual) health effects (SACKS *et al.*, 2018). In this equation, ΔY represents the change in the population's health response and takes into account: the incidence of the assessed effect for the base case (Y_o); the estimated effect (β) which is derived from epidemiological studies; the change in air quality between a base scenario and control (ΔQ); and the exposed population (Pop).

$$\Delta Y = Y_o \cdot \left(1 - e^{-\beta \cdot \Delta Q}\right) \cdot Pop \tag{6.1}$$

Subsequently, the economic value can be obtained by multiplying the values for reducing health effects by the estimated financial value per case. In this way, different scenarios simulating changes in air quality can be obtained. Andreão *et al*. (2018), Howard *et al*. (2019), and Fernandes *et al*. (2020) are studies that explored the use of such methodology in Brazil.

As an example of application of the methodology described above, the present study used the average annual concentrations of $PM_{2.5}$ (satellite-based estimates, simulations of chemical transport models and measurements at monitoring stations) brought by Brauer *et al*. (2016) in its study for the Global Burden of Diseases 2013, with spatial resolution of 0.1° × 0.1°, to estimate avoidable mortality for Brazilian municipalities for all causes (ICD-10: A00-Y98), cardiopulmonary (I00-I09; I11; I13; I20-I51; I60-I69; J09-J18; J40-J47), and lung cancer (C33-C34), using the RR from Pope III *et al.* (2019b) meta-analysis study, already presented earlier. From the data by Brauer *et al*. (2016), an annual $PM_{2.5}$ average concentration for each of the 5,572 municipalities was estimated (Figure 6.4a) using the geographic information system and the limits of each municipality. The 2013 mortality data were obtained from *Departamento de Informática do Sistema Único de Saúde* (DATASUS).

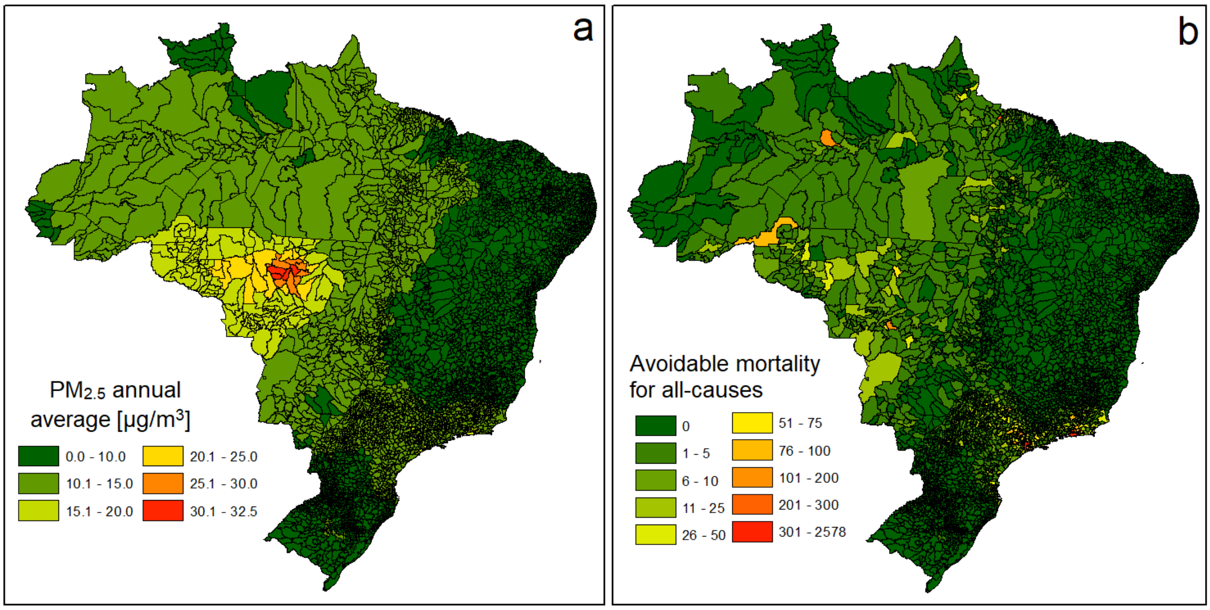

Figure 6.4 - (a) $PM_{2.5}$ annual average per municipality in 2013 from the concentrations of Brauer *et al*. (2016); (b) Avoidable all-cause mortality estimated for 2013.

Considering the maximum annual concentration recommended by WHO (10 µg m^{-3}), and applying Equation 6.1, a total of 15,638 ± 2,427 deaths for all causes (Figure 6.4b) could be avoided in 2013 in 2,238 municipalities that presented concentrations above WHO guideline. For cardiorespiratory causes, there were 7,980 ± 1,078 estimated deaths, and for lung cancer, 559 ± 137 deaths. These numbers translate the necessity for a more restrictive standard for fine particles in Brazil.

6.6 PM control strategies: design and efficiency

For the establishment of more restrictive standards of fine particles (and other air pollutants), the primary control mechanism is the reduction of atmospheric emissions. In São Paulo, for example, the Plan to Reduce Emissions from Stationary Sources (*Plano de Redução de Emissão de Fontes Estacionárias* - PREFE) and the Vehicle Pollution Control Plan (*Plano de Controle de Poluição Veicular* - PCPV) was planned for this purpose. Taking as an example the group of municipalities in the Metropolitan Area of São Paulo, 20.5% reduction targets for environmental ozone concentration were established (with 20.5% reduction in NOx and VOC emissions from fixed and mobile sources) and 4.8% in environmental concentrations and emissions of PM (with 4.8% reduction from both mobile and fixed sources).

However, it is possible to perceive in this control strategy the neglect of chemistry and atmospheric transport. The 4.8% reduction in PM emissions will not necessarily result in a 4.8% reduction in PM concentration. The relationship between local emissions and environmental concentrations is generally not linear, being a function of meteorology, as already demonstrated, and geographic conditions (topography), which can significantly change the conditions of dispersion and secondary formation of pollutants (KARAGULIAN *et al.*, 2015; ALBUQUERQUE *et al.*, 2018).

Therefore, control strategies, mainly of secondary pollutants, must consider the interaction between pollutants and emitting sources, meteorology, topography, land use and land cover. In this sense, chemical transport models are the tool designed for this purpose (WANG *et al.*, 2016; ALBUQUERQUE *et al.*, 2019 to test whether the planned emission reduction will result in the reduction of the environmental concentration of pollutants. The importance of a well-designed and modeled atmospheric emissions control strategy is emphasized, so that the health benefits resulting from improved air quality are, in fact, significant.

Considering that most of the fine particles are from secondary origin, actions to directly reduce PM emissions may not achieve the desired effect in ambient concentrations. As well, reducing some precursors of $PM_{2.5}$ can only result in a change in the chemical composition of PM, and no reduction in its mass concentration. In addition to the need to reduce $PM_{2.5}$ concentrations, other factors such as the chemical composition of PM and its concentration in the number of particles are also significant to assess health impacts.

6.7 Conclusion

There is still much to be done in Brazil in terms of air pollutants monitoring, especially fine particles. $PM_{2.5}$ epidemiological studies on Brazilian cities were based mainly on air quality modeling using the CCATT-BRAMS model. With the establishment of national standards for $PM_{2.5}$ by CONAMA Resolution 491/2018, although still much more permissive than WHO guidelines, it is expected that more cities start to monitor this pollutant, and therefore, new epidemiological studies can be carried out, in order to determine more comprehensively (territorial) its relationship with respiratory and cardiovascular diseases.

The availability of monitored data for research and population knowledge is still an issue to be faced in some cities. A consistent database, without missing data, is essential to work with time series. Concerning modeling, the validity of the estimates depends on how well the model fit the observational data. In Brazilian studies, the lack or the availability of measured environmental data can impair the validation of the modeling and its application. The lack of emission inventories is also a critical point to be addressed in Brazil. For a more coherent representation of the particles by the models, an accurate and chemically specified emission inventory is necessary.

To apply workable public policies that will generate positive results in terms of reducing the emission and environmental concentration of air pollutants, and as a consequence, improving population health, it is necessary to consider the interactions between pollutants, meteorology conditions, and topography for example. Restrictive air quality standards evaluation, human exposure and health effects are crucial factors to consider in urban air quality management, and air quality modeling is recommended for such investigation.

7. FINAL CONSIDERATIONS

Air quality management, especially in urban centers, refers to activities aimed to reduce air pollution to protect human health and the environment. Franco *et al*. (2019) mention some essential factors relation to urban air quality management: the recognition air pollution importance; the development of extensive knowledge of pollution conditions; a reliable and representative air quality monitoring; a clear knowledge of emission sources; information and requirements needed by users of environmental data; the understanding of air quality planning and its management as a comprehensive activity; control emissions; implementation of a response system to critical pollution episodes; and the establishment of a human capital with technical and managerial capacities to face the problem, represented by a stable, interdisciplinary and trained team. The present work goes further and demonstrates the need to formulate comprehensive long-term management plans with well-defined ambitious air quality standards. Therefore, two other essential factors for urban air quality management are endorsed: human exposure and health effects.

With the main objective of estimating the health benefits resulting from an improvement in air quality, at first, all annual averages of fine particulate matter available at the national level (until 2017) were used. To evaluate the health benefits in a higher number of cities, including those without monitoring, and the interaction of air pollutants between them, a numerical modeling tool was used (for 2015) for the four Brazilian Southeast metropolitan areas.

Although the Brazilian air quality standards were updated after almost three decades, the first intermediate standards established by CONAMA Resolution 491/2018 are still permissive compared to WHO guidelines. For the annual average of $PM_{2.5}$, for example, the first intermediate standard (20 $\mu g\ m^{-3}$) is double that of the guideline recommended by WHO. Furthermore, the lack of a deadline for the establishment of each goal and the final standard is still an issue to be faced.

There is still much to be done in Brazil in terms of monitoring air pollutants, particularly for fine particles, which is only monitored in a few cities in the Southeast. The availability of monitored data for research and for the knowledge of the population itself is still an obstacle in most Brazilian cities. With the adoption of WHO annual air quality guideline for the $PM_{2.5}$ (10 $\mu g\ m^{-3}$), between 2,380 ± 800 and 6,280 ± 1,820 deaths from all causes, it could be avoided in 2017 in only 15 cities evaluated in Brazil. These numbers show the importance of adopting deadlines for the implementation of each intermediate goal and final standard of CONAMA Resolution 491/2018, and improving the air quality monitoring, expanding throughout the national territory. $PM_{2.5}$ is also produced by the secondary formation in the atmosphere, reducing the concentration of other pollutants may result in a decrease in the formation of $PM_{2.5}$. Policies and investments that support cleaner transport, alternative energy generation, control of industrial emissions, and better management of municipal waste could reduce the main sources of fine particles and reduce exposure.

The integration of air quality modeling with emission control strategies, or with the direct assessment of air quality standards, as applied in this work, represents an advantageous approach to the management and assessment of air quality, to assess areas where pollutant concentrations may exceed air quality standards and estimate possible health gains.

For adequate air quality modeling, the importance of a representative emissions inventory for the area of interest was shown. In this work, given the large area, it has initially opted for the use of global emission inventories. Because the main global inventory (EDGAR) available for Brazil resulted in high PM concentrations for MASP, it was tested and used the use of a vehicular PM emissions calculated from the 2nd National Inventory of Atmospheric Emissions by Road Motor Vehicles was implemented, spatially distributed considering the urban areas of each Brazilian city and the population and fleet of each city, together with burning emissions. The resulting inventory can then be seen as a valuable environmental management tool for the region, especially those without any information on air pollutants emission.

The results with the application of WRF-Chem showed that cities around the capitals, with a high concentration of modeled $PM_{2.5}$, do not monitor this pollutant. Compliance with the WHO annual guideline would have the potential to avoid 32,000 ± 5,300 deaths from all causes in the four metropolitan regions of the Southeast in 2015. When a base of 100,000 inhabitants is used, MASP and MARJ showed similar results, while in MAGV, preventable deaths are higher than in MABH. Ischemic heart disease was primarily responsible for the estimated preventable deaths from cardiovascular disease.

Another highlight given in this work was the importance of using local cohort studies to estimate health benefits, which is not yet possible in Brazil. The choice of the cohort study that serves as a basis for the estimation of health benefits can generate considerable differences, which may be related to the chemical composition of the PM and its heterogeneous mixture of particle sizes. The use of international epidemiological study for local avoidable deaths estimation must consider the environmental characteristics of each study area (geographical location, emission sources, and mixtures of pollutants), variability between different populations, socioeconomic conditions, and exposure assessment methodology.

The epidemiological evidence on PM and public health is substantial, comprising hundreds of publications. However, in Brazil, until 2019, only 13 epidemiological studies of time series have been found in the literature relating $PM_{2.5}$ and respiratory diseases, and a smaller number with diseases of the circulatory system, which shows that there is still ample space for research in this area.

Therefore, for future studies, it is recommended to carry out epidemiological studies relating $PM_{2.5}$ and various health outcomes in the short, medium, and when possible, long-term, in the most diverse Brazilian cities, using chemical transport models.

It is also recommended to apply the methodology used in this research to estimate health benefits in other metropolitan areas. The elaboration of a national inventory, more detailed, spatialized, involving several economic sectors, is also a valuable tool to generate more precise and accurate results in air quality modeling.

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APPENDIX I

Table SI.1 – Average annual $PM_{2.5}$ concentration in Brazilian cities with monitoring ($\mu g\ m^{-3}$).

Cities	**2017**	**2016**	**2015**	**2014**	**2013**	**2012**	**2011**	**2010**	**2009**	**2008**	**2007**	**2006**	**2005**	**2004**	**2003**	**2002**	**2001**	**2000**
Belfort Roxo					20	18	20											
Campos dos Goytacazes			9	12	9	8												
Duque de Caxias	10	12	14	22	21	20	19											
Itaboraí			8	18	14													
Niterói		11	9	15	14	10	14											
Rezende		11	9	13	8		13											
Rio de Janeiro	13	13	12	14	14	12	16											
São João de Meriti	11	16	12	15	18	12	16											
Seropédica				15	10	10	11											
Volta Redonda					9		10											
Campinas	17	18																
Guarulhos	18																	
Piracicaba	13	13	13	15	14	15												
Ribeirão Preto	13																	
Santos	15.4	14.6	16	18	19	16												
São Bernardo do Campo	16.2	16.7	17															
São Caetano do Sul	18	16.8	19.6	14.5	18.2	20.1	22.9	19.3	16.3			20.7	21.5	20.9	20.6	21.8	22.9	
São José do Rio Preto	16	15.3	14.2	16	14		11.5	13.9	10.9	13.7								
São José dos Campos	12.1	12.4																
São Paulo	16.1	16.4	17.9	19.1	16.1	17.2	20.2	17.4	14.6	17	20.1	19.5	21.6	20.1	18.4	22.2	22.6	23.3
Taubaté	12.9	15.1																
Belo Horizonte				13.7	12.2													
Vila Velha	9.7	11	11.4															
Vitória	10.4	12	12															

Table SI.2 – Avoidable deaths for the cities of Espírito Santos state, with the standard deviation in parentheses.

Health outcome	Reference	Vila Velha			Vitória		
		2017	2016	2015	2017	2016	2015
All Causes	Pope *et al*. (2002)	0	14 (5)	19 (7)	4 (2)	21 (8)	21 (8)
	Krewski *et al*. (2009)	0	7 (2)	10 (3)	2 (1)	11 (4)	10 (4)
	Laden *et al*. (2006)	0	20 (6)	29 (8)	6 (2)	29 (8)	27 (8)
Non-Accidental	Cesaroni *et al*. (2013)	0	8 (1)	12 (1)	3 (0)	13 (2)	13 (2)
	Crouse *et al*. (2012)	0	30 (1)	42 (5)	10 (1)	47 (2)	45 (2)
Cardiovascular	Cesaroni *et al*. (2013)	0	4 (1)	5 (1)	1 (0)	5 (1)	5 (1)
	Crouse *et al*. (2012)	0	9 (1)	12 (1)	3 (0)	14 (1)	13 (1)
	Laden *et al*. (2006)	0	8 (2)	9 (2)	2 (1)	10 (3)	9 (2)
Ischemic Heart Disease	Pope *et al*. (2004)	0	5 (1)	5 (1)	1 (0)	7 (1)	6 (1)
	Krewski *et al*. (2009)	0	4 (1)	5 (1)	1 (0)	6 (1)	5 (1)
	Crouse *et al*. (2012)	0	7 (0)	9 (1)	2 (0)	11 (1)	9 (1)
	Cesaroni *et al*. (2013)	0	3 (0)	3 (1)	1 (0)	4 (1)	3 (1)
Lung Cancer	Pope *et al*. (2002)	0	1 (0)	1 (0)	0 (0)	2 (1)	1 (0)
	Krewski *et al*. (2009)	0	1 (0)	1 (0)	0 (0)	1 (0)	1 (0)
	Cesaroni *et al*. (2013)	0	0 (0)	0 (0)	0 (0)	1 (0)	1 (0)

Table SI.3 - Avoidable deaths for the cities of Rio de Janeiro state (part 1), with the standard deviation in parentheses.

Health outcome	**Reference**	**Belfort Roxo**			**Campos dos Goytacazes**			
		2013	**2012**	**2011**	**2015**	**2014**	**2013**	**2012**
All Causes	Pope *et al*. (2002)	169 (64)	135 (51)	164 (62)	0	39 (1)	0	0
	Krewski *et al*. (2009)	87 (29)	69 (23)	84 (29)	0	20 (7)	0	0
	Laden *et al*. (2006)	294 (87)	239 (70)	277 (82)	0	59 (17)	0	0
Non-Accidental	Cesaroni *et al*. (2013)	106 (13)	86 (11)	105 (13)	0	24 (3)	0	0
	Crouse *et al*. (2012)	363 (19)	297 (15)	359 (18)	0	84 (4)	0	0
Cardiovascular	Cesaroni *et al*. (2013)	43 (7)	37 (6)	40 (7)	0	9 (1)	0	0
	Crouse *et al*. (2012)	106 (8)	91 (7)	99 (8)	0	22 (2)	0	0
	Laden *et al*. (2006)	115 (31)	102 (28)	105 (29)	0	19 (5)	0	0
Ischemic Heart Disease	Pope *et al*. (2004)	48 (6)	43 (5)	42 (5)	0	8 (1)	0	0
	Krewski *et al*. (2009)	41 (6)	36 (5)	36 (5)	0	7 (1)	0	0
	Crouse *et al*. (2012)	74 (5)	67 (4)	66 (4)	0	13 (1)	0	0
	Cesaroni *et al*. (2013)	28 (5)	25 (4)	25 (4)	0	5 (1)	0	0
Lung Cancer	Pope *et al*. (2002)	6 (2)	5 (2)	5 (2)	0	2 (1)	0	0
	Krewski *et al*. (2009)	5 (2)	4 (1)	4 (1)	0	1 (0)	0	0
	Cesaroni *et al*. (2013)	2 (1)	2 (1)	2 (1)	0	1 (0)	0	0

Table SI.4 - Avoidable deaths for the cities of Rio de Janeiro state (part 2), with the standard deviation in parentheses.

Health outcome	Reference	Itaboraí				Duque de Caxias					
		2016	**2015**	**2014**	**2013**	**2016**	**2015**	**2014**	**2013**	**2012**	**2011**
All Causes	Pope *et al*. (2002)	0	0	66 (25)	34 (13)	66 (24)	131 (49)	381 (144)	363 (137)	318 (120)	199 (113)
	Krewski *et al*. (2009)	0	0	34 (11)	17 (6)	34 (11)	67 (22)	197 (67)	187 (63)	163 (55)	154 (52)
	Laden *et al*. (2006)	0	0	110 (32)	58 (17)	109 (31)	214 (61)	603 (180)	603 (179)	536 (159)	518 (152)
Non-Accidental	Cesaroni *et al*. (2013)	0	0	41 (5)	21 (3)	41 (5)	82 (10)	239 (31)	228 (29)	198 (25)	188 (24)
	Crouse *et al*. (2012)	0	0	142 (7)	74 (4)	148 (7)	291 (14)	812 (42)	776 (40)	677 (35)	641 (33)
Cardiovascular	Cesaroni *et al*. (2013)	0	0	16 (3)	8 (1)	19 (3)	37 (6)	107 (18)	97 (16)	83 (14)	78 (13)
	Crouse *et al*. (2012)	0	0	39 (3)	20 (2)	47 (4)	93 (7)	260 (21)	236 (19)	203 (16)	192 (15)
	Laden *et al*. (2006)	0	0	39 (11)	22 (6)	49 (12)	95 (25)	255 (71)	244 (68)	207 (57)	202 (55)
Ischemic Heart Disease	Pope *et al*. (2004)	0	0	17 (2)	9 (1)	18 (2)	36 (4)	90 (11)	84 (11)	82 (10)	74 (9)
	Krewski *et al*. (2009)	0	0	15 (2)	8 (1)	16 (2)	31 (4)	77 (12)	72 (11)	70 (11)	64 (10)
	Crouse *et al*. (2012)	0	0	27 (2)	14 (1)	30 (2)	58 (4)	139 (9)	131 (9)	128 (8)	116 (8)
	Cesaroni *et al*. (2013)	0	0	10 (2)	5 (1)	11 (2)	21 (4)	54 (10)	50 (9)	49 (9)	44 (8)
Lung Cancer	Pope *et al*. (2002)	0	0	3 (1)	1 (0)	2 (1)	5 (2)	16 (6)	14 (5)	15 (5)	13 (4)
	Krewski *et al*. (2009)	0	0	2 (1)	1 (0)	2 (1)	4 (1)	13 (4)	12 (4)	12 (4)	10 (3)
	Cesaroni *et al*. (2013)	0	0	1 (1)	1 (0)	1 (0)	2 (1)	6 (3)	6 (3)	6 (3)	5 (2)

Table SI.5 - Avoidable deaths for the cities of Rio de Janeiro state (part 3), with the standard deviation in parentheses.

Health outcome	Reference	Niterói						Resende				
		2016	**2015**	**2014**	**2013**	**2012**	**2011**	**2016**	**2015**	**2014**	**2013**	**2011**
All Causes	Pope *et al*. (2002)	24 (9)	0	118 (44)	95 (36)	0	94 (35)	5 (2)	0	13 (5)	0	12 (5)
	Krewski *et al*. (2009)	12 (4)	0	60 (20)	49 (16)	0	48 (16)	2 (1)	0	7 (2)	0	6 (2)
	Laden *et al*. (2006)	29 (8)	0	138 (40)	119 (34)	0	117 (33)	7 (2)	0	21 (6)	0	20 (6)
Non-Accidental	Cesaroni *et al*. (2013)	15 (2)	0	75 (9)	60 (8)	0	59 (7)	3 (0)	0	8 (1)	0	8 (1)
	Crouse *et al*. (2012)	54 (3)	0	261 (13)	212 (10)	0	209 (10)	10 (1)	0	29 (1)	0	27 (1)
Cardiovascular	Cesaroni *et al*. (2013)	5 (1)	0	29 (5)	25 (4)	0	23 (4)	1 (0)	0	4 (1)	0	3 (1)
	Crouse *et al*. (2012)	14 (1)	0	72 (6)	62 (5)	0	58 (4)	3 (0)	0	9 (1)	0	8 (1)
	Laden *et al*. (2006)	11 (3)	0	51 (13)	47 (12)	0	42 (11)	3 (1)	0	10 (3)	0	9 (2)
Ischemic Heart Disease	Pope *et al*. (2004)	8 (1)	0	40 (5)	31 (4)	0	28 (3)	2 (0)	0	5 (1)	0	3 (0)
	Krewski *et al*. (2009)	7 (1)	0	34 (5)	26 (4)	0	24 (4)	1 (0)	0	4 (1)	0	2 (0)
	Crouse *et al*. (2012)	13 (1)	0	64 (4)	50 (3)	0	45 (3)	3 (0)	0	7 (0)	0	4 (0)
	Cesaroni *et al*. (2013)	5 (1)	0	23 (4)	18 (3)	0	17 (3)	1 (0)	0	3 (0)	0	2 (0)
Lung Cancer	Pope *et al*. (2002)	2 (1)	0	7 (2)	6 (2)	0	6 (2)	0 (0)	0	1 (0)	0	1 (0)
	Krewski *et al*. (2009)	1 (0)	0	5 (2)	5 (1)	0	5 (2)	0 (0)	0	1 (0)	0	0 (0)
	Cesaroni *et al*. (2013)	1 (0)	0	3 (1)	2 (1)	0	2 (1)	0 (0)	0	0 (0)	0	0 (0)

Table SI.6 - Avoidable deaths for the cities of Rio de Janeiro state (part 4), with the standard deviation in parentheses.

Health outcome	Reference	Rio de Janeiro						
		2017	**2016**	**2015**	**2014**	**2013**	**2012**	**2011**
All Causes	Pope *et al*. (2002)	889 (331)	889 (331)	595 (221)	1,157 (432)	1,144 (427)	562 (209)	1,725 (646)
	Krewski *et al*. (2009)	453 (152)	453 (152)	302 (102)	590 (199)	584 (196)	286 (96)	883 (298)
	Laden *et al.* (2006)	1,162 (332)	1,162 (332)	780 (222)	1,525 (438)	1,517 (435)	768 (2018)	2,339 (678)
Non-Accidental	Cesaroni *et al*. (2013)	563 (71)	563 (71)	376 (47)	735 (93)	726 (91)	355 (45)	1,095 (138)
	Crouse *et al*. (2012)	1,995 (97)	1,995 (97)	1,339 (65)	2,590 (127)	2,557 (126)	1,264 (61)	3,819 (190)
Cardiovascular	Cesaroni *et al*. (2013)	233 (39)	233 (39)	156 (26)	296 (49)	296 (49)	145 (24)	438 (73)
	Crouse *et al*. (2012)	589 (45)	589 (45)	395 (30)	745 (57)	742 (57)	367 (28)	1,090 (84)
	Laden *et al*. (2006)	494 (127)	494 (127)	333 (85)	628 (163)	614 (160)	314 (80)	925 (245)
Ischemic Heart Disease	Pope *et al*. (2004)	243 (29)	243 (29)	164 (19)	297 (36)	315 (38)	156 (19)	473 (58)
	Krewski *et al*. (2009)	206 (30)	206 (30)	138 (20)	252 (37)	267 (39)	132 (19)	402 (59)
	Crouse *et al*. (2012)	392 (23)	392 (23)	265 (16)	475 (29)	504 (31)	253 (15)	749 (47)
	Cesaroni *et al*. (2013)	142 (25)	142 (25)	95 (16)	173 (30)	184 (32)	91 (16)	278 (49)
Lung Cancer	Pope *et al*. (2002)	50 (16)	50 (16)	33 (11)	64 (21)	65 (21)	33 (11)	93 (31)
	Krewski *et al*. (2009)	40 (12)	40 (12)	27 (8)	52 (16)	52 (16)	26 (8)	75 (24)
	Cesaroni *et al*. (2013)	19 (8)	19 (8)	13 (6)	24 (11)	24 (11)	12 (5)	36 (16)

Table SI.7 - Avoidable deaths for the cities of Rio de Janeiro state (part 5), with the standard deviation in parentheses.

Health outcome	**Reference**	**São João de Meriti**						
		2017	**2016**	**2015**	**2014**	**2013**	**2012**	**2011**
All Causes	Pope *et al*. (2002)	20 (7)	116 (43)	39 (14)	95 (36)	151 (57)	37 (14)	113 (42)
	Krewski *et al*. (2009)	10 (3)	59 (20)	20 (7)	49 (16)	77 (26)	19 (6)	58 (20)
	Laden *et al*. (2006)	32 (9)	186 (54)	64 (18)	155 (45)	253 (74)	64 (18)	189 (55)
Non-Accidental	Cesaroni *et al*. (2013)	12 (2)	74 (9)	25 (3)	60 (8)	96 (12)	24 (3)	72 (9)
	Crouse *et al*. (2012)	44 (2)	257 (13)	88 (4)	211 (10)	332(17)	85 (4)	253 (13)
Cardiovascular	Cesaroni *et al*. (2013)	5 (1)	31 (5)	11 (2)	23 (4)	37 (6)	10 (2)	27 (4)
	Crouse *et al*. (2012)	14 (1)	78 (6)	27 (2)	58 (4)	93 (7)	25 (2)	67 (5)
	Laden *et al*. (2006)	14 (4)	80 (21)	28 (7)	59 (15)	96 (26)	27 (7)	70 (19)
Ischemic Heart Disease	Pope *et al*. (2004)	5 (1)	32 (4)	11 (1)	22 (3)	34 (4)	11 (1)	27 (3)
	Krewski *et al*. (2009)	5 (1)	27 (4)	9 (1)	18 (3)	29 (4)	9 (1)	23 (3)
	Crouse *et al*. (2012)	9 (1)	50 (3)	18 (1)	34 (2)	53 (3)	18 (1)	43 (3)
	Cesaroni *et al*. (2013)	3 (1)	19 (3)	6 (1)	13 (2)	20 (4)	6 (1)	16 (3)
Lung Cancer	Pope *et al*. (2002)	1 (0)	5 (2)	2 (1)	4 (1)	6 (2)	2 (1)	5 (2)
	Krewski *et al*. (2009)	1 (0)	4 (1)	1 (0)	3 (1)	5 (2)	1 (0)	4 (1)
	Cesaroni *et al*. (2013)	0 (0)	2 (1)	1 (0)	2 (1)	2 (1)	1 (0)	2 (1)

Table SI.8 - Avoidable deaths for the cities of Rio de Janeiro state (part 6), with the standard deviation in parentheses.

Health outcome	**Reference**	**Saropédica**				**Volta Redonda**	
		2014	**2013**	**2012**	**2011**	**2013**	**2011**
All Causes	Pope *et al*. (2002)	14 (5)	0	0	3 (1)	0	0
	Krewski *et al*. (2009)	7 (2)	0	0	1 (0)	0	0
	Laden *et al*. (2006)	24 (7)	0	0	5 (1)	0	0
Non-Accidental	Cesaroni *et al*. (2013)	8 (1)	0	0	2 (0)	0	0
	Crouse *et al*. (2012)	30 (1)	0	0	6 (0)	0	0
Cardiovascular	Cesaroni *et al*. (2013)	4 (1)	0	0	1 (0)	0	0
	Crouse *et al*. (2012)	11 (1)	0	0	2 (0)	0	0
	Laden *et al*. (2006)	10 (3)	0	0	2 (0)	0	0
Ischemic Heart Disease	Pope *et al*. (2004)	5 (1)	0	0	1 (0)	0	0
	Krewski *et al*. (2009)	4 (1)	0	0	1 (0)	0	0
	Crouse *et al*. (2012)	8 (0)	0	0	1 (0)	0	0
	Cesaroni *et al*. (2013)	3 (1)	0	0	0 (0)	0	0
Lung Cancer	Pope *et al*. (2002)	0 (0)	0	0	0 (0)	0	0
	Krewski *et al*. (2009)	0 (0)	0	0	0 (0)	0	0
	Cesaroni *et al*. (2013)	0 (0)	0	0	0 (0)	0	0

Table SI.9 - Avoidable deaths for the cities of São Paulo state (part 1), with the standard deviation in parentheses.

Health outcome	**Reference**	**Campinas**		**Guarulhos**	**Ribeirão Preto**
		2017	**2016**	**2017**	**2017**
All Causes	Pope *et al*. (2002)	266 (100)	303 (114)	320 (18)	73 (27)
	Krewski *et al*. (2009)	136 (46)	155 (53)	164 (55)	37 (12)
	Laden *et al*. (2006)	345 (101)	391 (115)	524 (153)	99 (28)
Non-Accidental	Cesaroni *et al*. (2013)	168 (21)	192 (24)	202 (26)	45 (6)
	Crouse *et al*. (2012)	583 (29)	662 (33)	700 (35)	160 (8)
Cardiovascular	Cesaroni *et al*. (2013)	76 (13)	87 (15)	105 (18)	18 (3)
	Crouse *et al*. (2012)	188 (15)	213 (17)	260 (20)	44 (3)
	Laden *et al*. (2006)	150 (40)	170 (46)	274 (74)	36 (9)
Ischemic Heart Disease	Pope *et al*. (2004)	98 (12)	111 (14)	126 (16)	20 (2)
	Krewski *et al*. (2009)	83 (12)	94 (14)	107 (16)	17 (2)
	Crouse *et al*. (2012)	154 (10)	174 (11)	198 (13)	33 (2)
	Cesaroni *et al*. (2013)	58 (10)	66 (12)	74 (13)	12 (2)
Lung Cancer	Pope *et al*. (2002)	14 (5)	16 (5)	17 (6)	5 (2)
	Krewski *et al*. (2009)	11 (4)	13 (4)	14 (4)	4 (1)
	Cesaroni *et al*. (2013)	5 (2)	6 (3)	6 (3)	2 (1)

Table SI.10 - Avoidable deaths for the cities of São Paulo state (part 2), with the standard deviation in parentheses.

Health outcome	**Reference**	**Piracicaba**					
		2017	**2016**	**2015**	**2014**	**2013**	**2012**
All Causes	Pope *et al*. (2002)	43 (16)	43 (16)	43 (16)	71 (27)	56 (21)	69 (26)
	Krewski *et al*. (2009)	22 (7)	22 (7)	22 (7)	36 (12)	29 (10)	35 (12)
	Laden *et al*. (2006)	61 (17)	61 (17)	58 (17)	98 (28)	78 (22)	96 (28)
Non-Accidental	Cesaroni *et al*. (2013)	28 (3)	28 (3)	27 (3)	45 (6)	35 (4)	43 (5)
	Crouse *et al*. (2012)	97 (5)	97 (5)	97 (5)	158 (8)	124 (6)	152 (8)
Cardiovascular	Cesaroni *et al*. (2013)	8 (1)	8 (1)	8 (1)	12 (2)	11 (2)	13 (2)
	Crouse *et al*. (2012)	20 (2)	20 (2)	19 (1)	30 (2)	28 (2)	32 (2)
	Laden *et al*. (2006)	18 (5)	18 (5)	15 (4)	23 (6)	23 (6)	24 (6)
Ischemic Heart Disease	Pope *et al*. (2004)	7 (1)	7 (1)	6 (1)	10 (1)	9 (1)	12 (1)
	Krewski *et al*. (2009)	6 (1)	6 (1)	5 (1)	8 (1)	8 (1)	10 (1)
	Crouse *et al*. (2012)	11 (1)	11 (1)	10 (1)	16 (1)	14 (1)	18 (1)
	Cesaroni *et al*. (2013)	4 (1)	4 (1)	4 (1)	6 (1)	5 (1)	7 (1)
Lung Cancer	Pope *et al*. (2002)	3 (1)	3 (1)	3 (1)	4 (1)	4 (1)	4 (1)
	Krewski *et al*. (2009)	2 (1)	2 (1)	2 (1)	4 (1)	3 (1)	3 (1)
	Cesaroni *et al*. (2013)	1 (0)	1 (0)	1 (0)	2 (1)	1 (1)	1 (1)

Table SI.11 - Avoidable deaths for the cities of São Paulo state (part 3), with the standard deviation in parentheses.

Health outcome	Reference	Santos					
		2017	**2016**	**2015**	**2014**	**2013**	**2012**
All Causes	Pope *et al*. (2002)	129 (48)	110 (41)	137 (51)	177 (66)	203 (76)	131 (49)
	Krewski *et al*. (2009)	66 (22)	56 (19)	70 (24)	91 (31)	104 (35)	67 (23)
	Laden *et al*. (2006)	139 (40)	119 (34)	154 (45)	199 (58)	236 (70)	153 (44)
Non-Accidental	Cesaroni *et al*. (2013)	84 (11)	71 (9)	89 (11)	115 (15)	131 (17)	85 (11)
	Crouse *et al*. (2012)	291 (14)	249 (12)	309 (15)	394 (20)	449 (23)	296 (15)
Cardiovascular	Cesaroni *et al*. (2013)	39 (7)	33 (6)	41 (7)	54 (9)	63 (11)	41 (7)
	Crouse *et al*. (2012)	97 (7)	83 (6)	102 (8)	132 (10)	155 (12)	103 (8)
	Laden *et al*. (2006)	64 (17)	55 (14)	73 (19)	90 (24)	108 (29)	73 (19)
Ischemic Heart Disease	Pope *et al*. (2004)	48 (6)	41 (5)	43 (5)	64 (8)	73 (9)	48 (6)
	Krewski *et al*. (2009)	41 (6)	35 (5)	37 (5)	54 (8)	63 (9)	41 (6)
	Crouse *et al*. (2012)	76 (5)	66 (4)	68 (4)	100 (6)	114 (7)	76 (5)
	Cesaroni *et al*. (2013)	28 (5)	24 (4)	25 (4)	38 (7)	44 (8)	28 (5)
Lung Cancer	Pope *et al*. (2002)	7 (2)	6 (2)	8 (3)	13 (4)	15 (5)	9 (3)
	Krewski *et al*. (2009)	6 (2)	5 (2)	7 (2)	10 (3)	12 (4)	7 (2)
	Cesaroni *et al*. (2013)	3 (1)	2 (1)	3 (1)	5 (2)	6 (3)	4 (2)

Table SI.12 - Avoidable deaths for the cities of São Paulo state (part 4), with the standard deviation in parentheses.

Health outcome	**Reference**	**São Bernardo do Campo**			**São José dos Campos**		**Taubaté**	
		2017	**2016**	**2015**	**2017**	**2016**	**2017**	**2016**
All Causes	Pope *et al*. (2002)	148 (55)	160 (60)	152 (57)	43 (16)	50 (18)	32 (12)	57 (21)
	Krewski *et al*. (2009)	76 (26)	82 (28)	78 (26)	22 (7)	25 (8)	16 (6)	29 (10)
	Laden *et al.* (2006)	223 (65)	241 (70)	225 (66)	64 (18)	73 (21)	46 (13)	80 (23)
Non-Accidental	Cesaroni *et al*. (2013)	94 (12)	102 (13)	96 (12)	27 (3)	31 (4)	21 (3)	36 (5)
	Crouse *et al*. (2012)	327 (16)	353 (18)	334 (17)	97 (5)	110 (5)	73 (4)	126 (6)
Cardiovascular	Cesaroni *et al*. (2013)	48 (8)	51 (9)	47 (8)	9 (1)	10 (2)	8 (1)	15 (2)
	Crouse *et al*. (2012)	119 (9)	128 (10)	118 (9)	23 (2)	26 (2)	21 (2)	37 (3)
	Laden *et al*. (2006)	110 (29)	118 (31)	111 (30)	20 (5)	23 (6)	19 (5)	33 (9)
Ischemic Heart Disease	Pope *et al*. (2004)	49 (6)	53 (7)	55 (7)	8 (1)	9 (1)	11 (1)	19 (2)
	Krewski *et al*. (2009)	42 (6)	45 (7)	47 (7)	7 (1)	8 (1)	9 (1)	16 (2)
	Crouse *et al*. (2012)	78 (5)	84 (5)	87 (5)	13 (1)	14 (1)	18 (1)	30 (2)
	Cesaroni *et al*. (2013)	29 (5)	31 (5)	32 (6)	5 (1)	5 (1)	6 (1)	11 (2)
Lung Cancer	Pope *et al*. (2002)	10 (3)	11 (4)	8 (3)	3 (1)	3 (1)	1 (0)	3 (1)
	Krewski *et al*. (2009)	8 (3)	9 (3)	6 (2)	2 (1)	3 (1)	1 (0)	2 (1)
	Cesaroni *et al*. (2013)	4 (2)	4 (2)	3 (1)	1 (0)	1 (1)	1 (0)	1 (0)

Table SI.13 - Avoidable deaths for the cities of São Paulo state (part 5), with the standard deviation in parentheses.

Health outcome	Reference	São Caetano do Sul (part 1)						
		2009	**2006**	**2005**	**2004**	**2003**	**2002**	**2001**
All Causes	Pope *et al*. (2002)	49 (18)	84 (32)	85 (32)	87 (33)	77 (29)	84 (32)	95 (36)
	Krewski *et al*. (2009)	25 (8)	43 (15)	44 (15)	45 (15)	40 (13)	43 (15)	49 (17)
	Laden *et al.* (2006)	56 (16)	95 (28)	100 (30)	106 (31)	101 (30)	112 (33)	132 (40)
Non-Accidental	Cesaroni *et al*. (2013)	32 (4)	56 (7)	56 (7)	57 (7)	50 (6)	55 (7)	63 (8)
	Crouse *et al*. (2012)	110 (6)	189 (10)	188 (10)	193 (10)	171 (9)	186 (10)	210 (11)
Cardiovascular	Cesaroni *et al*. (2013)	13 (2)	28 (5)	29 (5)	31 (5)	26 (4)	32 (5)	38 (7)
	Crouse *et al*. (2012)	33 (3)	68 (5)	69 (6)	75 (6)	64 (5)	77 (6)	93 (8)
	Laden *et al*. (2006)	23 (6)	42 (12)	45 (13)	51 (14)	49 (13)	62 (17)	75 (21)
Ischemic Heart Disease	Pope *et al*. (2004)	18 (2)	41 (5)	37 (5)	36 (5)	36 (5)	43 (5)	44 (6)
	Krewski *et al*. (2009)	15 (2)	35 (5)	32 (5)	31 (5)	31 (5)	37 (6)	38 (6)
	Crouse *et al*. (2012)	28 (2)	63 (4)	57 (4)	56 (4)	56 (4)	66 (4)	68 (5)
	Cesaroni *et al*. (2013)	10 (2)	25 (4)	22 (4)	22 (4)	22 (4)	26 (5)	27 (5)
Lung Cancer	Pope *et al*. (2002)	3 (1)	4 (1)	6 (2)	5 (2)	5 (2)	5 (2)	5 (2)
	Krewski *et al.* (2009)	2 (1)	3 (1)	5 (2)	4 (1)	4 (1)	4 (1)	4 (1)
	Cesaroni *et al*. (2013)	1 (0)	2 (1)	2 (1)	2 (1)	2 (1)	2 (1)	2 (1)

Table SI.14 - Avoidable deaths for the cities of São Paulo state (part 6), with the standard deviation in parentheses.

Health outcome	Reference	São Caetano do Sul (part 2)							
		2017	**2016**	**2015**	**2014**	**2013**	**2012**	**2011**	**2010**
All Causes	Pope *et al*. (2002)	69 (26)	59 (22)	76 (29)	35 (13)	65 (24)	77 (29)	99 (38)	73 (28)
	Krewski *et al*. (2009)	35 (12)	30 (10)	39 (13)	18 (6)	33 (11)	40 (13)	51 (17)	38 913)
	Laden *et al*. (2006)	68 (20)	58 (17)	78 (23)	36 (10)	65 (19)	77 (23)	101 (30)	72 (21)
Non-Accidental	Cesaroni *et al*. (2013)	45 (6)	39 (5)	50 (6)	23 (3)	43 (5)	51 (6)	65 (8)	48 (6)
	Crouse *et al*. (2012)	156 (8)	134 (7)	171 (9)	81 (4)	147 (7)	174 (9)	220 (11)	164 (8)
Cardiovascular	Cesaroni *et al*. (2013)	20 (3)	17 (3)	22 (4)	10 (2)	18 (3)	23 (4)	29 (5)	23 (4)
	Crouse *et al*. (2012)	49 (4)	42 (3)	54 (4)	26 (2)	44 (3)	57 (5)	71 (6)	55 (4)
	Laden *et al*. (2006)	27 (7)	24 (6)	35 (9)	16 (4)	30 (8)	36 (10)	46 (13)	34 (9)
Ischemic Heart Disease	Pope *et al*. (2004)	26 (3)	23 (3)	29 (4)	12 (2)	22 (3)	28 (3)	33 (4)	27 (3)
	Krewski *et al*. (2009)	23 (3)	19 (3)	25 (4)	11 (2)	19 (3)	24 (4)	29 (4)	23 (3)
	Crouse *et al*. (2012)	41 (3)	36 (2)	45 (3)	20 (1)	34 (2)	43 (3)	51 (3)	42 (3)
	Cesaroni *et al*. (2013)	16 (3)	13 (2)	17 (3)	7 (1)	13 (2)	17 (3)	20 (4)	16 (3)
Lung Cancer	Pope *et al*. (2002)	3 (1)	3 (1)	5 (2)	2 (1)	4 (1)	4 (1)	5 (2)	4 (1)
	Krewski *et al*. (2009)	3 (1)	2 (1)	4 (1)	2 (1)	3 (1)	3 (1)	4 (1)	3 (1)
	Cesaroni *et al*. (2013)	1 (1)	1 (1)	2 (1)	1 (0)	1 (1)	1 (1)	2 (1)	2 (1)

Table SI.15 - Avoidable deaths for the cities of São Paulo state (part 7), with the standard deviation in parentheses.

Health outcome	**Reference**	**São José do Rio Preto**								
		2017	**2016**	**2015**	**2014**	**2013**	**2011**	**2010**	**2009**	**2008**
All Causes	Pope *et al*. (2002)	105 (39)	93 (35)	70 (26)	100 (37)	65 (24)	24 (9)	60 (22)	13 (5)	52 (19)
	Krewski *et al*. (2009)	54 (18)	47 (16)	36 (12)	51 (17)	33 (11)	12 (4)	31 (10)	7 (2)	26 (9)
	Laden *et al*. (2006)	134 (39)	119 (34)	88 (25)	132 (38)	88 (25)	33 (9)	82 (23)	18 (5)	75 (22)
Non-Accidental	Cesaroni *et al*. (2013)	66 (8)	58 (7)	45 (6)	63 (8)	41 (5)	15 (2)	38 (5)	8 (1)	33 (4)
	Crouse *et al*. (2012)	230 (11)	204 (10)	157 (8)	218 (11)	145 (7)	54 (3)	135 (7)	30 (1)	116 (6)
Cardiovascular	Cesaroni *et al*. (2013)	28 (5)	25 (4)	19 (3)	27 (4)	17 (3)	7 (1)	18 (3)	4 (1)	16 (3)
	Crouse *et al*. (2012)	70 (5)	62 (5)	48 (4)	67 (5)	44 (3)	17 (1)	45 (3)	10 (1)	40 (3)
	Laden *et al*. (2006)	57 (15)	51 (13)	38 (10)	54 (14)	39 (10)	13 (3)	36 (9)	8 (2)	34 (9)
Ischemic Heart Disease	Pope *et al*. (2004)	32 (4)	28 (3)	22 (3)	33 (4)	22 (3)	7 (1)	20 (2)	4 (1)	17 (2)
	Krewski *et al*. (2009)	27 (4)	24 (4)	19 (3)	28 (4)	18 (3)	6 (1)	17 (2)	4 (1)	15 (2)
	Crouse *et al*. (2012)	51 (3)	45 (3)	36 (2)	53 (3)	35 (2)	11 (1)	32 (2)	7 (0)	28 (2)
	Cesaroni *et al*. (2013)	19 (3)	17 (3)	13 (2)	20 (3)	13 (2)	4 (1)	12 (2)	3 (0)	10 (2)
Lung Cancer	Pope *et al*. (2002)	5 (2)	5 (2)	4 (1)	5 (2)	4 (1)	1 (0)	3 (1)	1 (0)	3 (1)
	Krewski *et al*. (2009)	4 (1)	4 (1)	3 (1)	4 (1)	3 (1)	1 (0)	3 (1)	1 (0)	2 (1)
	Cesaroni *et al*. (2013)	2 (1)	2 (1)	2 (1)	2 (1)	2 (1)	0 (0)	1 (1)	0 (0)	1 (1)

Table SI.16 - Avoidable deaths for the cities of São Paulo state (part 8), with the standard deviation in parentheses.

Health outcome	**Reference**	**São Paulo (part 1)**						
		2006	**2005**	**2004**	**2003**	**2002**	**2001**	**2000**
All Causes	Pope *et al*. (2002)	3,216 (1,211)	3,751 (1,418)	3,405 (1,284)	2,767 (1,040)	3,937 (1,490)	4,035 (1,528)	4,284 (1,625)
	Krewski *et al*. (2009)	1,653 (559)	1,934 (656)	1,752 (593)	1,420 (480)	2,032 (689)	2,084 (707)	2,215 (752)
	Laden *et al.* (2006)	4,810 (1,420)	5,656 (1,688)	5,242 (1,553)	4,380 (1,286)	6,306 (1,888)	6,222 (1,987)	7,097 (2,137)
Non-Accidental	Cesaroni *et al*. (2013)	2,032 (258)	2,371 (303)	2,148 (273)	1,737 (220)	2,459 (314)	2,512 (321)	2,676 (342)
	Crouse *et al*. (2012)	6,976 (355)	8,051 (416)	7,357 (376)	6,001 (303)	8,343 (432)	8,517 (442)	9,056 (472)
Cardiovascular	Cesaroni *et al*. (2013)	1,016 (172)	1,201 (204)	1,123 (190)	909 (153)	1,317 (224)	1,367 (233)	1,481 (253)
	Crouse *et al*. (2012)	2,490 (198)	2,916 (235)	2,747 (219)	2,241 (176)	3,192 (258)	3,312 (268)	3,578 (291)
	Laden *et al*. (2006)	2,109 (576)	2,496 (694)	2,419 (664)	2,000 (540)	2,839 (793)	2,996 (840)	3,294 (929)
Ischemic Heart Disease	Pope *et al*. (2004)	1,210 (152)	1,391 (177)	1,355 (171)	1,133 (141)	1,605 (205)	1,643 (211)	1,783 (230)
	Krewski *et al*. (2009)	1,034 (156)	1,192 182)	1,159 (175)	967 (145)	1,376 (210)	1,409 (216)	1,531 (236)
	Crouse *et al*. (2012)	1,884 (122)	2,145 (143)	2,105 (138)	1,778 (114)	2,471 (166)	2,526 (170)	2,730 (186)
	Cesaroni *et al*. (2013)	720 (128)	833 (149)	808 (144)	672 (119)	963 (173)	988 (198)	1,074 (194)
Lung Cancer	Pope *et al*. (2002)	179 (61)	204 (70)	181 (62)	149 (51)	206 (71)	215 (74)	224 (78)
	Krewski *et al*. (2009)	145 (46)	165 (53)	146 (47)	120 (38)	166 (54)	174 (56)	182 (59)
	Cesaroni *et al*. (2013)	69 (31)	80 (36)	70 (32)	58 (26)	80 (37)	84 (38)	88 (40)

Table SI.17 - Avoidable deaths for the cities of São Paulo state (part 9), with the standard deviation in parentheses.

Health outcome	**Reference**	**São Paulo (part 2)**					
		2012	**2011**	**2010**	**2009**	**2008**	**2007**
All Causes	Pope *et al*. (2002)	2,657 (997)	3,786 (1,428)	2,707 (1,016)	1,657 (619)	2,434 (913)	3,448 (1,300)
	Krewski *et al*. (2009)	1,362 (460)	1,949 (660)	1,388 (469)	846 (285)	1,247 (421)	1,775 (601)
	Laden *et al.* (2006)	3,745 (1,093)	5,260 (1,559)	3,874 (1,132)	2,423 (698)	3,582 (1,044)	5,075 (1,504)
Non-Accidental	Cesaroni *et al*. (2013)	1,686 (213)	2,416 (307)	1,715 (217)	1,049 (132)	1,541 (195)	2,184 (278)
	Crouse *et al*. (2012)	5,847 (293)	8,262 (423)	5,945 (299)	3693 (182)	5,353 (268)	7,477 (382)
Cardiovascular	Cesaroni *et al*. (2013)	813 (137)	1,172 (198)	841 (142)	524 (88)	783 (132)	1,095 (185)
	Crouse *et al*. (2012)	2,015 (157)	2,867 (229)	2,082 (163)	1,314 (101)	1,942 (152)	2,676 (213)
	Laden *et al*. (2006)	1,691 (452)	2,336 (642)	1,718 (460)	1102 (288)	1,670 (446)	2,288 (628)
Ischemic Heart Disease	Pope *et al*. (2004)	974 (120)	1,361 (172)	1,000 (124)	617 (75)	943 (116)	1,319 (166)
	Krewski *et al*. (2009)	830 (123)	1,164 (176)	852 (127)	524 (77)	803 (119)	1,128 (17)
	Crouse *et al*. (2012)	1,536 (97)	2,116 (139)	1,575 (100)	986 (60)	1,488 (94)	2,050 (134)
	Cesaroni *et al*. (2013)	575 (101)	812 (145)	591 (104)	361 (63)	556 (98)	786 (14)
Lung Cancer	Pope *et al*. (2002)	158 (53)	218 (75)	155 (52)	98 (33)	143 (48)	206 (7)
	Krewski *et al*. (2009)	127 (40)	176 (56)	124 (39)	78 (25)	115 (36)	166 (53)
	Cesaroni *et al*. (2013)	60 (27)	85 (38)	59 (27)	37 (17)	55 (25)	80 (36)

Table SI.18 - Avoidable deaths for the cities of São Paulo state (part 10), with the standard deviation in parentheses.

Health outcome	**Reference**	**São Paulo (part 3)**				
		2017	**2016**	**2015**	**2014**	**2013**
All Causes	Pope *et al*. (2002)	2,509 (939)	2,630 (985)	3,120 (1,172)	3,508 (1,320)	2,312 (865)
	Krewski *et al*. (2009)	1,284 (433)	1,346 (454)	1,601 (541)	1,803 (610)	1,183 (399)
	Laden *et al.* (2006)	3,379 (980)	3,537 (1,028)	4,176 (1,223)	4,727 (1,393)	3,229 (936)
Non-Accidental	Cesaroni *et al*. (2013)	1,615 (204)	1,693 (214)	1,995 (253)	2,241 (285)	1,472 (186)
	Crouse *et al*. (2012)	5,634 (281)	5,899 (294)	6,893 (347)	7,702 (391)	5,136 (256)
Cardiovascular	Cesaroni *et al*. (2013)	775 (130)	812 (136)	950 (160)	1,066 (180)	716 (120)
	Crouse *et al*. (2012)	1,932 (150)	2023 (157)	2,352 (185)	2,625 (208)	1,787 (138)
	Laden *et al*. (2006)	1,609 (426)	1683 (447)	1,930 (519)	2,153 (586)	1,495 (395)
Ischemic Heart Disease	Pope *et al*. (2004)	904 (111)	946 (116)	1118 (139)	1,246 (156)	850 (104)
	Krewski *et al*. (2009)	769 (113)	805 (119)	954 (142)	1,064 (160)	724 (107)
	Crouse *et al*. (2012)	1438 (90)	1503 (94)	1,764 (112)	1,952 (126)	1352 (84)
	Cesaroni *et al*. (2013)	531 (93)	557 (98)	662 (117)	740 (131)	500 (88)
Lung Cancer	Pope *et al*. (2002)	144 (48)	151 (51)	189 (64)	205 (70)	138 (46)
	Krewski *et al*. (2009)	116 (37)	121 (38)	152 (48)	165 (53)	111 (35)
	Cesaroni *et al*. (2013)	55 (25)	58 (26)	73 (33)	79 (36)	53 (24)

Table SI.19 - Avoidable deaths for the cities of Minas Gerais, with the standard deviation in parentheses.

Health outcome	**Reference**	**Belo Horizonte**	
		2014	**2013**
All Causes	Pope *et al*. (2002)	292 (109)	172 (64)
	Krewski *et al*. (2009)	149 (50)	87 (29)
	Laden *et al.* (2006)	410 (117)	246 (70)
Non-Accidental	Cesaroni *et al*. (2013)	182 (23)	107 (13)
	Crouse *et al*. (2012)	640 (31)	380 (18)
Cardiovascular	Cesaroni *et al*. (2013)	67 (11)	40 (7)
	Crouse *et al*. (2012)	168 (13)	101 (8)
	Laden *et al*. (2006)	135 (35)	79 (20)
Ischemic Heart Disease	Pope *et al*. (2004)	47 (6)	30 (4)
	Krewski *et al*. (2009)	40 (6)	25 (4)
	Crouse *et al*. (2012)	76 (5)	48 (3)
	Cesaroni *et al*. (2013)	28 (5)	17 (3)
Lung Cancer	Pope *et al*. (2002)	16 (5)	9 (3)
	Krewski *et al*. (2009)	12 (4)	7 (2)
	Cesaroni *et al*. (2013)	6 (3)	3 (2)

APPENDIX II

Population

Figure SII.1 shows the total population per thousand inhabitants over 25 years old in each city investigated. Figure SII.2 shows the children population between 1 and 9 years old. Figure SII.3 presents the population over 1-year-old. The number is per thousand inhabitants.

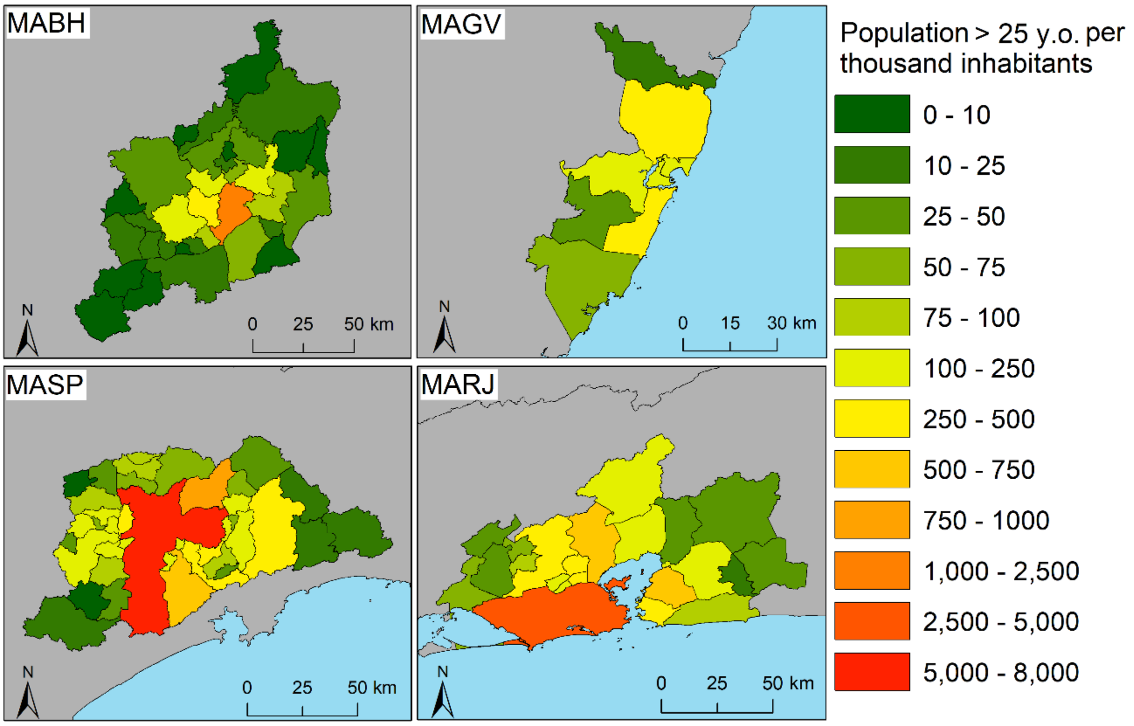

Figure SII.1 - Population over 25 years old per thousand inhabitants.

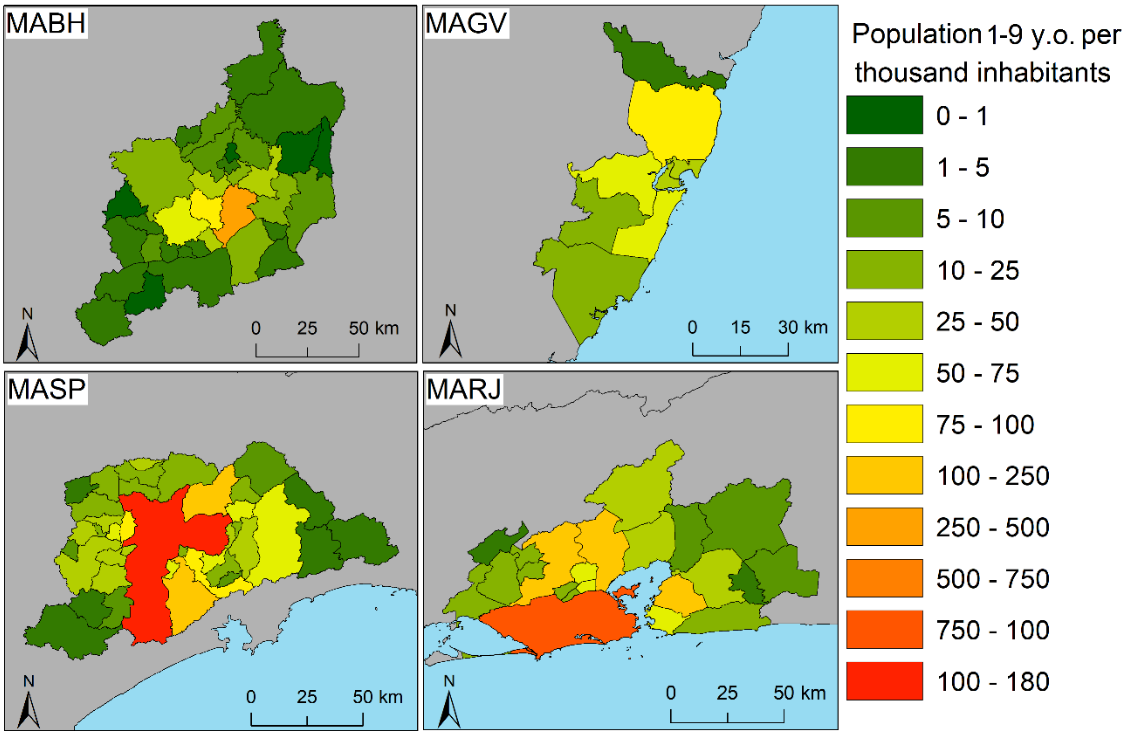

Figure SII.2 - Population between 1 and 9 years old per thousand inhabitants.

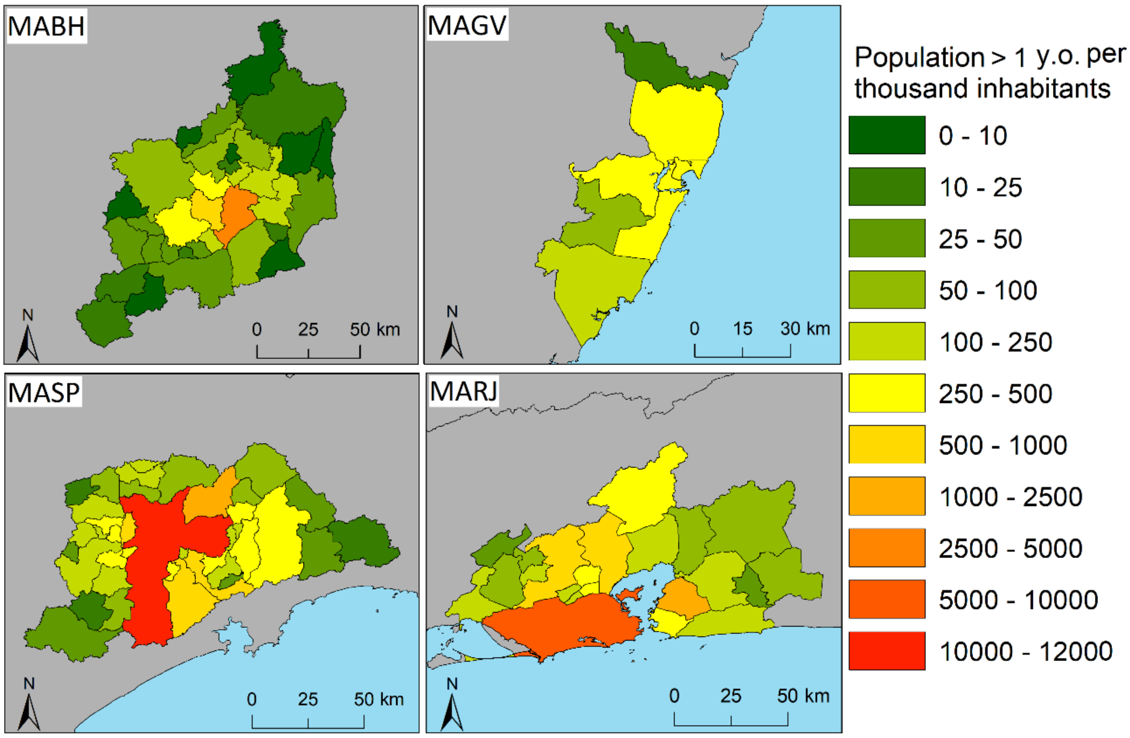

Figure SII.3 - Population over 1 year old per thousand inhabitants.

Incident Rate

Figure SII.4 shows the annual incident rate (2015) per 100,000 inhabitants for mortality due to all causes, non-accidental causes, cardiovascular, IHD, and lung cancer, for people above 25 years old. For all causes and non-accidental causes, the MARJ presented a higher incident rate than the other MA. MABH obtained larger variability. In this MA specifically, the cities that make up it are diverse, including small cities, distant from the capital, with a population fewer than 10,000 inhabitants (Figure SII.1), and populous cities, like those around the capital.

It can be observed that IHD is around half of the cardiovascular incident rate. For Lung cancer, all four MA presented similar results, with the median incident rate varying from 15 to 20 thousand. For this outcome, the capitals presented higher values than the median.

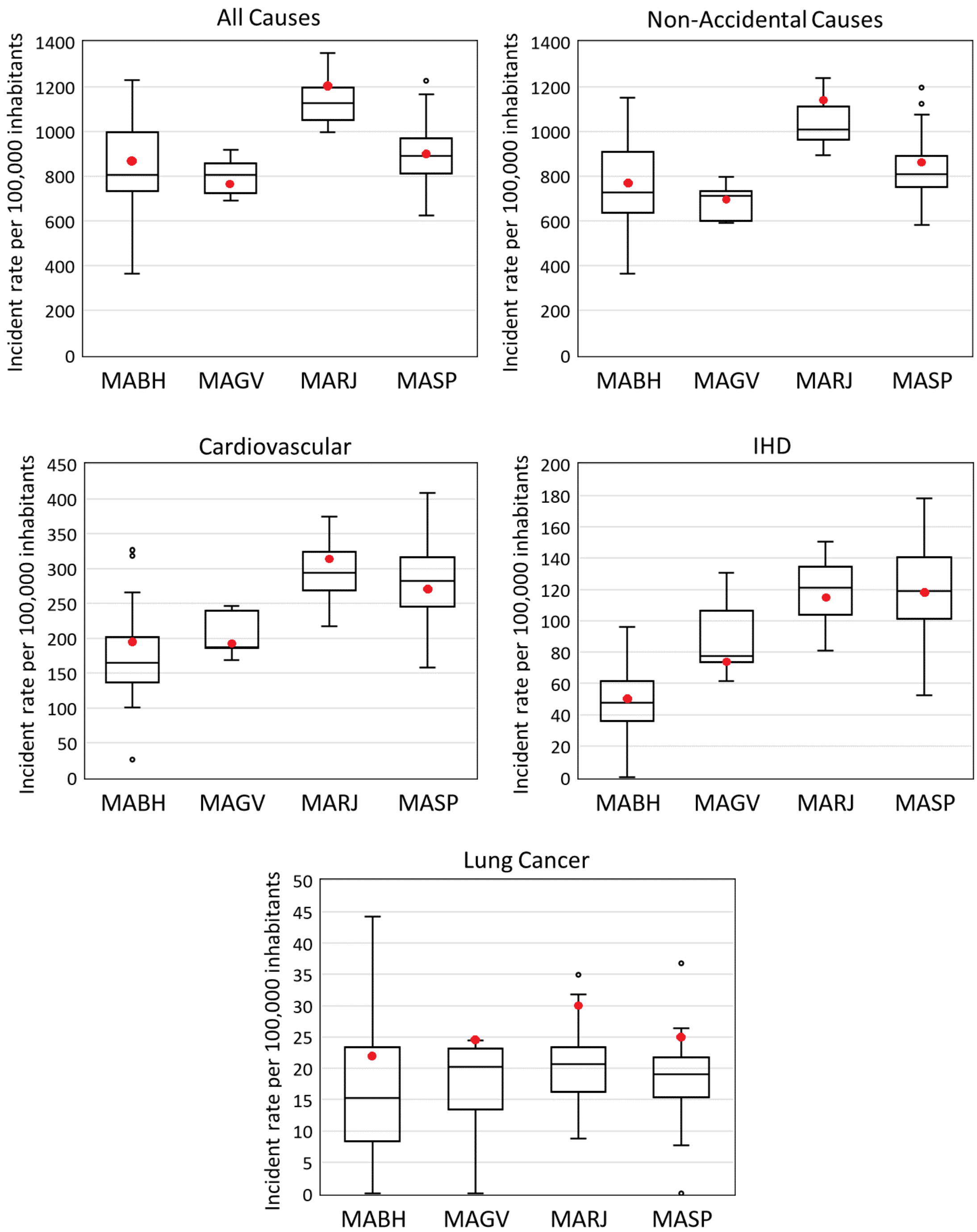

Figure SII.4 - Annual Incident rate (2015) per 100,000 inhabitants for people above 25 years old for MABH, MAGV, MARJ, and MASP. Red dots represent the incident rate for the capitals: Belo Horizonte (MABH), Vitoria (MAGV), Rio de Janeiro (MARJ), and São Paulo (MASP).

Figure SII.5 presents the monthly incident rate for children (1-9 years old) for respiratory diseases (ICD-10: chapter X) per 1,000 inhabitants for the four MA. Overall, during the dry season (April-September), there is an increase in the incident rate. In MABH, MAGV, and MASP, the capital presented an incident rate above the median. In MARJ, for most of the year, the incident rate was below the median.

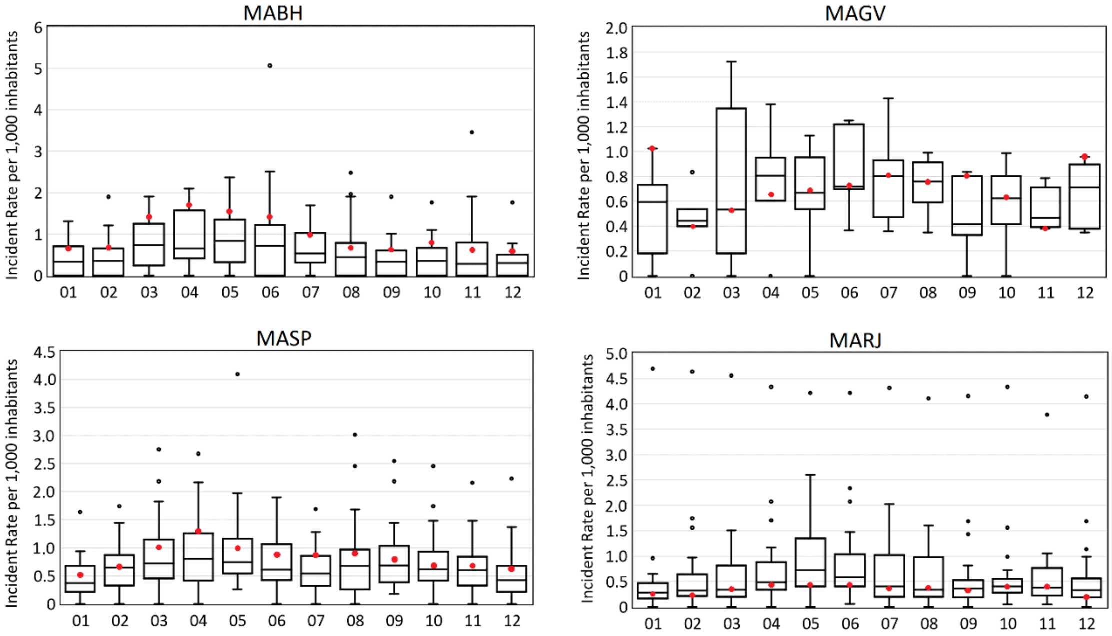

Figure SII.5 - Monthly incident rate per 1,000 inhabitants for respiratory diseases (ICD-10: chapter X) for children between 1 and 9 years old for MABH, MAGV, MARJ, and MASP. Red dots represent the incident rate for the capitals: Belo Horizonte (MABH), Vitoria (MAGV), Rio de Janeiro (MARJ), and São Paulo (MASP).

Figure SII.6 shows the monthly incident rate for people > 1-year old for respiratory diseases (ICD-10: chapter X) per 1,000 inhabitants for the four MA. Overall, during wintertime, the incident rate increases. Belo Horizonte presented an incident rate higher than the other capitals. Rio de Janeiro, otherwise, present lower values. About half of the cities present a higher incident rate than the capitals in each MA.

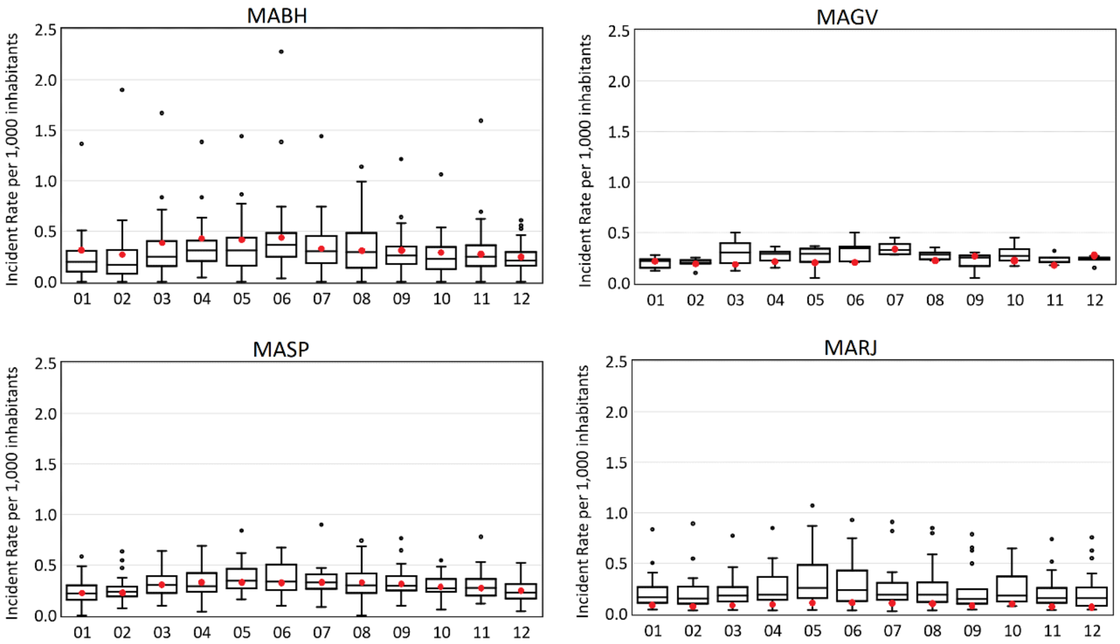

Figure SII.6 - Monthly incident rate per 1,000 inhabitants for respiratory diseases (ICD-10: chapter X) for people over 1-year-old for MABH, MAGV, MARJ, and MASP. Red dots represent the incident rate for the capitals: Belo Horizonte (MABH), Vitoria (MAGV), Rio de Janeiro (MARJ), and São Paulo (MASP).

Figure SII.7 presents the monthly incident rate for the circulatory system diseases (ICD-10: chapter IX) for the elderly (>65 years old) per 1,000 inhabitants, for the four MA. Overall, during the dry season (April-September), there is an increase in the incident rate. In MABH and MAGV, the capital (red dots in Figure SII.7) presented higher values than the other cities in most of the year. In MASP and MARJ, the capitals presented an incident rate slighted above the median.

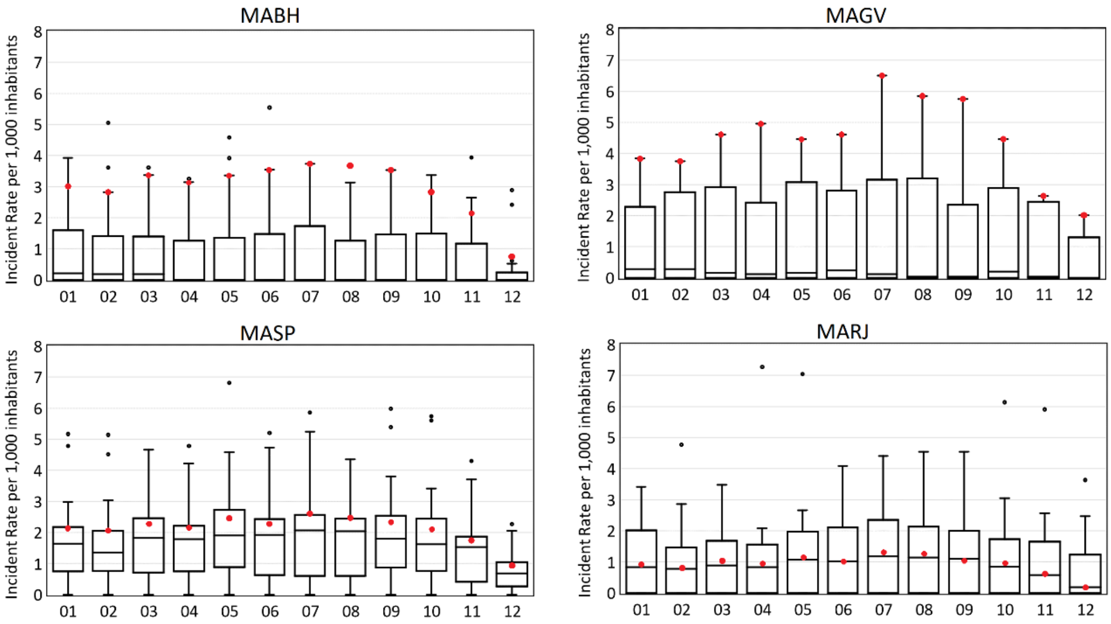

Figure SII.7 - Monthly incident rate per 1,000 inhabitants for circulatory system diseases (ICD-10: chapter IX) for the elderly (>65 years old) per 1,000 inhabitants, for MABH, MAGV, MARJ, and MASP. Red dots represent the incident rate for the capitals: Belo Horizonte (MABH), Vitoria (MAGV), Rio de Janeiro (MARJ), and São Paulo (MASP).

Meteorological validation

For modeling validation, hourly meteorological parameters (temperature, specific humidity, wind speed, and wind direction) were compared to 32 automatic meteorological monitoring stations of the National Institute of Meteorology in Brazil (INMET) (SM - Figure SII.8), using the statistical indices and benchmarks suggested by Emery et al. (2001): Mean Bias (MB); Mean Error (ME); Root Mean Square Error (RMSE); and Index of Agreement (IOA). The circular correlation coefficient (CCC) was also calculated for wind direction. To evaluate the performance of the meteorological model simulations, the resulting criteria for complex conditions suggested by Ramboll (2018) and LADCO and WDNR (2018) was used. For those benchmarks that did not present criteria for complex conditions, the criteria for simple conditions suggested by Emery et al. (2001) was used instead.

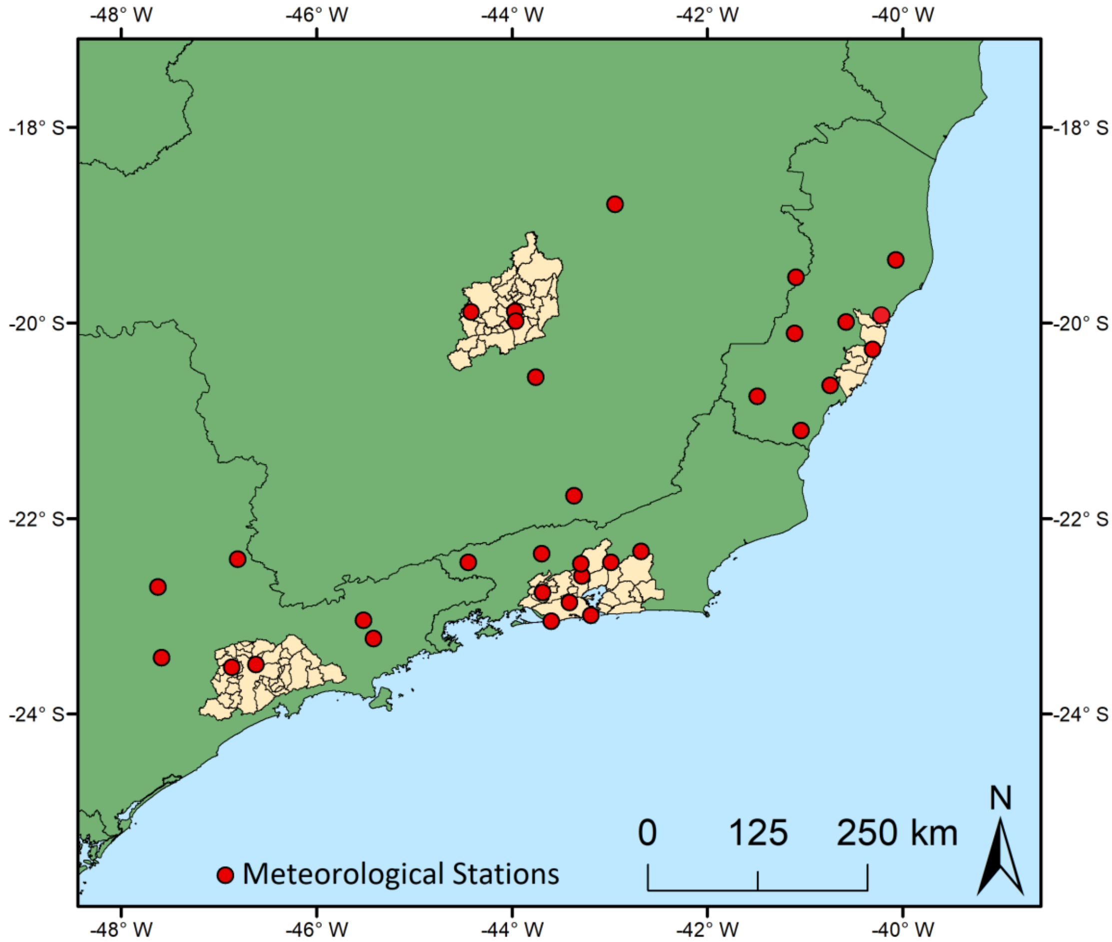

Figure SII.8 - Location of the 32 meteorological stations used to validate the meteorology.

The statistical indices and benchmarks obtained from the comparison between modelled and monitored meteorological parameters are presented in Figure SII.9. Most of the surface stations agreed with the modelled specific humidity and temperature for all four MA. A smaller variability on the indices was observed for MABH, the metropolitan area with better benchmarks.

The median wind speed was overestimated in all MA. Despite this tendency, also reported in the literature (SHIMADA *et al*., 2011; ALBUQUERQUE *et al*., 2018; ANDREÃO *et al*., 2020), the modelled wind speed values agreed with most of the surface stations, within benchmarks, except for IOA, where the median values were around between 0.5 and 0.6.

The wind direction, one of the most complex indicators to represent in complex situations, presented better indices for MABH. For MARJ and MASP, otherwise, a large variability was observed, with a smaller number of monitoring stations agreeing with the modeling.

From the MB index, it is notable that the model tends to move the wind direction anticlockwise, compared to the observed wind direction, in particular for MARJ and MASP, which may have contributed to poor CCC values. Jiménez and Dudhia (2013) showed that modelled wind direction depends on wind speed, being easier to represent the wind direction for higher wind speeds. A larger variability of wind direction was also observed in monitoring data, which contributes to the difficulty of its representation. The wind roses comparing the modelled and observed wind direction are presented in Figures SII.10 to SII.13.

The same meteorological parametrizations were used for the four MA (Table 5.3). Considering the complex topography of the MA simulated, varying from sea level to areas with more than 1,000 m, it can be interpreted from the meteorological validation an adequate representation of the conditions for MABH and MAGV, and reasonable representation for MARJ and MASP, especially when considering a comparison between values from a point (surface meteorological monitoring station) and an area of 25 km^2 (grid cell where the monitoring station is located).

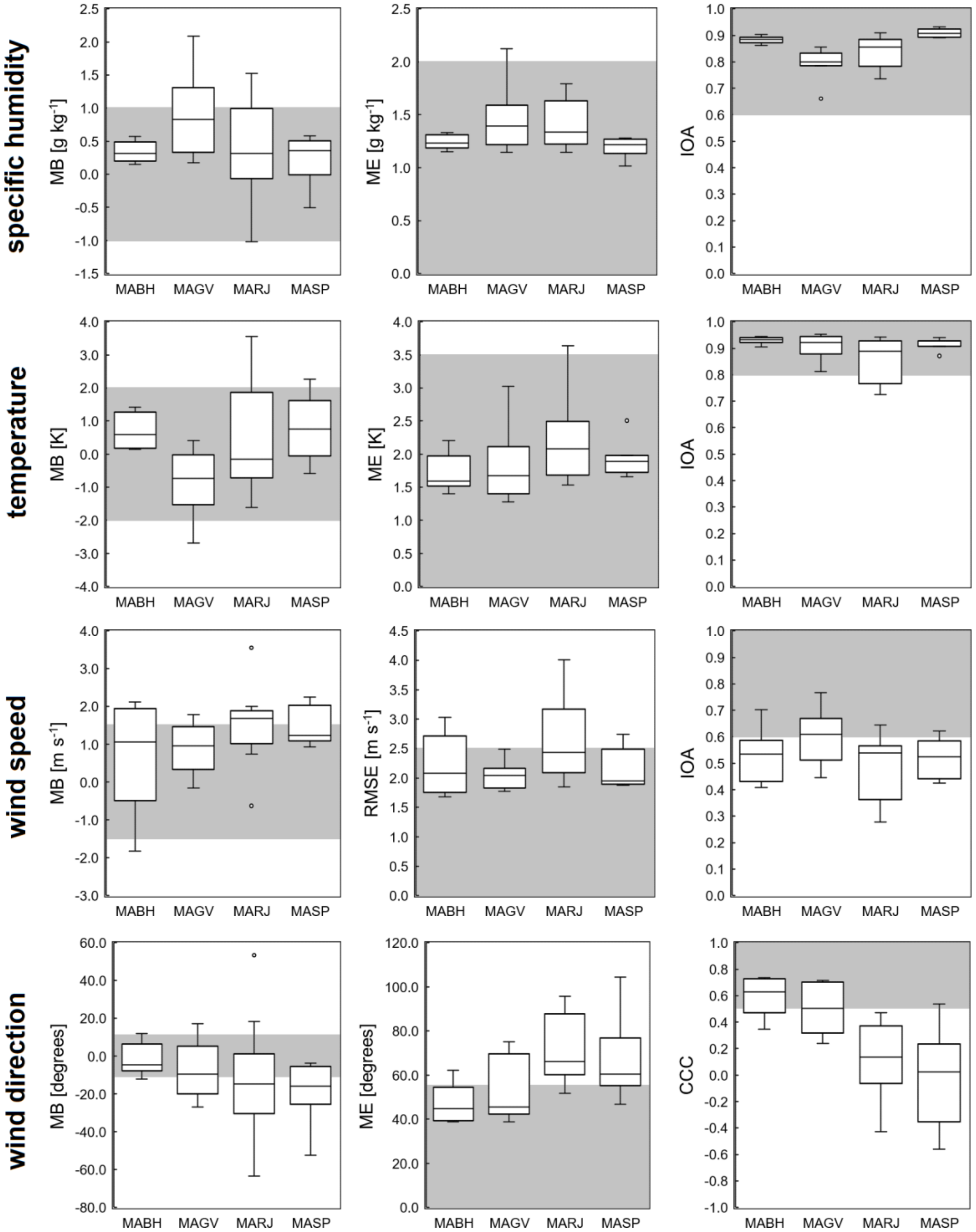

Figure SII.9 - Meteorological WRF validation against surface stations. The gray area represents the criteria benchmark. MB wind direction and IOA benchmarks are for simple conditions. The CCC benchmark is based on Andreão et al. (2020).

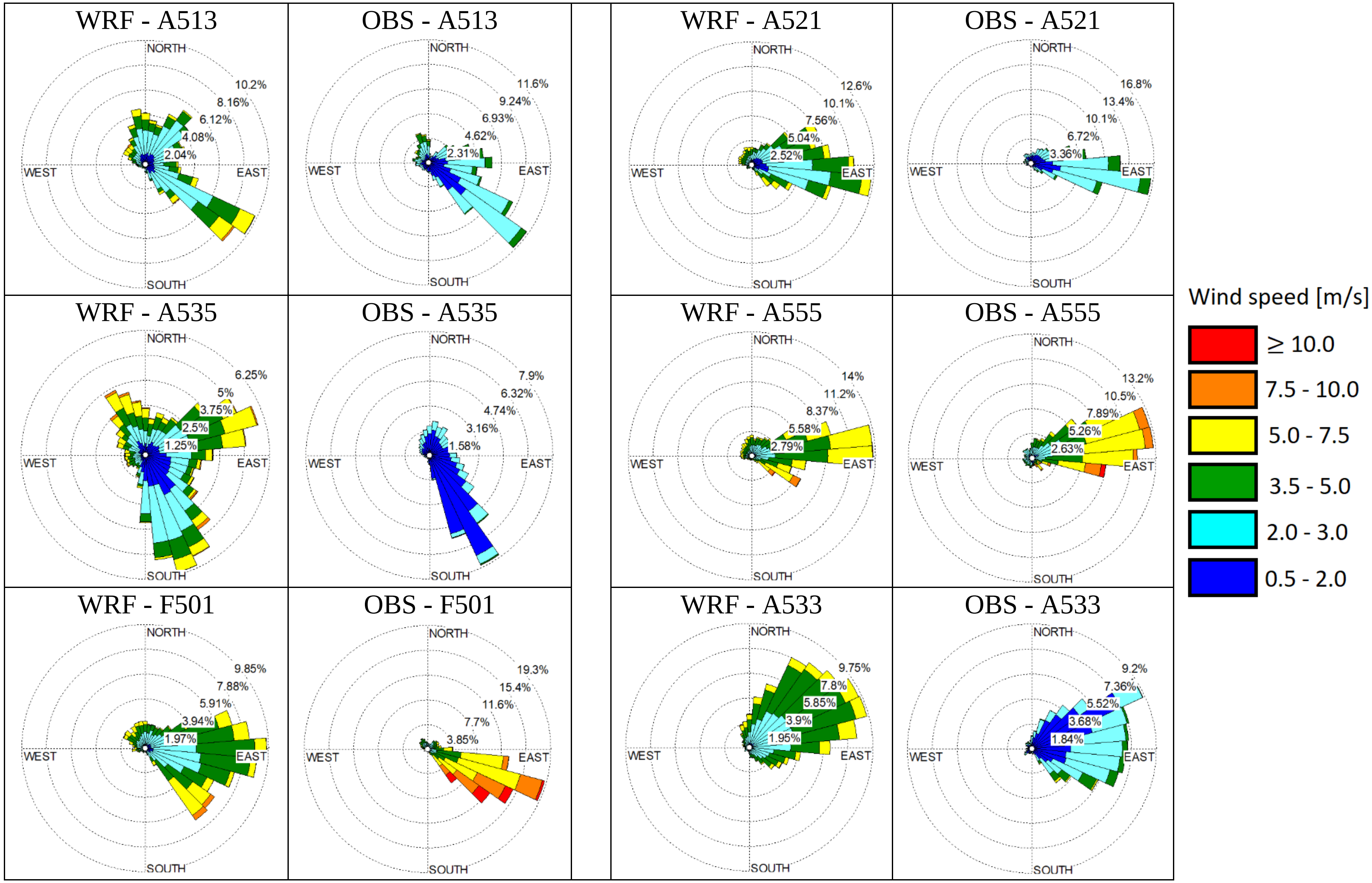

Figure SII.10 - Wind rose for modelled and observed wind speed and wind direction for six MABH monitoring stations.

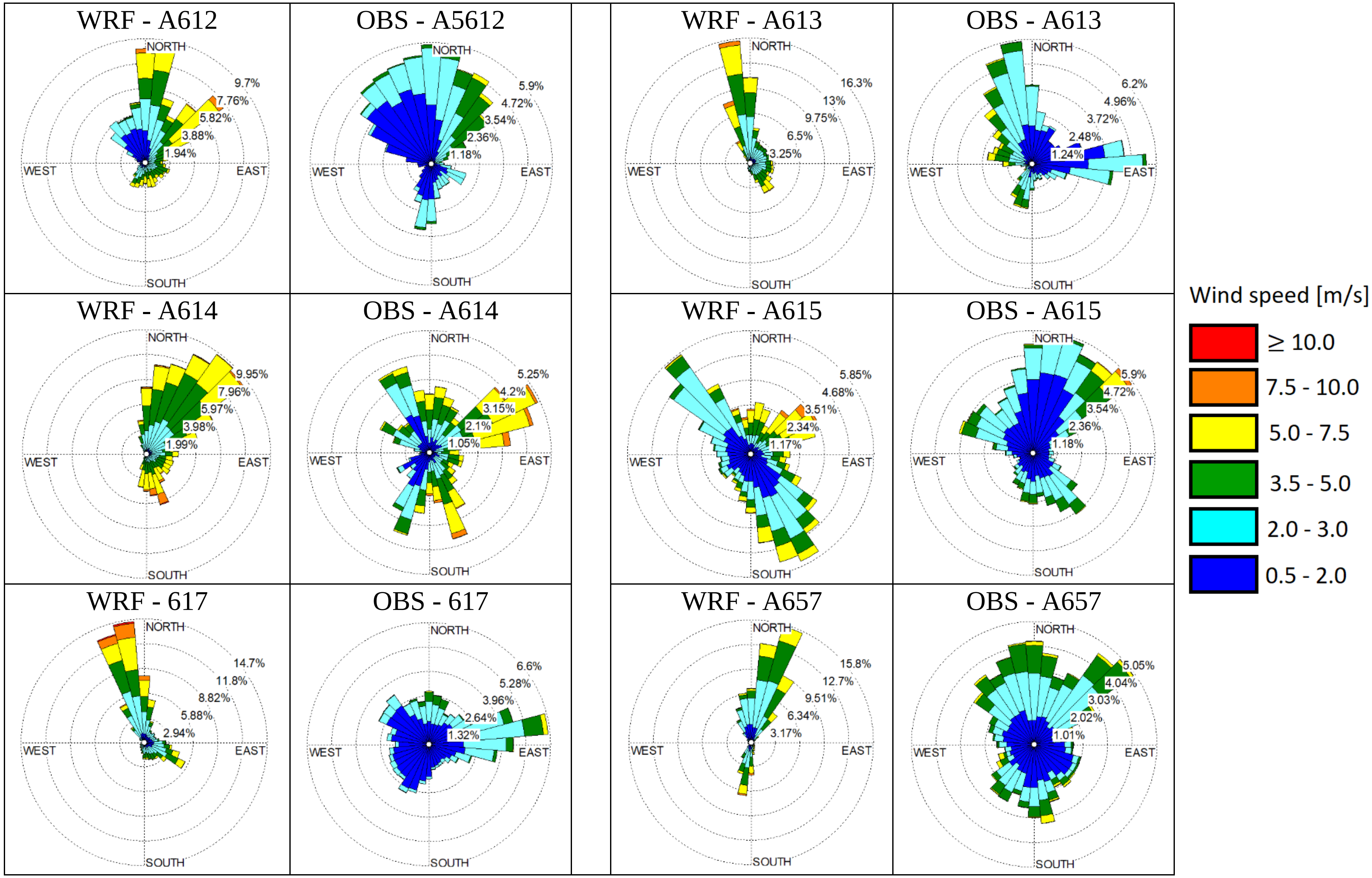

Figure SII.11 - Wind rose for modelled and observed wind speed and wind direction for six MAGV monitoring stations.

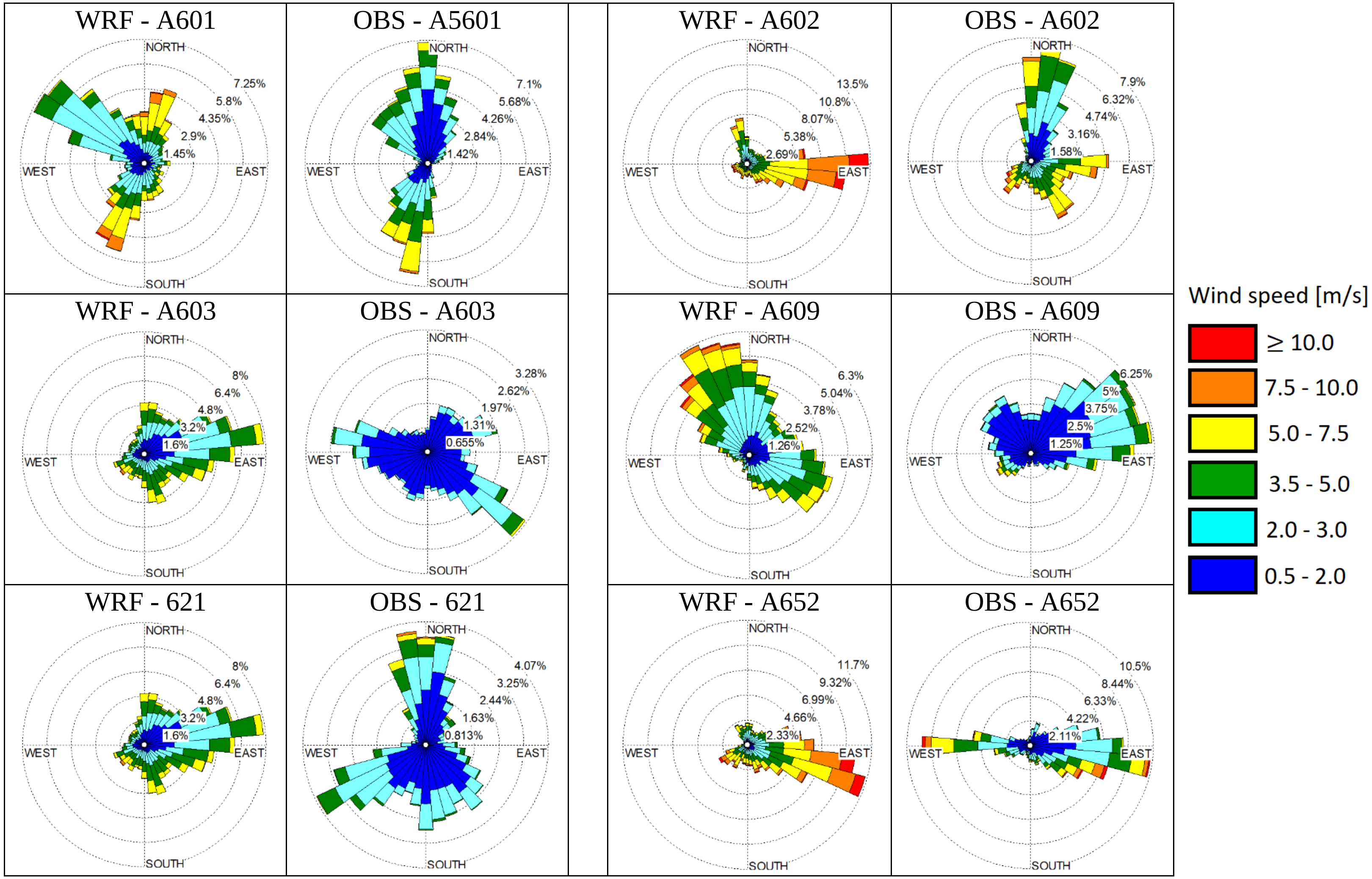

Figure SII.12 - Wind rose for modelled and observed wind speed and wind direction for six MARJ monitoring stations.

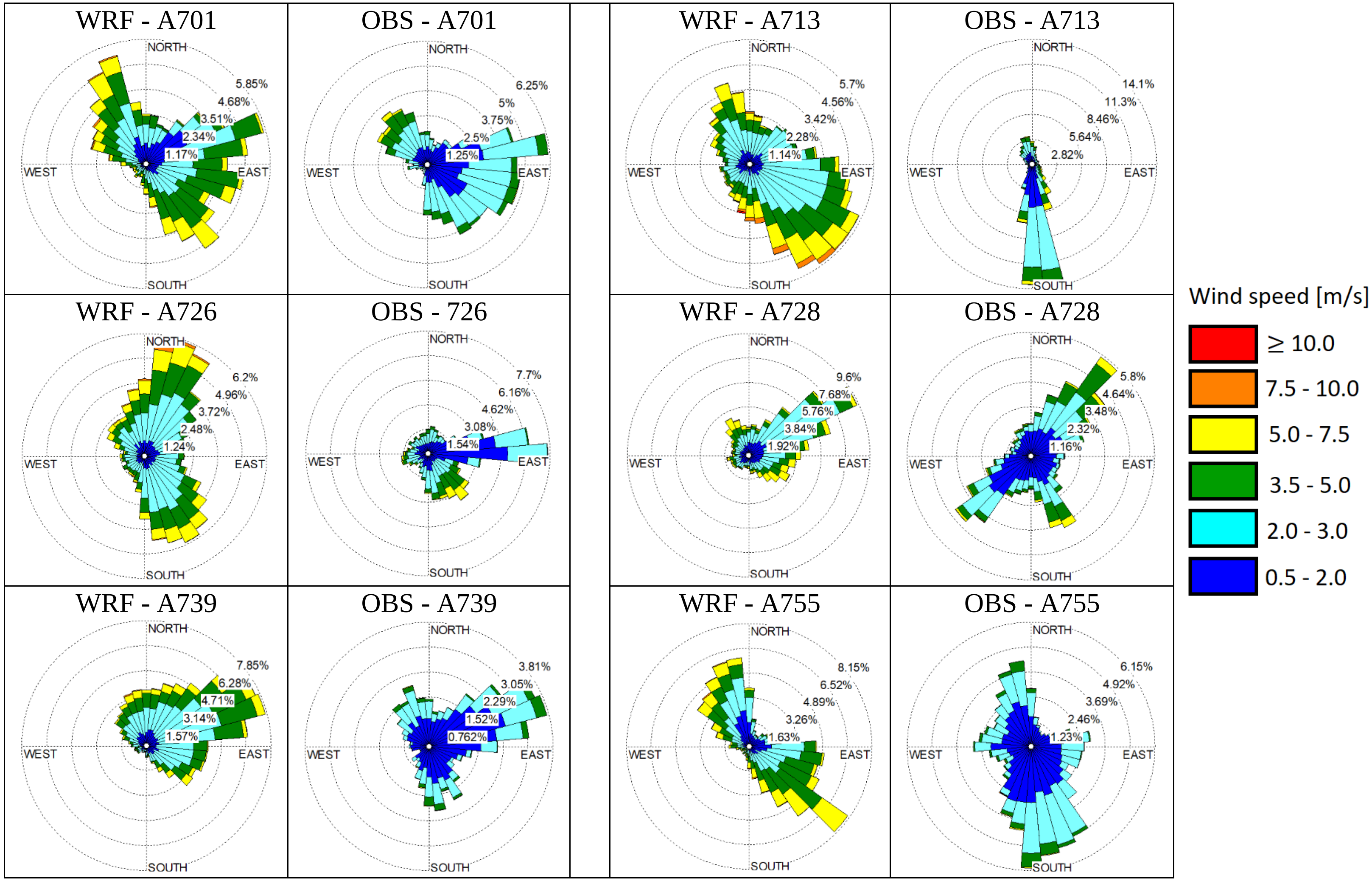

Figure SII.13 - Wind rose for modelled and observed wind speed and wind direction for six MASP monitoring stations.

For precipitation, Figure SII.14 shows a comparison between modelled and observed monthly accumulated rainfall for the four MA. Overall, WRF-Chem tends to overestimate the rainfall during wet-season, where it can be observed a large variability among the meteorological station's grid cells (WRF in x-axis in Figure SII.14). The observed rainfall was better represented during the dry season. A clear seasonality can be observed in MABH, MARJ, and MASP, which was represented by WRF-Chem. In MAGV, January was unusual dry. A larger difference between modelled and observed data was obtained in February, March, May, November, and December.

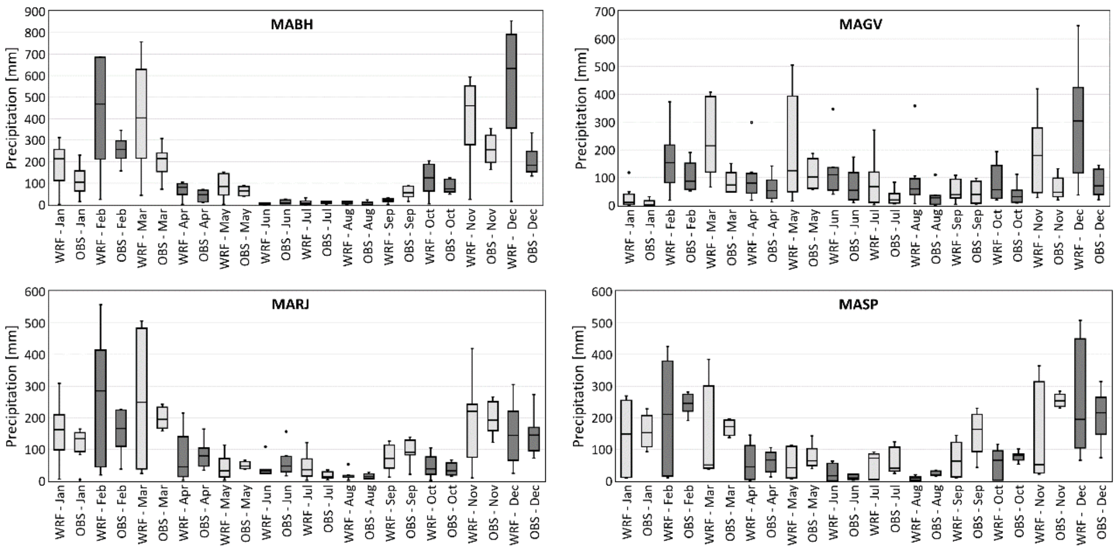

Figure SII.14 - Monthly accumulated rainfall for MABH, MAGV, MARJ, and MASP.