

UNIVERSIDADE FEDERAL DE MINAS GERAIS
Escola de Engenharia
Programa de Pós-Graduação em Saneamento, Meio Ambiente e Recursos Hídricos

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**FROM GROUND-LEVEL MONITORING TO REMOTE SENSING: A Comprehensive
Assessment of Air Quality During COVID-19 Mobility Restrictions**

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Assessment of Air Quality During COVID-19 Mobility Restrictions**

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Às 14:00 horas do dia 30 de junho de 2023, reuniu-se na Escola de Engenharia da UFMG a Comissão Examinadora de TESE DE DOUTORADO, indicada pelo Colegiado do PROGRAMA DE PÓS-GRADUAÇÃO EM SANEAMENTO, MEIO AMBIENTE E RECURSOS HÍDRICOS, para julgar, em exame final, o trabalho final intitulado:

"FROM GROUND-LEVEL MONITORING TO REMOTE SENSING: A COMPREHENSIVE ASSESSMENT OF AIR QUALITY DURING COVID-19 MOBILITY RESTRICTIONS"

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Abrindo a sessão, a Orientadora, Prof^a. Taciana Toledo de Almeida Albuquerque, após dar conhecimento aos presentes o teor das Normas Regulamentares do Trabalho Final, passou a palavra ao candidato para apresentação de seu trabalho. Seguiu-se a arguição pelos examinadores com a respectiva defesa pelo candidato. Logo após, a Comissão se reuniu sem a presença do candidato e do público, para julgamento e expedição do resultado final. Foram atribuídas as seguintes indicações:

BANCA EXAMINADORA	INSTITUIÇÃO	INDICAÇÃO
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Prof ^a . Dra. Judith Johanna Hoelzemann	UFRN	APROVADO

Prof. Dr. Thiago Nogueira	USP	APROVADO
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Pelas indicações, o candidato foi considerado APROVADO, devendo proceder a pequenas alterações no trabalho, para a sua edição definitiva, a ser entregue no prazo de 2 meses.

O resultado final foi comunicado publicamente ao candidato pelo Presidente da Comissão. Nada mais havendo a tratar, o Presidente encerrou a reunião e lavrou a presente ATA, que será assinada por todos os membros participantes da Comissão Examinadora. Belo Horizonte, 30 de junho de 2023.

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ABSTRACT

The mobility restrictions implemented to control COVID-19, in addition to reducing disease transmission rates, resulted in improved air quality. Thus, given this scenario, this work aims to assess how mobility restrictions during the pandemic affected air quality in Brazil. In addition, the research seeks to determine how capable remote sensors are to monitor air quality in this region. For this purpose, an analysis of the impact of mobility restrictions on air quality was carried out considering the pollutants particulate matter (PM), carbon monoxide (CO), sulfur dioxide (SO₂), nitrogen oxides (NO_x), and ozone (O₃). Mobility trend and atmospheric conditions data sets were also analyzed. For the evaluation of the sensor, tropospheric nitrogen dioxide (NO₂) density data obtained from the TROPospheric Monitoring Instrument (TROPOMI) sensor and Aerosol Optical Depth (AOD) from the Moderate-Resolution Imaging Spectroradiometer (MODIS) sensor were considered, seeking to evaluate their relationship with concentrations of NO₂ and PM, respectively. Furthermore, AOD data from the Multi-Angle Implementation of Atmospheric Correction (MAIAC) were compared with those retrieved by the Dark Target (DT) and Deep Blue (DB) algorithms. The assessment of mobility restrictions in Brazil showed that, in general, there was an improvement in air quality during this period. During the first 30 days of restrictions, a substantial reduction in primary pollutants was observed, with CO concentrations decreasing by around 40% from average levels from 2015 to 2019. On the other hand, there was no substantial decrease in atmospheric pollutants for the State of São Paulo. Significant reductions were observed mainly in areas impacted by vehicle emissions, as is the case of the Metropolitan Region of São Paulo, which reached an average decrease of 29% in CO, 28% in NO_x, 40% in NO, 19% in SO₂, 15% in PM_{2.5} and 8% in PM₁₀ concentrations. The atypical meteorological conditions between the evaluated years prevented assertively establishing the real impact of mobility restrictions on air quality in São Paulo. As for the remote sensing information, the AOD data obtained through the MAIAC algorithm showed low and non-significant correlations with MP ($R < 0.2$ and $\alpha > 0.05$) for the state of São Paulo. For tropospheric NO₂, good correlations were observed with ground-level concentrations; some stations even presented correlations greater than 0.80. The correlations between the NO₂ data were generally stronger in densely populated places, consequently presenting more significant variability in the measured values. The evaluation of AOD retrieval algorithms revealed that all products display varying levels of dependence on land cover types, view geometry, and spatiotemporal aerosol loads. Overall, DB and MAIAC algorithms exhibited greater accuracy across a broader spectrum of land cover types. However, it is essential to note that these algorithms did not perform well in representing the urban class. Finally, the study shows that mobility restrictions had a significant positive impact on reducing air pollutants, highlighting the potential importance of remote sensing products as a crucial tool for supporting air quality monitoring.

Keywords: MODIS. TROPOMI. Aerosol optical depth. Mobility restriction. Air quality. COVID-19.

RESUMO

As restrições de mobilidade implementadas para o controle da COVID-19 além de reduzirem as taxas de transmissão da doença, resultaram na melhora da qualidade do ar. Assim, dado o eventual cenário, o objetivo deste trabalho é avaliar de qual forma as restrições de mobilidade, durante o período de pandemia, afetaram a qualidade do ar no Brasil. Além disso, o trabalho busca averiguar quão hábeis são os sensores remotos para monitorar a qualidade do ar nesta região. Para isso, foi realizada a análise do impacto das restrições de mobilidade na qualidade do ar considerando os poluentes material particulado (MP), monóxido de carbono (CO), dióxido de enxofre (SO₂), óxidos de nitrogênio (NO_x) e ozônio (O₃). Também foram analisados os dados de tendência de movimentação e de condições atmosféricas. Para a avaliação dos sensores, foram considerados dados de densidade de dióxido de nitrogênio (NO₂) troposférico obtidos através do sensor TROPOspheric Monitoring Instrument (TROPOMI) e profundidade óptica do aerossol (do inglês Aerosol Optical Depth - AOD) do sensor Moderate-Resolution Imaging Spectroradiometer (MODIS), buscando avaliar sua relação com concentrações de NO₂ e MP, respectivamente. Além disso, dados de AOD do Multi-Angle Implementation of Atmospheric Correction (MAIAC) foram comparados com aqueles recuperados pelos algoritmos Dark Target (DT) e Deep Blue (DB). A avaliação das restrições de mobilidade no Brasil demonstrou que, em geral, houve melhora na qualidade do ar durante este período. Durante 30 dias de restrições, observou-se uma redução substancial dos poluentes primários, com as concentrações de CO diminuindo cerca de 40% em relação aos níveis médios de 2015 a 2019. Por outro lado, não houve diminuição substancial dos poluentes atmosféricos para o Estado de São Paulo. As reduções significativas foram observadas principalmente nas áreas impactadas por emissões veiculares, como é o caso da Região Metropolitana de São Paulo, que atingiu uma redução média de 29% em CO, 28% em NO_x, 40% em NO, 19% em SO₂, 15% em MP_{2,5} e 8% nas concentrações de MP₁₀. As condições meteorológicas atípicas entre os anos avaliados impediram estabelecer assertivamente o real impacto das restrições de mobilidade na qualidade do ar no estado. Quanto as informações de sensoriamento remoto avaliadas, os dados de AOD obtidos através do algoritmo MAIAC apresentaram baixas, e não-significativas, correlações com MP ($R < 0,2$ e $\alpha > 0,05$) para o estado de São Paulo. Para o NO₂ troposférico, observou-se boas correlações com concentrações a nível de solo, algumas estações chegaram a apresentar correlações maiores que 0.80. De forma geral, as correlações entre os dados de NO₂ foram mais fortes em locais mais densamente povoados e que conseqüentemente apresentam maior variabilidade nos valores medidos. A avaliação dos algoritmos de recuperação de AOD revelou que todos os produtos exibem níveis variados de dependência dos tipos de cobertura do solo, geometria da vista e cargas de aerossóis espaço-temporais. No geral, os algoritmos DB e MAIAC exibiram maior precisão em um espectro mais amplo de tipos de cobertura da terra. No entanto, é essencial observar que esses algoritmos não tiveram um bom desempenho na representação da classe urbana. Por fim, o estudo mostra que as restrições de mobilidade tiveram um impacto positivo significativo na redução de poluentes atmosféricos, destacando o valor potencial dos produtos de sensoriamento remoto como uma ferramenta crucial para apoiar o monitoramento da qualidade do ar.

Palavras-chave: MODIS. TROPOMI. Profundidade óptica do aerossol. Restrição de mobilidade. Qualidade do ar. COVID-19.

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1 INTRODUCTION

The COVID-19 pandemic caused by the Sars-CoV-2 virus has led to one of the most significant global health crises in recent history (PAK et al., 2020; THE LANCET, 2020b). The highly infectious nature of the virus through the air demanded a worldwide alert for measures to control its spread. The disease's rapid spread in a given region was accompanied by a concomitant crisis in the healthcare system, leading to the adoption of various measures such as the use of masks and the reduction of collective activities through social distancing (BLUMENTHAL et al., 2020; DE OLIVEIRA ANDRADE, 2020). Mobility restrictions were found to be effective in controlling the transmission of the virus and resulted in a reduction in air pollution in several regions (WANG et al., 2020b). These measures have been implemented globally and have provided a unique opportunity to study the impact of reduced human activities on air quality. Recent studies have shown a positive correlation between mobility restrictions and air quality improvement, highlighting the need for continued efforts to reduce air pollution for long-term public health benefits (BAUWENS et al., 2020; CHOSSIÈRE et al., 2021; HE; WANG; YIM, 2021; ZAMBRANO-MONSERRATE; RUANO; SANCHEZ-ALCALDE, 2020).

The reduction of air pollutants directly impacts human life since the deterioration of air quality is associated with high morbidity and mortality rates from cardiovascular and respiratory diseases (BEELEN et al., 2014; CHEN; GOLDBERG; VILLENEUVE, 2008; IPCC, 2007; POPE et al., 2002). High levels of air pollutants cause approximately five million deaths annually, mainly due to the health damage related to ozone (O₃) and particulate matter (MP) exposure (STANAWAY et al., 2018). In the case of fine particulate matter (PM_{2.5}) this pollutant has become one of the main subjects of international research on air pollution (PENG et al., 2016). Exposures to high PM_{2.5} levels significantly contribute to the global increase in certain diseases, causing approximately 3 million premature deaths in 2013 (FOROUZANFAR et al., 2015).

Studies focused on analyzing information about the concentration of gases and particles in the atmosphere are usually developed by using air quality monitoring stations, which are punctual, generating inevitable spatial gaps in-ground monitoring. Although this type of monitoring provides real-time data with high temporal resolution, the available monitoring resources restrict spatial coverage and resolution (LIN et al., 2015). In Brazil, for example, until 2014, air quality measurements were carried out in only 1.7% of municipalities (RÉQUIA;

KOUTRAKIS; ROIG, 2015). The country has only 0.03 stations per 1,000 km², while the US has 0.5, Japan 4.9, and Germany 5.18 (RÉQUIA; KOUTRAKIS; ROIG, 2015). In this context, remote sensing has been explored in recent years as a tool that can play an important role in mapping the dynamics of various atmospheric pollutants.

Remote sensing techniques used to measure air pollution and other chemically reactive trace gases in the lower atmosphere began to be developed in 1981 when the first carbon monoxide (CO) measurements were carried out (REICHLE et al., 1986). Since then, the ability to estimate by remote sensors has increased significantly. New sensors have made it possible to determine concentrations of sulfur dioxide (SO₂) (FIOLETOV et al., 2016; KROTKOV et al., 2016), nitrogen oxides (NO e NO₂ - NO_x) (BECHLE; MILLET; MARSHALL, 2013; BUCSELA et al., 2013; KROTKOV et al., 2016), O₃ (CUESTA et al., 2013; FISHMAN et al., 1990; FISHMAN; WOZNIAK; CREILSON, 2003), carbon compounds (CH₄ e CO₂) (BARKLEY; MONKS; ENGELEN, 2006; PARKER et al., 2011; SCHEPERS et al., 2012) e PM (PM₁₀ e PM_{2.5}) (VAN DONKELAAR; MARTIN; PARK, 2006; LIN et al., 2018; PÉREÉ et al., 2009). With the emergence of the COVID-19 pandemic, part of the air quality studies were based on satellite data applications. These studies essentially evaluated NO₂ through the Tropospheric Monitoring Instrument (TROPOMI) and Ozone Monitoring Instrument (OMI) sensors, and particulate matter based on its relationship with Aerosol Optical Depth (AOD) data extracted from the Moderate-Resolution Imaging Spectroradiometer sensors (MODIS) (e.g., BAUWENS et al., 2020; DIAMOND; WOOD, 2020; FAN et al., 2020a; FILONCHYK; HURYNOVICH; YAN, 2021; NAKADA; URBAN, 2020; ZAMBRANO-MONSERRATE; RUANO; SANCHEZ-ALCALDE, 2020).

The NO₂ data obtained by satellite represent the density of NO₂ molecules in the total column (troposphere + stratosphere) or in the troposphere. Thus, as NO₂ is a short-lived pollutant emitted mainly by fossil fuel combustion, its highest levels and density will be observed close to the surface. This enables comparisons between the data obtained for tropospheric NO₂ density and the NO₂ concentrations measured by air quality stations (BASSANI et al., 2021; CERSOSIMO; SERIO; MASIELLO, 2020; VÎRGHILEANU et al., 2020; ZHENG et al., 2019). On the other hand, AOD is a measure that represents the optical properties of aerosols and is not a quantitative measure of particulate matter fractions (coarse and fine particulate matter, PM₁₀ and PM_{2.5}, respectively). In addition, satellite-derived AOD data represents the

total column. Given the above, integrated techniques to estimate PM_{10} and $PM_{2.5}$ concentrations at ground level have recently been investigated (HOFF; CHRISTOPHER, 2009).

Thus, although the monitoring of air pollutants by remote sensing has great potential for assessing impacts on health and the environment, its capacity will depend on the quality of the measurement. In this way, the use of local information to evaluate the quality of remote sensing products is essential, even to propose more appropriate adjustments to the atmospheric conditions of the region of interest. Finally, Brazil, due to its immense territoriality and the strong impact of industrial-urban emissions and biomass burning, is undoubtedly a country that can benefit immensely from developing techniques based on remote sensing for monitoring air pollution conditions.

2 RESEARCH OBJECTIVES

2.1 Main Objective

This study aims to evaluate the spatiotemporal dynamics of air pollutant concentrations in Brazil during the COVID-19 pandemic. In addition, the research proposes the evaluation of remote sensing products aimed at mapping air pollutants in the lower atmosphere.

2.2 Specific Objectives

In more specific terms, the objectives of the study are:

- Assess the influence of state-imposed mobility restrictions during the COVID-19 outbreak on regional air quality and identify potential changes in pollutant levels;
- Statistically evaluate how pollutant concentrations behaved locally in response to mobility restrictions imposed during the COVID-19 pandemic in the state of São Paulo;
- Evaluate whether traditionally used remote sensing products for mapping particulate matter and nitrogen dioxide have the potential for air quality monitoring in the state of São Paulo;
- Identify available Aerosol Optical Depth (AOD) products, assess their accuracy, and investigate factors impacting their retrieval quality.

3 THEORETICAL BACKGROUND

3.1 Air Pollution

Atmospheric pollution can be defined as the condition in which the concentration of gases or particles (liquid and solid) is sufficiently high, above their environmental levels, to produce adverse effects on humans, animals, vegetation, or materials (SEINFELD; PANDIS, 2016). Traditionally, air pollution was seen as a phenomenon characteristic of industrialized regions and large urban centers (SEINFELD; PANDIS, 2016), since industrial activities, as well as vehicles, emit gases such as CO₂, nitrogen oxides (NO_x), particulate matter (PM), among others. However, it is currently known that these pollutants have a high dispersive character and can be transported across countries and to other geographic borders (LIN et al., 2015). Therefore, it is clear that large urban centers are just "hotspots" in a continuum of concentrations of trace gases and particles over the entire Earth's atmosphere (SEINFELD; PANDIS, 2016).

Atmospheric pollutants can be separated according to their source type into two groups: primary and secondary pollutants. Primary pollutants are those emitted directly by the source [e.g., carbon monoxide (CO), CO₂, methane (CH₄), sulfur dioxide (SO₂), NO_x, hydrocarbons, and some types of PM] and secondary are those formed in the atmosphere through chemical reactions [e.g., hydrogen peroxide (H₂O₂), sulfuric acid (H₂SO₄), nitric acid (HNO₃), sulfur trioxide (SO₃), nitrates (NO₃⁻), sulfates (SO₄²⁻), ozone (O₃), and some types of MP] (VALLERO, 2014). Primary pollutants can react in the atmosphere with other compounds, thus forming secondary pollutants, being therefore, also called precursors. Most air pollutants have natural and anthropogenic sources, which depend on the species considered. Anthropogenic sources can also be classified as stationary sources (or fixed), when they occupy limited areas (e.g., industries, fires, etc.), and mobile sources, which are geographically dispersed (e.g., vehicles in general). The main air quality indicator substances, as well as their sources and effects on human health and the environment are presented in Table 3.1.

Table 3.1 – Overview of common air pollutants: sources and impacts on human health and environment.

Pollutant	Main Sources	Impacts on human health	Impacts on the environment
Inhalable fine particles (PM _{2.5})	Emitted as a primary pollutant during combustion (industrial, vehicular, and biomass burning), or formed in the atmosphere as a secondary pollutant of sulfate and nitrate, among others.	Harmful to the respiratory and cardiovascular systems. It can cause premature mortality.	It changes radiative transfer in the atmosphere, potentially altering radiative forcing on a global scale.
Inhalable coarse particles (PM ₁₀) and Smoke	Emitted as a primary pollutant during combustion (industrial, vehicular, and biomass burning), resuspended dust, or formed in the atmosphere as a secondary pollutant.	Harmful to the respiratory system.	It changes radiative transfer in the atmosphere, potentially altering radiative forcing on a global scale.
Total suspended particles (TSP)	Industrial processes, motor vehicles (exhaust), resuspended dust, and biomass burning.	Harmful to the respiratory system.	It changes radiative transfer in the atmosphere, potentially altering radiative forcing on a global scale.
Sulfur dioxide (SO ₂)	Primary pollutant emitted during the combustion of sulfur-containing fuels (coal, diesel, fuel oil) and in industrial processing (oil refineries, pulp and paper production, fertilizers).	It aggravates asthma by causing shortness of breath, coughing and inflammation of the respiratory tract.	It causes acidification of soils and aquatic ecosystems. It can damage forest ecosystems. Contributes to the formation of particulate matter.
Nitrogen oxides (NO, NO ₂)	Primary and secondary pollutants resulting from fuel combustion in motor vehicles, industrial processes, thermal power plants and incinerations. Minor emissions occur naturally in soils.	It can cause respiratory diseases (cough, sore throat) and, in high concentration, inflames the airways and reduces the functioning of the lungs.	Contributes to the eutrophication of aquatic ecosystems. It can lead to the formation of acid rain, damage to vegetation and crops.
Carbon monoxide (CO)	Primary pollutant from limited (incomplete) oxygen combustion in motor vehicles, industrial processes and domestic heating.	Interferes with oxygen uptake, resulting in oxygen deprivation. Low to moderate doses cause headaches, impair brain function and reduce manual dexterity. At high concentrations, death occurs.	It affects animals in the same way as humans. When oxidized, it contributes to the formation of CO ₂ and O ₃ (greenhouse gases).
Ozone (O ₃)	Secondary pollutant formed photochemically by solar radiation on NO _x and Volatile Organic Compounds (VOCs).	It damages the respiratory tract and impairs lung function. Prolonged exposure can result in decreased lung capacity and premature mortality.	High concentrations can damage vegetation. O ₃ is a greenhouse gas that changes short-term radiative transfer in the troposphere.

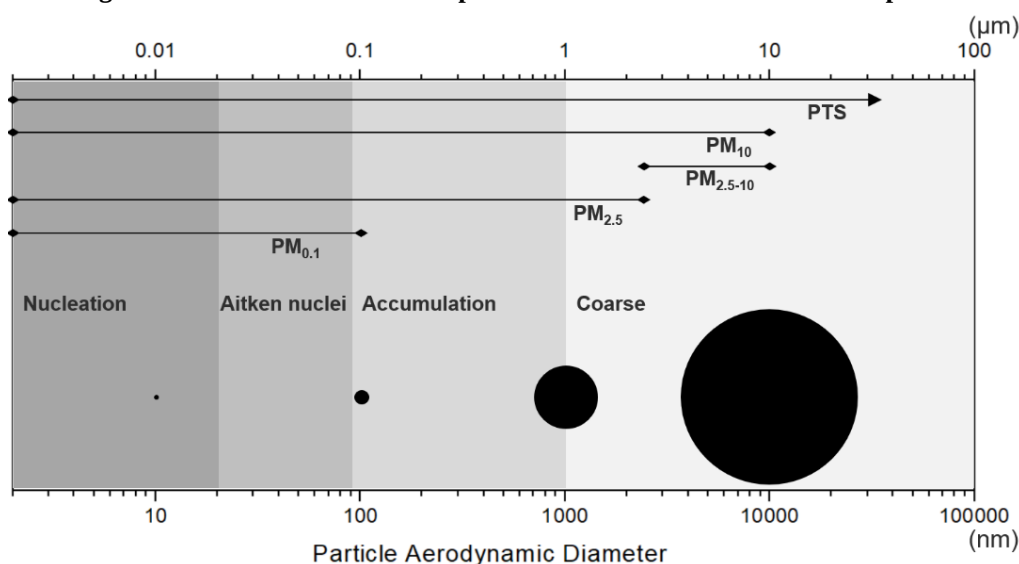
Source: CETESB, (2018) e Oke et al. (2017).

Among the main air pollutants, particulate matter is defined as a solid or liquid material or a mixture of phases and represents a wide variety of micrometer-sized particles that are suspended in the air (COLBECK, 2008). Over time and space, PM properties can be moderated by atmospheric physical and chemical processes such as condensation, evaporation, and coagulation (KULMALA et al., 2004). In addition, PM can be classified by its emission sources (Primary or secondary - Table 3.1) or according to their sizes (aerodynamic diameter)(COLBECK, 2008; KULMALA et al., 2004).

Concerning sizes, the PM can be called “nucleation mode”, when it has an aerodynamic diameter between 3 and 20 nm; “Aitken nuclei mode”, when presenting a diameter between 20 and 90 nm; “accumulation mode”, with a diameter between 90 and 1000 nm; and “coarse mode”, which have diameters greater than 1000 nm (KULMALA et al., 2004). Nucleation-mode particles are responsible for high concentrations in number, but due to their small size, they represent little of the total PM mass. These particles are subject to coagulation and/or condensation of vapors and then come to represent the accumulation mode. Given removal by precipitation or dry deposition, accumulation mode particles have a typical atmospheric lifetime

of about 1 to 2 weeks. Coarse mode particles are formed by mechanical wear processes and comprise soil-related and industrial dust and sea air (COLBECK, 2008; SEINFELD; PANDIS, 2016; WALLACE; HOBBS, 2005). Still, according to the particles size, PM can also be classified as (SEINFELD; PANDIS, 2016): (1) Total Suspended Particles (TSP), which have a diameter smaller than 100 μm ; (2) Inhalable particles (PM_{10}), which have diameters smaller than 10 μm ; (3) Coarse inhalable particles ($\text{PM}_{2.5-10}$), when diameters vary between 2.5 and 10 μm ; (4) Fine inhalable particles ($\text{PM}_{2.5}$), which have a diameter of less than 2.5 μm ; and (5) Ultrafine inhalable particles ($\text{PM}_{0.1}$), if they are smaller than 0.1 μm . Figure 3.1 presents the different dimensions of aerosols present in the atmosphere.

Figure 3.1 - Size classifications of particulate matter found in the atmosphere.



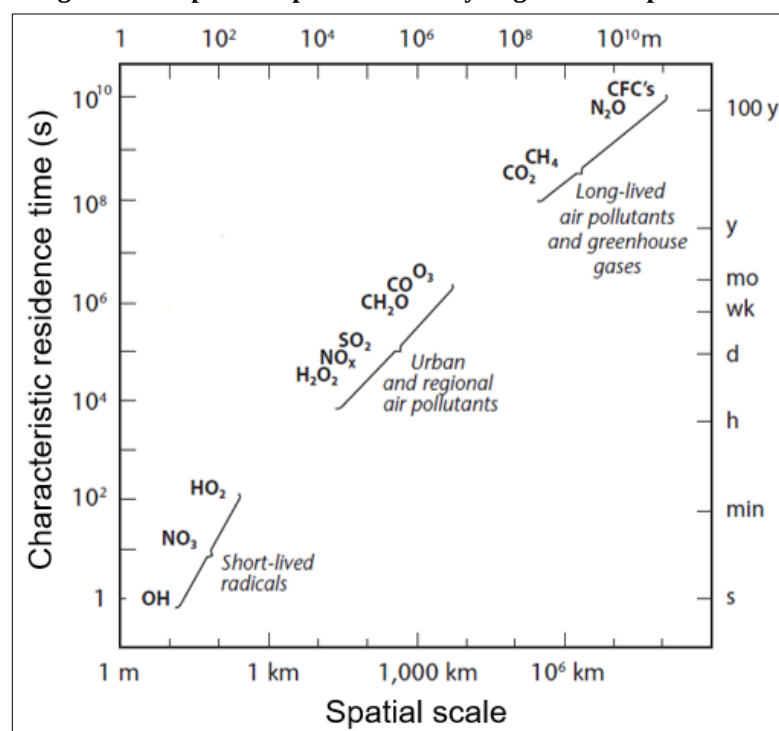
Source: Oke et al. (2017).

In addition to particulate pollutants, there are pollutants in gaseous form in the atmosphere, called “trace gases”. These gases comprise a huge range of species that have mole fractions less than 10^{-6} , 1 part per million by volume (ppmv) (SEINFELD; PANDIS, 2016). Even in small amounts, they play a crucial role in the global radioactive balance, in the thermodynamics and chemical properties of the atmosphere, and in the net radiation that reaches the earth's surface (SEINFELD; PANDIS, 2016; YAMASOE; CORRÊA, 2016). The primary gaseous pollutants in the atmosphere can be categorized as gases containing sulfur, nitrogen, and carbon compounds and tropospheric ozone (SEINFELD; PANDIS, 2016).

The impact of pollutants depends not only on the damage caused to health and the environment but also on their lifetime and geographical distribution. The pollutant lifetime is directly related to its spatial coverage (geographical distribution) and the dose to which the population is

exposed (damage to health). Figure 3.2 presents the typical atmospheric residence times for some gaseous pollutants and greenhouse gases. Greenhouse gases have long lifetimes (years), so they have a global geographic distribution and are transported over long distances. NO_x , on the other hand, reacts quickly when present in the atmosphere, having a residence time of approximately one day, which is characterized as a short-moderate lifetime pollutant. Thus, pollutants with this characteristic have a more local spatial coverage and can be transported over shorter distances when compared to other pollutants, such as aerosols, which have a residence time of weeks (depending on their composition) (SEINFELD; PANDIS, 2016).

Figure 3.2 – Spatiotemporal variability of gaseous air pollutants.



Source: Oke et al. (2017).

As seen, air pollutants, depending on the concentration, can cause damage to human health and the environment. Thus, rules are needed to regulate emission and air quality standards. In Brazil, the legislation that regulates air quality standards is Resolution 491 of 2018 by CONAMA (Conselho Nacional do Meio Ambiente) (BRASIL, 2018). This legislation updated the air quality standards regulated by CONAMA resolutions 03 of 1990 and 05 of 1989 (BRASIL, 1989, 1990), considering as reference the values recommended by the World Health Organization (WHO) in 2005. This resolution brings the maximum limits allowed in the atmosphere for pollutants: particulate matter (PTS, PM_{10} , and $\text{PM}_{2.5}$), SO_2 , NO_2 , O_3 , smoke, CO, and lead (Pb). The implementation of resolution 491 takes place in four phases that are

progressively more rigorous; three phases are considered intermediate, followed by the final phase, with the first phase coming into force on the date of publication of the legislation and the other phases still have no defined dates to be implemented. Other important resolutions are CONAMA 382 of 2006 and 436 of 2011, which deal with the maximum emission limits of air pollutants from stationary sources (BRASIL, 2006, 2011).

3.2 Remote Sensing

Over the last few decades, there has been a substantial increase in the use of remote sensing in applied studies (HANSEN; LOVELAND, 2012; YU et al., 2014). This fact is related to the continued technological development and the reduction of data acquisition costs that occurred since the launch of the first satellite (Sputnik 1), still in 1957 (CRACKNELL; VAROTSOS, 2007; GREKOUSIS; MOUNTRAKIS; KAVOURAS, 2015; TOTH; JÓZKÓW, 2016). The first satellite data had a restricted range of applications. Currently, several operational solutions can be extracted from remote sensing data, such as: biodiversity studies and ecological applications (FAUVEL et al., 2020; NAGENDRA et al., 2013), natural hazard assessment (HOQUE et al., 2017; SAMANTA; PAL; PALSAMANTA, 2018), hydrological and climatic dynamics (HOOKER; DUVEILLER; CESCATTI, 2018; SOUZA et al., 2020; WEST; QUINN; HORSWELL, 2019), land cover monitoring (GREKOUSIS; MOUNTRAKIS; KAVOURAS, 2015; PÉREZ-HOYOS et al., 2017; RUDKE et al., 2021b, 2019), and atmospheric pollutants monitoring (AVITABILE et al., 2016; BIRDSEY et al., 2013; LI et al., 2021).

The sensors attached to the satellites are responsible for acquiring information of interest. These can be divided into photographic sensors, radar, laser, spectrometers, and radiometers. The use of specific data depends on the type of application of interest, the size and dynamics of the object or phenomenon under study (ELSAYED; RISCHBECK; SCHMIDHALTER, 2015). Depending on how data is acquired, sensors can be divided into two groups: passive and active. The main difference is that passive sensors rely on solar radiation as a light source, and active sensors are equipped with components that emit light (ELSAYED; RISCHBECK; SCHMIDHALTER, 2015). These sensors are coupled to satellites that have different orbits, which are associated with the function to which the sensor was designated. The main mapping orbitals are geostationary orbit (GEO), Low Earth Orbit (LEO), Medium Earth Orbit (MEO), as polar orbit, and Sun-synchronous orbit (SSO).

Regardless of the sensor's characteristics, some attributes describe and determine whether the application of data obtained by the sensor is appropriate for a particular study. Temporal, spatial, spectral, and radiometric resolution are considered key concepts used in the characterization of remote sensing data.

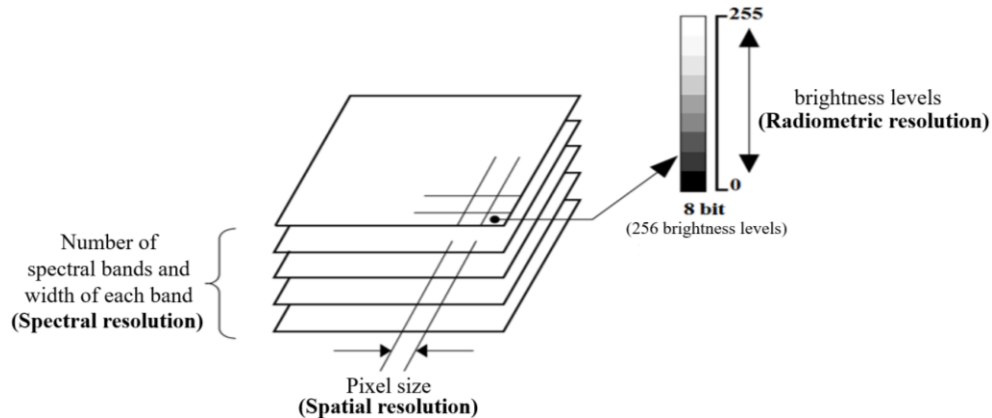
Temporal resolution is related to how often a given area is imaged (e.g., 1 day, 4 hours, etc.). Thus, better temporal resolutions are obtained, in general, by geostationary sensors and orbits close to the pole. High temporal resolutions are required to study phenomena of significant variability, such as atmospheric pollution. In contrast, low temporal resolutions are generally used in studies that require more detail on the surface (greater spatial resolution) and therefore demand more time for acquisition.

Spatial resolution indicates the size of the smallest distinguishable surface that can be recorded separately in an image (Figure 3.3) - this unit of area is called a pixel (CAMPBELL; WYNNE., 2011). Therefore, a sensor with a spatial resolution of 30 meters has pixels that distinguish objects with an equivalent of 900 m² (30 m x 30 m) or larger (FLORENZANO, 2002).

Spectral resolution measures the width of a sensor's spectral bands (GITAS et al., 2012). A high spectral resolution indicates that each detector receives energy in a narrower range of wavelengths (LILLESAND; KIEFER; CHIPMAN, 2014). Other features of this type of resolution are the number and position of spectral bands (spectral region) (Figure 3.3). The orbital sensors used for environmental applications work mainly in the Ultraviolet (UV), Visible (VIS), and Infrared (IR) spectral regions. Each of these regions can be separated into bands of lower spectral range, just as VIS can be divided according to the regions referring to the spectrum of green, red, and blue colors. The IR region can be separated into Near Infrared (NIR), Shortwave Infrared (SWIR), and Thermal Infrared (TIR).

The radiometric resolution expresses the ability of the sensor to capture different intensities or digital levels of the electromagnetic field of objects (GITAS et al., 2012). Radiometric resolution is regularly expressed in number of binary digits (bits). Therefore, a sensor with an 8-bit radiometric resolution has 256 brightness levels (RICHARDS; JIA, 2006) (Figure 3.3).

Figure 3.3 - Overview of technical specifications of remote sensors for data collection



3.2.1 Satellite-based monitoring of atmospheric pollution

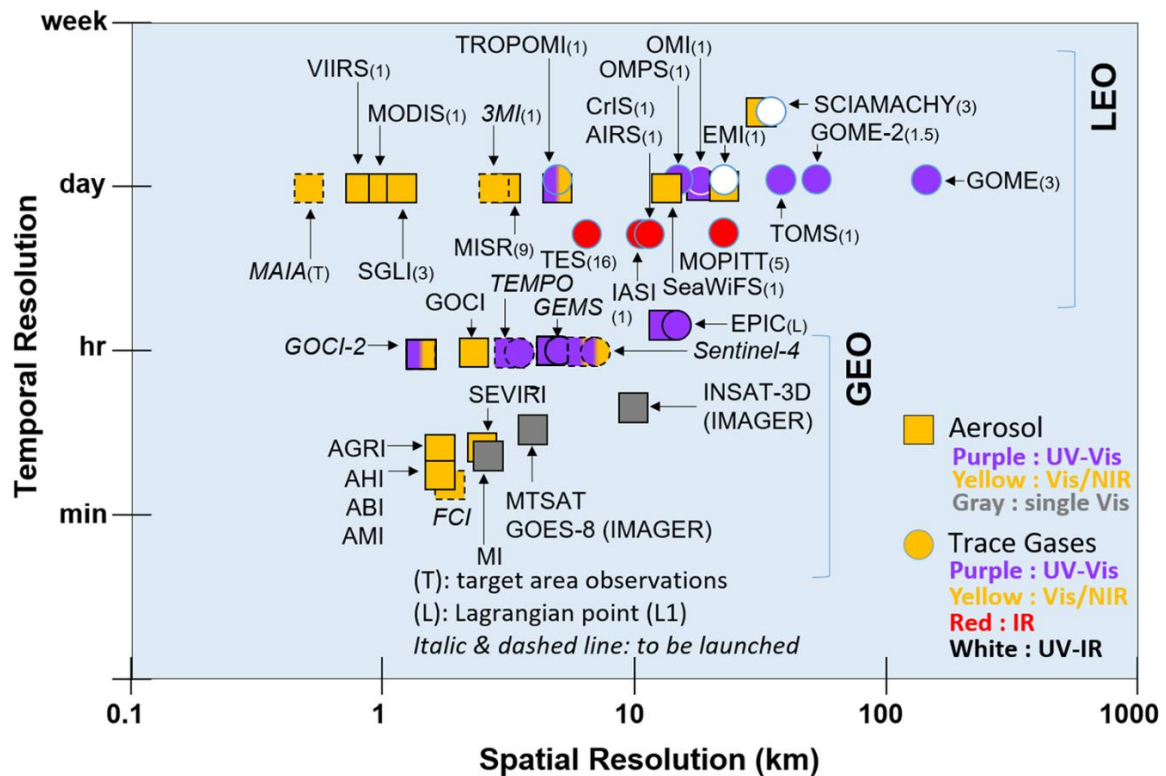
With the increase in the concentrations of several pollutants across the globe and the beginning of epidemiological studies that indicated the association between air pollution and the rise in morbidity and mortality rates due to respiratory diseases (BELL; DAVIS, 2001; FREITAS et al., 2004; GOUVEIA, 2000; NEMERY; HOET; NEMMAR, 2001), interest in regional air quality has grown. Thus, methods for monitoring air quality based on systematic observations became in demand, especially in more industrialized areas. The main objects of interest are in remotely measuring concentrations of ozone, aerosols, and their precursors (NO_2 , SO_2 , formaldehyde (HCHO), etc.). In this context, air quality monitoring networks began to be implemented, and remote sensors capable of assisting in monitoring began to be produced.

Therefore, with the premise of monitoring large areas, the first satellites capable of measuring atmospheric pollutants were launched in the 1970s. This is the case of the sensors Solar Backscatter Ultraviolet radiometer (SBUV) e Total Ozone Mapping Spectrometer (TOMS), which were launched in 1978, coupled to the NIMBUS-7 satellite, and which could perform O_3 measurements (total column). The first operational aerosol products were developed with data from the Advanced Very High Resolution Radiometer (AVHRR) sensor, launched in 1978 on the TIROS N platform (LEE et al., 2009). Furthermore, although not initially designed for this purpose, TOMS has provided aerosol measurements (TORRES et al., 2002), together with the Stratospheric and Mesospheric Sounder (SAMS), also coupled to NIMBUS-7 satellite. Since then, the technology used in the sensors has evolved significantly, allowing more detailed acquisition of these pollutants, and providing the retrieval of other trace gases, such as NO_2 , SO_2 , HCHO, and CO (KIM et al., 2020). In this sense, remote sensing has been playing an essential role in understanding several spatial aspects related to the current status of air quality,

as well as future trends, providing information on quantities of pollutants, emissions, and long-range transport (regional/global scales), which are challenging to observe only with point stations (KIM et al., 2020).

In addition to the evolution related to the range of mapped pollutants, there was also an improvement in the spatial resolution of the sensors, which can currently be mapped at medium resolutions [2-30 m; e.g., HYperspectral Camera (HYC), launched in 2019], and in the temporal resolution, with the implementation of geostationary satellites. Figure 3.4 presents the relationship between the spatial and temporal resolution of some sensors.

Figure 3.4 – Temporal and spatial resolution of sensors capable of monitoring gases and particles present in the atmosphere. For LEO instruments, the numbers in parentheses represent the revisit time in days. For planned missions, mission names are in *italics*, and symbol outlines are in dashed lines.



Source: KIM et al. (2020).

Currently, on a global scale (LEO satellites), the MODerate resolution Imaging Spectroradiometer (MODIS) sensors, coupled to the Aqua and Terra satellites, represent the primary source of aerosol data, especially Aerosol Optical Depth (AOD), for continental areas. The sensors have 36 spectral bands, of which seven are widely used for AOD data retrieval (wavelengths range from 0.620 to 2.155 μm). The sensor has spatial resolutions that vary

according to the spectral band analyzed, ranging from 250, 500 and 1,000 meters, and it maps the entire Earth's surface every 1 or 2 days (more often near the poles).

Regarding trace gases, Ozone Monitoring Instrument (OMI) and TROPospheric Monitoring Instrument (TROPOMI) are currently the most used sensors for global mappings (GKATZELIS et al., 2021). On board the Aura satellite, the OMI sensor was launched in 2003, with a spatial resolution of 13x24 km and a temporal resolution of 1 day. The products derived from OMI allow the mapping of NO₂ (total and tropospheric columns), SO₂ (total column), Aerosol index, HCHO (total column), and O₃ (total and tropospheric columns). The TROPOMI sensor, coupled to the Sentinel 5P satellite, was launched in 2017 and can capture information on the elements mapped by OMI, in addition to the height of the aerosol layer, CO (total column), and CH₄ (total column). The temporal resolution of TROPOMI is one day, and the spatial resolution at the nadir (perpendicular to the Earth's surface) was approximately 7 km until August 6, 2019, passing to 5.5 km due to the reduction in radiance co-addition time (LUDEWIG et al., 2020). In the orbit direction, the TROPOMI sensor varied resolutions, being around 3.5 km for bands 2-6, around 7 km for bands 7-8, and approximately 28 km for band 1, generating gas-trace products in different resolutions (LUDEWIG et al., 2020).

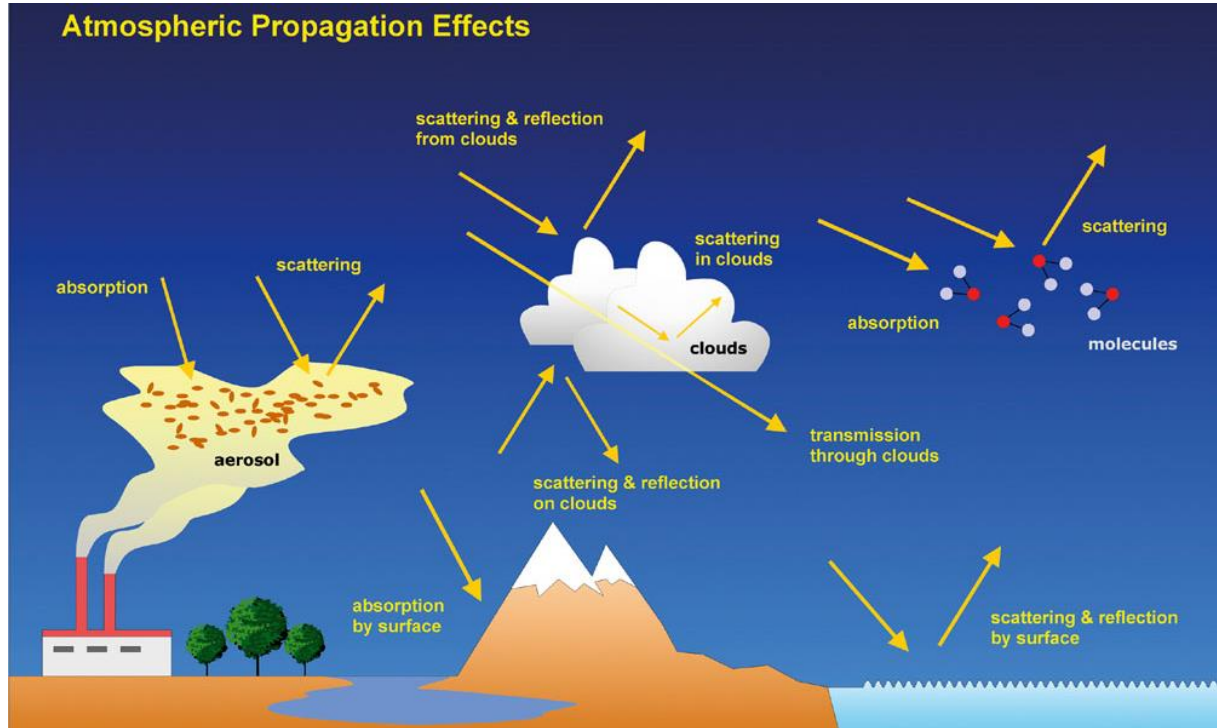
With the COVID-19 pandemic, MODIS, OMI, and TROPOMI sensors have become widely used to assess regional air quality changes. According to Gkatzelis et al. (2021), until October 2020, one third of the studies published on this topic used satellite data, with OMI and TROPOMI sensors being widely used to verify NO₂ reductions and MODIS used to evaluate particulate matter.

3.2.2 Atmospheric composition and remote sensing

Compared to other types of monitoring, satellite observations have the advantage of providing information on a regional-global scale, thus assessing air quality conditions in remote areas. For this, data retrieval from trace gases and aerosols is based on inferences regarding the amount of electromagnetic radiation acquired by sensors at the Top Of the Atmosphere (TOA). This radiation travels through the atmospheric column until the sensor captures it, so the spectral radiance measured on the satellite already considers all interactions with the constituents of the atmosphere (trace gases, aerosols, clouds) (BOVENSMANN et al., 2011), as shown in Figure 3.5. In this context, the main challenge of remote sensing is the quantitative derivation of these constituents from the spectral radiance measured in the TOA by the sensor. Given the

atmospheric constituents, this derivation depends on knowledge of the absorption, emission, and scattering of solar electromagnetic radiation in the atmosphere.

Figure 3.5 - Solar radiation interactions with Earth's atmosphere and surface.



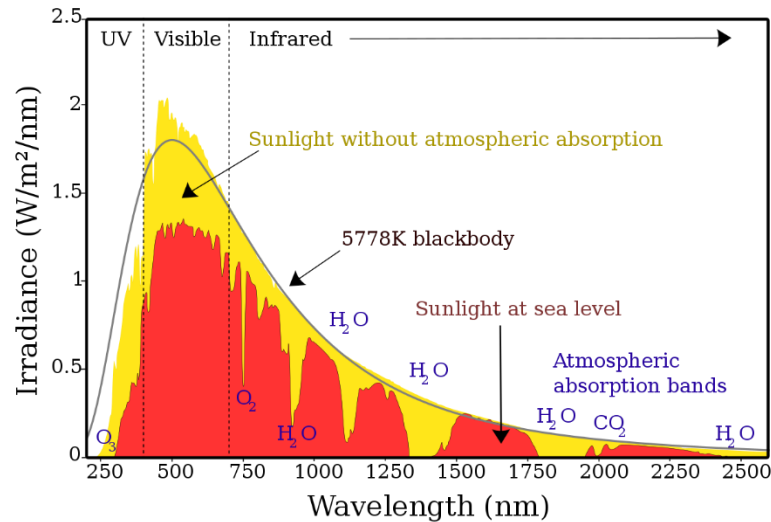
Source: Bovensmann et al. (2011).

In remote sensing air quality monitoring, most sensors that collect relevant data are passive (DUNCAN et al., 2014). These sensors map trace gases and aerosols considering radiance in the electromagnetic spectrum regions referring to the ultraviolet (UV), visible (VIS), and infrared (IR) wavelengths (DUNCAN et al., 2014).

As the Sun is the source of electromagnetic radiation for passive sensors, it is crucial to understand how it interacts with the matter in the atmosphere. The Sun radiates approximately like a blackbody, which means it absorbs almost all the incident radiation upon it (YAMASOE; CORRÊA, 2016). Therefore, considering its surface temperature ($\sim 5780\text{K}$) and Planck's law for a blackbody, it is possible to indicate that solar radiation peaks in the visible spectral range (Figure 3.6). Solar radiation essentially corresponds to the spectral regions of VIS (46%) and NIR (46%), and a smaller portion (8%) is characterized by wavelengths shorter than VIS (e.g., UV radiation) (YAMASOE; CORRÊA, 2016). In these regions, radiative transfer through the atmosphere is governed by (see Figure 3.5): (1) Scattering by air molecules (Rayleigh and Raman scattering); (2) Scattering and absorption by aerosols and cloud particles (Mie

Scattering); (3) Absorption and emission by trace gases; (4) Refraction due to the density gradient in the atmosphere; (5) Surface reflection and absorption (BOVENSMANN et al., 2011).

Figure 3.6 – Spectral distribution curves of solar irradiance observed at the top of the atmosphere (yellow) and the surface (red). The black line represents the theoretical curve for a blackbody at 5778K.



Source: Wikimedia.

In the atmosphere, scattering occurs due to molecules, aerosol particles, and clouds containing water droplets and ice crystals (Figure 3.5). The most characteristic scattering phenomenon in the atmosphere results in the blue color of the sky. This is due to the molecular scattering of radiation by N_2 and oxygen in all directions (scattering), which is especially strong for shorter wavelengths. This phenomenon occurs to the detriment of Rayleigh scattering, which occurs when the diameters of gas molecules are much smaller than the wavelength of the incident radiation, and the intensity of this scattering is inversely proportional to the fourth power of the wavelength (λ^{-4}) (YAMASOE; CORRÊA, 2016). Thus, as the blue color is represented by the shortest wavelength (within the field of visible radiation), its intensity prevails, and the sky tends to present this color. For aerosols and cloud particles that have diameters comparable to or even larger than the wavelength of the incident radiation, the resulting scattering is called Mie scattering. In this type of scattering, the resulting radiation occurs due to a large number of coherently excited elementary emitters (molecules, for example) in a particle (BURROWS; PLATT; BORRELL, 2011). When compared to Rayleigh scattering, the main difference presented in Mie scattering is, generally, the lower wavelength dependence and strong dominance of the forward direction in the scattered light; as the particles become larger, the

scattered radiation is concentrated more in the forward directions(BURROWS; PLATT; BORRELL, 2011; YAMASOE; CORRÊA, 2016).

The processes of absorption and emission by gases also occur in the atmosphere. Absorption is the process by which incident electromagnetic radiation is absorbed by the molecule, which undergoes an internal change generated by energy (raising the energy state of the molecule) (BURROWS; PLATT; BORRELL, 2011). In this process, there is a physical alteration of the medium crossed, which in the case of the atmosphere is mainly related to the increase in temperature or internal energy (YAMASOE; CORRÊA, 2016). In the atmosphere, CO₂, O₂, O₃, and H₂O molecules absorb radiation at different wavelengths, resulting in different curves of radiation measured at the top of the atmosphere and at sea level, as shown in Figure 3.6. As the gases present in the atmosphere are made up of groups of atoms with chemical bonds, the processes of interaction with radiation are governed by the principles of quantum mechanics. This means that it is possible to quantify the internal energy of molecules and that interactions with radiation can change energy states in a discrete form (BURROWS; PLATT; BORRELL, 2011). Thus, when radiation hits an atom, its electrons can move to a higher energy state, and the atom becomes in an excited state (YAMASOE; CORRÊA, 2016). As the atom seeks the stability state, the electron that moves to a higher energy level tends to return to the lower energy level quickly - in this process, energy emission occurs (YAMASOE; CORRÊA, 2016). As molecules have various forms of internal energy, the molecular absorption and emission spectrum is more complex. It involves three distinct types of energy state transitions: electronic, vibrational, and rotational (see Section 3.3).

The radiation refraction in the atmosphere occurs due to the different densities through which the radiation travels. Thus, the incident radiation deviates from its typical trajectory when it crosses the different atmospheric layers. Snell's Law can be used to measure how much deviation will occur in the radiation (MALM, 2016). This law considers the incidence angle of radiation and the refraction indices of the two atmospheric levels at which the intention is to quantify the angle of refraction.

In addition to being scattered and absorbed in the atmosphere, incident electromagnetic radiation may undergo reflection and absorption upon reaching the surface. Thus, the radiation flux on a given surface will depend on the wavelength of the incident electromagnetic radiation and the surface coverage. In this way, the albedo, the ratio between the reflected diffuse and

incident radiation, generally depends on the wavelength (BURROWS; PLATT; BORRELL, 2011). Albedo ranges from zero (all photons are absorbed) to one (all photons are reflected), and most surfaces on Earth (except snow or ice) have albedo in the UV/VIS spectral range between 2 and 30% (BURROWS; PLATT; BORRELL, 2011).

The phenomena mentioned above depend on the wavelength of the incident radiation; that is, they occur differently for each line of the radiation spectrum. For this reason, the study of a particular component of the atmosphere and surface depends on the knowledge of the typical behavior that occurs when the energy interacts in given spectral lines (or spectral interval) and the phenomena mentioned (YAMASOE; CORRÊA, 2016). In remote sensing, the characteristic of these phenomena can be detected by comparing the radiation that returns to the sensor after iterations with the atmosphere and surface and the unaffected extraterrestrial solar irradiance (at the TOA) (BOVENSMANN et al., 2011). Therefore, the quantitative analysis of the constituents requires (BOVENSMANN et al., 2011): (1) high quality solar and terrestrial radiation spectra (spectral and radiometric); (2) accurate modeling of the sunlight transfer through the atmosphere to the sensor (radiative transfer model or direct model); and (3) techniques to relate the measured TOA spectra with the constituent properties.

3.3 Remote detection of trace gases by satellite

As previously mentioned, depending on the energy/frequency of the radiation incident on gas molecules, they undergo excitation and react through three energy states: molecular rotation, atomic vibration within the molecule, or electronic transitions between energy states. Based on these processes, which generate discrete absorption spectral lines, it is possible to determine the chemical species present in the atmosphere through remote sensing (BURROWS; PLATT; BORRELL, 2011).

Among the three energy states of the molecule, rotation occurs when the molecule absorbs energy of the same magnitude as its internal energy, typically in the far-infrared and microwave regions (wavelengths of approximately 10^3 to 10^5 μm). As a result, the molecule gains internal energy to rotate around an axis passing through its center of gravity (YAMASOE; CORRÊA, 2016). On the other hand, by absorbing radiation in the infrared spectrum (wavelengths of approximately 1 to 20 μm), the molecule gives rise to vibrational energy states. This occurs due to the oscillation/vibration of the bonds between atoms, causing them to vibrate around their equilibrium positions when disturbed (YAMASOE; CORRÊA, 2016). The transition to the

electronic energy state occurs due to the transfer of electrons to more external orbitals of the atom (as discussed in Section 3.3.2). The radiation required for this type of transition covers the ultraviolet and visible spectral regions (wavelengths shorter than 1 μm), when considering typical gas molecules in the Earth's atmosphere (YAMASOE; CORRÊA, 2016).

It is the transitions between these energy states that are associated with the interaction of molecules with electromagnetic radiation and provide the basis for the characterization of molecules through spectroscopic methods (BURROWS; PLATT; BORRELL, 2011). As mentioned earlier, the quantitative analysis of trace gases requires high-quality solar and terrestrial radiation spectra. This means that it is necessary to measure spectral regions of interest with a high spectral resolution to detect the spectral absorption lines that are formed when the gas absorbs specific wavelengths (BOVENSMANN et al., 2011; CHIMOT, 2018). With this purpose, passive sensors designed with nadir viewing are typically spectrometers. Spectrometers are devices that measure the intensity of energy as a function of wavelength (radiance spectrum) to create a spectrum of detected wavelengths (DUNCAN et al., 2014). Upon reaching the instrument's detector, individual photons are converted into electrons - a way of measuring the amount of energy entering at various wavelengths (DUNCAN et al., 2014). As mentioned, passive sensors detect gases and aerosols especially in the IR, visible and UV wavelength regions; this is because gases and aerosols absorb IR wavelengths (e.g., water vapour) or scatter visible and UV wavelengths (e.g., dust, NO_2) (DUNCAN et al., 2014).

Given the aforementioned, one of the most complex steps is related to the path of the emergent radiation, or more precisely, the average length of all possible paths due to the radiative processes that occur in the atmosphere and on the surface (scattering and absorption) (CHIMOT, 2018). These paths can be simulated through radioactive transfer equations implemented in models (Radiative Transfer Models – RTM). These models provide synthetic radiances, as would be measured by the sensor, for a specific state of the atmosphere and thus help to investigate how measurements of the satellite spectrum are affected by the atmosphere (BURROWS; PLATT; BORRELL, 2011; CHIMOT, 2018). The RTMs are an essential part of any trace gas retrieval process. Depending on the imposed application, more complex models can be applied, such as the full physical RTM model, or relatively simplified models like Beer's Law (also known as Beer-Lambert Law), as considered in Differential Optical Absorption Spectroscopy (DOAS; see Section 3.3.1).

To convert the data collected by the sensor (radiance spectra) into quantitative information about a specific gas (e.g., density of a particular molecule in the vertical column), it is necessary to perform a conversion of the spectral information using methods called "retrieval". Through the retrieval, an atmospheric state x is obtained (state vector that contains the numerical densities of the atmospheric constituents defined in discrete levels of altitude) from a measurement y (for satellites that observe the atmosphere with an angle of view in nadir the measurement y is represented by the intensity measured in the selected spectral window, which will depend on the atmospheric constituent of interest). Thus, the retrieval of gas information can be obtained through the relationship between x and y , which requires an RTM (F) to describe the dependency between the variables, where (BRASSEUR; JACOB, 2017):

$$y = F(x) + \varepsilon_0 \quad (3.1)$$

where: ε_0 represents observation errors in the measured signal relative to the RTM.

More effectively, x does not describe the physical representation of what occurs in the atmosphere but rather represents a set x' of physics approximations that explain most of the measurement y . Thus, it is necessary to consider a subset of all quantities that influence the measurement of the RTM – here represented by the parameter vector b ; some examples of b are satellite observation geometry (solar zenith angle, view zenith angle), cloud properties, surface properties, the presence of other tracers and aerosols, pressure, temperature, spectral line strengths, and instrument aspects (ESKES; BOERSMA, 2003). In this way, the x of Equation 3.1 is now represented as $x=(x', b)$, and the equation is now defined as (ESKES; BOERSMA, 2003):

$$y = F(x', b') + \frac{dF}{db}(b - b') + \varepsilon_0 \quad (3.2)$$

where: b' is the best estimate of these parameters in the RTM; and $\frac{dF}{db}(b - b')$ describes the errors resulting from uncertainties in the parameters b of the model.

As the radiation measured by the sensor contains information about the entire atmospheric profile and the Earth's surface, it is very complex to resolve all the subtle structures of the atmosphere accurately. In this case, the amount of independent information is generally low, so an infinite number of atmospheric states (or state vectors) can lead to the same single measurement (CHIMOT, 2018). This lack of detail prevents data retrieval along a gradient but

allows sufficiently distinct sets to be separated. This is the case with atmospheric NO₂, which cannot be segregated along an atmospheric vertical profile when derived from spaceborne sensors like OMI or TROPOMI. However, its total density (stratosphere + troposphere) or tropospheric column can be estimated (CHIMOT, 2018).

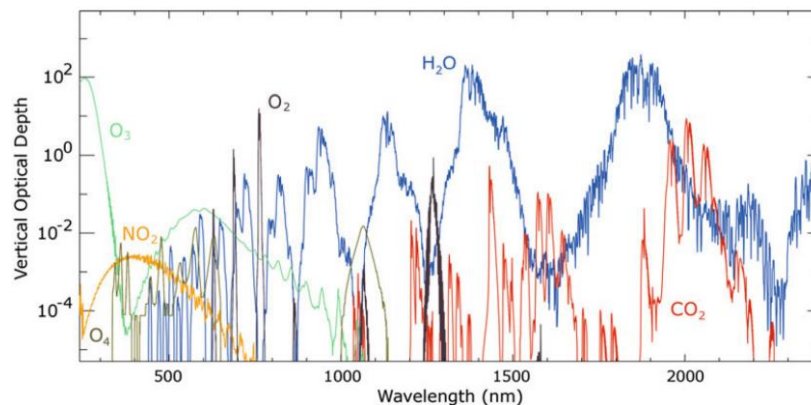
3.3.1 Tropospheric NO₂ retrieval: the DOAS method

Several trace gases in the atmosphere have structured absorption spectra in the UV-VIS and near-infrared spectral regions (Figure 3.7). For molecules with this characteristic, the Differential Optical Absorption Spectroscopy (DOAS) method (PLATT; PERNER, 1983) can be employed to retrieve their atmospheric abundances. In passive sensors, the DOAS method is based on the application of Beer's law (BRASSEUR; JACOB, 2017). When measuring trace gases, Beer's Law states that the radiant intensity passing through a homogeneous medium decreases as a function of the distance traveled (s), gas concentration, and spectral absorption intensity (CHIMOT, 2018):

$$I(\lambda, \theta, \phi) = I_0(\lambda, \theta, \phi) \cdot \exp \left(\int_s -\sigma(\lambda) \cdot \rho(s) \cdot ds \right) \quad (3.3)$$

where: $I_0(\lambda, \theta, \phi)$ represents the initial intensity, $I(\lambda, \theta, \phi)$, the measured radiation intensity (or radiance); $\rho(s)$, the density of the trace gas; and $\sigma(\lambda)$, the absorption cross section of a given gas measured in a laboratory. The product of $\sigma(\lambda) \cdot \rho(s) \cdot ds$ gives the optical thickness.

Figure 3.7 – Highly radiation-absorbing atmospheric constituents at ultraviolet-visible and near-infrared wavelengths.



Source: Bovensmann et al. (2011).

Equation 3.3 can be rewritten to consider the simultaneous presence of multiple absorbing constituents along with Rayleigh and Mie scattering, and it is then expressed as follows (BURROWS; PLATT; BORRELL, 2011):

$$I(\lambda, \theta, \phi) = I_0(\lambda, \theta, \phi) \cdot \exp \left\{ - \sum_i \sigma_i(\lambda) SCD_i - \sigma_{Ray}(\lambda) SCD_{Ray} - \sigma_{Mie}(\lambda) SCD_{Mie} \right\} \quad (3.4)$$

Where, SCD represents the Slant Column Density, which is the term $\int_s \rho(s) \cdot ds$ in Equation 3.3; and i is the index of the different absorbing constituents.

The various DOAS techniques rely on the same key concept, which consists of separating the wavelength-dependent absorption signal into two components: the low-frequency and the high-frequency parts (BURROWS; PLATT; BORRELL, 2011). In this case, the high-frequency component of the spectrum is used for the retrieval of atmospheric constituent absorptions, while the low-frequency part is treated as a closure term and is approximated by a low-order polynomial or another smoothing function (BURROWS; PLATT; BORRELL, 2011). Therefore, the absorption cross sections of the absorbing constituents are separated into the slow-varying part $\sigma^*(\lambda)$ and the differential part $\sigma'(\lambda)$: $\sigma(\lambda) = \sigma^*(\lambda) + \sigma'(\lambda)$, so that Equation 3.4 becomes:

$$I(\lambda, \theta, \phi) = I_0(\lambda, \theta, \phi) \cdot \exp \left\{ - \sum_i \sigma'_i(\lambda) SCD_i - \sum_i \sigma^*_i(\lambda) SCD_i - \sigma_{Ray}(\lambda) SCD_{Ray} - \sigma_{Mie}(\lambda) SCD_{Mie} \right\} \quad (3.5)$$

Rayleigh and Mie scattering cross sections, surface reflection, and low frequency can be modeled by a polynomial. In addition, the spectral fitting is performed within a predefined spectral window [e.g., 405-465 nm for TROPOMI NO₂ (VAN GEFFEN et al., 2019)] on the spectral reflectance R . DOAS spectral fit is obtained using a least squares fit approach applied to the logarithm of R at 405–465 nm (CHIMOT, 2018; VAN GEFFEN et al., 2019), which is rewritten from Equation 3.5:

$$\ln(R(\lambda)) = \sum_i \sigma'_i(\lambda) SCD_i + \text{polynomial} \quad (3.6)$$

Through this equation it is then possible to recover the total SCD of NO₂. To separate stratospheric (SCD^{strat}) and tropospheric (SCD^{trop}) contributions, NO₂ SCDs are assimilated based on the TM5-MP chemical transport model (VERHOELST et al., 2021). Finally, the Vertical Column Density (VCD) of NO₂ can be obtained through the equations:

$$SCD^{strat} = VCD^{strat} * M^{strat} \quad (3.7)$$

$$SCD^{trop} = VCD^{trop} / M^{trop} \quad (3.8)$$

where, M^{strat} e M^{trop} are the stratospheric and tropospheric Air-Mass Factor (AMF), respectively. These factors are derived from a look-up table of altitude-dependent AMF variables and actual daily information on the vertical distribution of NO₂ from the TM5-MP model; altitude-dependent AMF variables are satellite geometry, terrain height, cloud fraction and height, and surface albedo (VAN GEFFEN et al., 2019).

3.4 Remote detection of aerosols by satellite

As previously mentioned, in a cloud-free atmosphere, the radiance at TOA is caused by molecular scattering and absorption, surface reflectance, and scattering and absorption by aerosol particles (CHOWDHARY et al., 2019). Based on this principle, passive sensors analyze TOA radiance to extract aerosol optical properties (CHANG, 2012). In remote sensing, assumptions are made about surface reflectivity, aerosol size distribution, and chemical composition of aerosol particles (LUCCA, 2009).

3.4.1 Optical properties of aerosols

The light passing through the atmosphere is attenuated by the scattering and absorption of molecules and particles present in the atmosphere. To describe the variation of spectral radiance along an optical path due to attenuation or extinction, Beer's law is used. The extinction coefficient (σ_e) is defined as the change in radiance per unit thickness of the layer. It can be considered as the sum of the scattering coefficient (σ_s) and the absorption coefficient (σ_a) by particles and gases, both dependent on the wavelength (λ) (MALM, 2016):

$$\sigma_e(\lambda) = \sigma_s(\lambda) + \sigma_a(\lambda) \quad (3.9)$$

the relative contribution to aerosol particle extinction is commonly expressed in terms of simple albedo $\omega_0(\lambda)$, defined as (CHAUVIGNÉ et al., 2016):

$$\omega_0(\lambda) = \frac{\sigma_{sp}(\lambda)}{\sigma_{ep}(\lambda)} \quad (3.10)$$

So, when $\omega_0(\lambda) = 1$, all extinction is caused by scattering, and when $\omega_0(\lambda) = 0$, all extinction is caused by absorption. Most types of atmospheric aerosol have $\omega_0(\lambda) > 0.7$, that is, the radiation scattering effect is more important than the absorption one; becoming simpler the

detection of aerosols in regions on dark surfaces due to reducing the contribution of reflection by the cover (YAMASOE, 2004).

The aerosol optical depth (AOD) of the atmospheric column is defined as the aerosol extinction coefficient (σ_{ep}) along the vertical column (surface to the TOA):

$$AOD(\lambda) = \int_0^{TOA} \sigma_{ep}(\lambda, h) dh \quad (3.11)$$

The angular distribution of scattered photons is characterized by the phase function $p(\theta)$, which represents the scattering probability in each direction with respect to the incident (LUCCA, 2009). Several parameters can be derived from the phase function, such as the asymmetry parameter (g), which describes how much radiation is forward-scattered relative to the total scattering:

$$g(\lambda) = \frac{1}{2} \int_0^{TOA} \cos(\theta) p(\lambda, \theta) \sin(\theta) d\theta \quad (3.12)$$

If the scattering of light is isotropic (symmetric), g is null ($g=0$) and the scattering angle is approximately 90° . If all the radiation is forward-scattered ($\theta = 0^\circ$) $g = 1$ and $g = -1$ if the radiation is completely backscattered ($\theta = 180^\circ$) (LUCCA, 2009).

The scattering of solar radiation varies with the size parameter (x) of particles and gases, which is defined as the ratio of the particle size to the wavelength of the incident radiation. For spherical particles, x is defined as follows (BOHREN, 2006):

$$gx = 2\pi r/\lambda \quad (3.13)$$

The variation of the type of scattering experienced by solar radiation as a function of the size parameter is presented in Table 3.2. To describe the optical properties of the particles it is necessary to identify the proper scattering type.

Table 3.2 - Relationship between the type of solar radiation scattering and the particle size parameter.

x	Scattering type
$\ll 1$	Rayleigh - scattering by small molecules or particles in the atmosphere relative to the incident λ .
≈ 1	Mie - scattering by homogeneous spherical particles of size close to the incident λ .
$\gg 1$	Geometric - large atmospheric particle scattering in relation to incident λ .

Source: Lucca (2009).

Knowing the size distribution, it is also possible to define a parameter that describes the distribution of particles independently of models and mathematical adjustments. The parameter that best describes the radiative properties of a given size distribution is the effective radius r_e , defined as (REMER et al., 2013b):

$$r_e = \frac{\int_0^\infty r^3 n(r) dr}{\int_0^\infty r^2 n(r) dr} \quad (3.14)$$

3.4.2 Principles of aerosol detection by using satellite

To quantify particles in the atmosphere, sensors coupled to satellites measure the TOA radiance, which is affected by aerosol optical properties. In the following, reflectance will be used instead of radiance, being defined as $\pi L \cos(\theta_0)/F_0$, where L is the radiance, F_0 is the extraterrestrial solar irradiance, and θ_0 is the solar zenith angle (LUCCA, 2009).

When mapping dark surfaces, it is assumed that the surface is homogeneous, making these the simplest cases for retrieving aerosol optical properties (e.g., oceans) (REMER et al., 2013b). The simplicity of the equation, in this case, is because the TOA reflectance is solely caused by scattering from aerosols and molecules in the atmosphere (Rayleigh scattering) (REMER et al., 2013b). Aerosol and Rayleigh scattering will depend on Sun/Satellite geometry; described by the solar zenith angle θ_0 , satellite's zenith angle θ , and relative azimuth angle between the Sun and the satellite ($\phi - \phi_0$) (LUCCA, 2009). The contribution of angular aerosol effects to TOA reflectance depends on aerosols' optical properties and vertical distribution.

When the land cover is not dark, the scattered and reflected radiations from the surface in the atmosphere contribute to TOA reflectance, making the quantification of AOD more complex. The reflectance of a parallel, flat, cloud-free, and horizontally homogeneous atmosphere overlaid on an isotropically reflecting surface (Lambertian) can be expressed as follows:

$$\rho(\lambda) = \rho_{\text{atm}}(\lambda) + \frac{\rho_{\text{sfc}}(\lambda)}{1 - \rho_{\text{sfc}}(\lambda) \cdot r(\lambda)} T(\lambda) \quad (3.15)$$

where: ρ_{atm} is the path reflectance due to scattering in the atmosphere; ρ_{sfc} is the surface albedo; r is the spherical albedo of the atmosphere; and T is the sum of direct and diffuse transmittance through the atmosphere. All variables in Equation 3.15 depend on the Sun/satellite geometry.

In this case, the effect of aerosols on the TOA reflectance is twofold: (1) increasing the path reflectance (ρ_{atm}) and (2) reducing the transmittance (T); as a result, the contribution of surface reflection decreases. When the surface albedo is small, the first effect dominates, but for higher surface albedos (e.g., urban areas), the reduction in transmittance becomes increasingly important.

In wavelengths corresponding to the VIS and NIR spectra, the Earth's surface has a much higher albedo than the ocean (REMER et al., 2013a). In this case, high albedo values on the Earth's surface dominate the TOA reflectance in the VIS and NIR spectra. Thus, distinguishing between surface and atmospheric contributions is one of the main challenges in aerosol retrieval on land. Due to this fact, for a long time, aerosol retrieval over land surface yielded poorer results compared to ocean mapping (REMER et al., 2005). Currently, proposed algorithms can separate atmospheric and surface contributions, demonstrating high performance (LEVY et al., 2013; LYAPUSTIN et al., 2018; REMER et al., 2013a).

3.4.3 Satellite sensors used for AOD retrieval

Due to technical and scientific advancements in remote sensing, satellites are increasingly used to gather information on aerosol properties (e.g., AOD, columnar particle concentration, and particle sizes). Fraser et al. (1984) successfully conducted the first retrieval of AOD using observations from the Geostationary Operational Environmental Satellites (GOES) in the eastern United States. Since then, several algorithms for AOD retrieval have been developed using remote sensing data, including data from the MODerate-resolution Imaging Spectroradiometer (MODIS), the Multi-angle Imaging SpectroRadiometer (MISR), the Geostationary Operational Environmental Satellites (GOES), the Polarization and Directionality of the Earth's Reflectance (POLDER), the Sea-viewing Wide Field-of-view Sensor (SeaWiFS), the Ozone Monitoring Instrument (OMI), and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). Table 3.3 presents the characteristics of the main sensors used for AOD retrieval.

Table 3.3 – Characteristics of the primary sensors capable of detecting aerosol optical depth

Sensor ¹	Satellite ²	Orbit	Spatial resolution (AOD)	Temporal resolution (AOD)*	Spatial scale	Data initial	Data final
MODIS	Aqua/Terra	LEO	1 - 10 km	Daily	Global	2000	present
VIIRS	Suomi-NPP	LEO	0.75 - 6km	Daily	Global	2011	present
MISR	Terra	LEO	17.6 km	Monthly	Global	2000	present
SeaWiFS	OrbView-2	LEO	13.5 km	Daily	Global	1997	2010
SCIAMACHY	ENVISAT	LEO	60×30 km	Daily	Global	2002	2012
OMI	Aura	LEO	24x13 km	Daily	Global	2004	present
POLDER/ PARASOL	ADEOS I ADEOS-II	LEO	1/6°	Daily	Global	1996	2013
GOME-2	MetOp	LEO	10x40 km	Daily	Global	2006	present
GOCI	COMS	GEO	6 km	1 hr.	Regional (East Asia)	2010	present
ABI	GOES-R	GEO	2 km	5 and 15 min	Regional (North and South America and the Atlantic Ocean to the west coast of Africa)	2016	present
AHI	Himawari-8	GEO	5 km	10 min and 1hr.	Regional (East Asia, Southeast Asia, parts of South Asia, and Oceania)	2014	present
AGRI	FY-4	GEO	2 km	15 min	Regional (China [3–55° N, 70–140° E])	2016	present
MI	COMS	GEO	4 km	15 min	Full-disk (North hemisphere)	2010	present
CCD	INSAT	GEO	10 km	30 min	Regional (Asian sector [-10–45°N, 45–105° E])	2010	present

*More refined temporal resolution; LEO (Low-earth orbiting), GEO (Geostationary earth orbit), (1) Sensor: MODIS, MODerate resolution Imaging Spectroradiometer; VIIRS, Visible Infrared Imaging Radiometer Suite; MISR, Multi-angle Imaging SpectroRadiometer; SeaWiFS, Sea-viewing Wide Field-of-view Sensor; OMI, Ozone Monitoring Instrument; POLDER, Polarization and Directionality of the Earth's Reflectance instrument; PARASOL, Polarization and Anisotropy of Reflectances for Atmospheric Sciences coupled with Observations from a Lidar; GOME, Global Ozone Monitoring Experiment; GOCI, Geostationary Ocean Color Imager; ABI, Advanced Baseline Imager; AHI, Advanced Himawari Imager; AGRI, Advanced Geosynchronous Radiation Imager; MI, Meteorological Imager; CCD, Charge Coupled Device; (2) Satellite: Suomi-NPP, Suomi National Polar-orbiting Partnership; ENVISAT, ENVironmental SATellite; ADEOS, Advanced Earth Observing Satellite; MetOp, Meteorological Operational satellite programme; COMS, Communication, Ocean and Meteorological Satellite; GOES, Geostationary Operational Environmental Satellites; FY-4, FengYun-4; INSAT, Indian National Satellite System.

Source: GOES-R, Lalitaporn et al. (2013), Choi et al. (2016), Kim et al. (2016), Formenti et al. (2018), NOAA (2018), Dubovik et al. (2019), Sogacheva et al. (2019) e ZHANG et al. (2019a, 2019b).

Among the primary sensors, MISR, aboard the Terra satellite, provides nine views of each pixel, with spatial resolution ranging from 275 m to 1.1 km and four spectral bands between 0.446 μm and 0.866 μm (VAN DONKELAAR et al., 2016). The MISR instrument's bandwidth is approximately 380 km, taking about a week for complete global coverage at mid-latitudes. MISR has demonstrated spectral stability and has been used for AOD data retrieval applications over the past two decades (DI GIROLAMO et al., 2004; KAHN et al., 2005; VAN DONKELAAR et al., 2016; VAN DONKELAAR; MARTIN; PARK, 2006).

The SeaWiFS instrument was in operation between 1997 and 2010. With a bandwidth of 1500 km, this sensor provided almost daily global observation in 8 spectral bands, between 0.402 and 0.885 μm , with a minimum spatial resolution of 1.1 km. Its radiometric calibration remained stable throughout its operational lifespan. Previous studies suggest that SeaWiFS

v003 data are suitable for estimating interannual changes in AOD at global and regional scales (HSU; CHANG; LIN, 2016; LI et al., 2009).

The MODIS sensors have 36 spectral bands with different spatial resolutions (250, 500, and 1000 m) and an imaging swath width of 2,330 km, providing near-daily global coverage for each instrument (MHAWISH et al., 2019). The sensor has been capturing images of the Earth's surface since 2000 and currently has three algorithms for AOD retrieval. Thus, due to the maturity of AOD retrieval algorithms and near-daily data coverage, as well as providing a long data record, the sensor has been widely used in scientific research aiming to estimate PM_{2.5} data from satellite observations (VAN DONKELAAR et al., 2016; VAN DONKELAAR; MARTIN; PARK, 2006; WU; GRAAF; MENENTI, 2016).

3.4.4 MODIS-AOD datasets

On board the Terra and Aqua satellites, MODIS has acquired TOA data since 1999 and 2002, respectively, being the first sensor designed to provide global aerosol optical characteristics. The MODIS sensor has 36 spectral bands, ranging in wavelength from 0.4 μm to 14.4 μm on both satellites. The sensor has a temporal resolution of 1 to 2 days and moderate spatial resolutions of 250 m, 500 m, and 1000 m. Out of the 36 spectral bands available in the sensor, seven have primary usage focused on aerosols, with wavelengths ranging from 0.47 μm to 2.13 μm (corresponding to the VIS, NIR, and SWIR regions of the spectrum) (NICHOL; BILAL, 2016). The characteristics of these bands are presented in Table 3.4.

Table 3.4 - Characteristics of the primary MODIS sensor spectral bands used in aerosol retrievals

MODIS - band	Wavelength range (μm)	Central wavelength (μm)	Spatial resolution (m)
1	0.620 – 0.670	0.646	250
2	0.841 – 0.876	0.855	250
3	0.459 – 0.479	0.466	500
4	0.545 – 0.565	0.553	500
5	1.230 – 1.250	1.243	500
6	1.628 – 1.652	1.632	500
7	2.105 – 2.155	2.119	500

Source: Adaptado de Levy et al. (2007).

In the last decades, the AOD retrieval algorithms of the MODIS sensor have undergone several modifications. Currently, three distinct and independent algorithms are routinely used for extracting this information, namely: Dark Target (DT) (LEVY et al., 2013; LEVY; REMER; DUBOVIK, 2007; REMER et al., 2005; TANRÉ et al., 1997), Deep Blue (DB) (HSU et al.,

2013, 2004), and Multiangle Implementation of Atmospheric Correction (MAIAC) (LYAPUSTIN et al., 2011, 2018).

3.4.4.1 DT

The Dark Target (DT) algorithms used for AOD retrieval from the MODIS sensor over land and ocean are based on the algorithm developed for AOD retrieval from the Advanced Very-High-Resolution Radiometer (AVHRR) sensor (REMER et al., 2013b). The DT algorithms for AOD retrieval over the ocean (DT-ocean) and land (DT-land), as adequately described in Levy et al. (2013), were initially designed for the retrieval of MODIS sensor data and have been applied immediately after the launch of the Terra satellite.

The DT-ocean algorithm is based on converting TOA reflectance from dark surface targets into aerosol information. To achieve this, six bands of the MODIS sensor are used (0.55, 0.65, 0.86, 1.24, 1.63, and 2.11 μm). The strategy for AOD retrieval is based on a Look-Up Table (LUT) approach, where radiative transfer calculations are pre-computed for various values of aerosol and surface parameters (REMER et al., 2005). The spectral reflectance observations at the six wavelengths are compared with the LUT reflectance, which contains four possibilities for the fine mode and five possibilities for the coarse mode (each possibility, along with its characterization, is presented in Remer et al. (2005)). Thus, the reflectances are used with the LUT, taking into account the specific illumination and sensor viewing geometry, to obtain three parameters: (1) total aerosol optical depth (AOD) at 0.55 μm , (2) fraction of this optical depth that is due to the fine mode, and (3) effective radius of the aerosol size distribution integrated in the atmospheric column (CORREIA et al., 2007). Using the obtained value for the aerosol optical depth at 0.55 μm and the spectral dependency model presented in Remer et al. (2005), it is obtained the AOD value at the other wavelengths.

The DT-land algorithm, like DT-ocean, compares TOA measurements with LUT values. Additionally, DT-land is also based on retrieving the AOD from TOA over a dark surface target (REMER et al., 2013b). Over the ocean, DT-ocean assumes a homogeneous surface, making assumptions about the spectral reflectance of aerosols easier. However, the reflectance is highly variable over land due to different types of land cover (varying vegetation and urbanization), which requires more complex assumptions. Therefore, as some land surface targets are highly reflective, making certain spectral bands impractical, the retrieval is limited to three bands (0.47, 0.66, and 2.12 μm) (LEVY et al., 2013). To retrieve AOD data, the DT-land algorithm

selects dark pixels (500 m resolution) with TOA reflectance ranging from 0.01 to 0.25 in the 2.12 μm band (MHAWISH et al., 2019). In addition, after cloud, water, snow/ice, and bright pixel masking, the algorithm also selects and discards pixels using the 0.65 μm band (discarding the darkest 20% and the brightest 50% of pixels), aiming to eliminate issues with cloud shadows, residual cloud contamination, or unique surface conditions (CORREIA et al., 2007). Finally, the reflectance of the remaining pixels is calculated as an average (within each 10x10 km or 3x3 km grid) and used to derive the AOD (MHAWISH et al., 2019). The DT-land algorithm uses three fine modes (low, moderate, and highly absorbing) and one coarse mode, with the selection dependent on the season and geographic location (CORREIA et al., 2007).

Based on the DT algorithms, AOD products are generated at two spatial resolutions (10 and 3 km). In summary, both products share the same retrieval protocol, except for selecting pixels for which the retrieval is performed. In the 10 km product, 20 x 20 pixels of 500 m are arranged in each 10x10 km grid cell; the 3 km product combines measurements from 6 x 6 pixels in each 3x3 km grid cell (REMÉR et al., 2013a). The retrieval quality is determined by the number of pixels used in each cell of the 10x10 km or 3x3 km grid.

3.4.4.2 DB

Unlike DT, the Deep Blue (DB) algorithm can be applied to recover AOD over bright surfaces, including deserts, and is sensitive to aerosol absorption and extinction (REMÉR et al., 2013b). DB retrieves AOD under the assumption that the reflectance of bright surfaces (e.g., urban areas and deserts) is relatively lower in the blue band (0.412 μm) compared to longer visible wavelengths (HSU et al., 2004). In addition to being applied to bright surfaces, the new version of the algorithm (described in detail by Hsu et al. (2013)) extends the retrievals to vegetated and/or dark surfaces. For this purpose, the surface reflectance is determined using a hybrid approach based on a global surface reflectance database in visible bands (e.g., 0.412, 0.47, and 0.65 μm), depending on the location, season, and land cover type (HSU et al., 2013). The surface reflectance dataset used by DB is based on the minimum reflectivity approach and grouped by scattering angle, season, and normalized difference vegetation index (NDVI). In vegetated/dark regions, DB follows a similar approach to DT. The hybrid method scales the surface reflectance by applying near-surface atmospheric correction near the locations with surface AOD measurements (AERONET network, AEROSOL ROBOTIC NETWORK). For this purpose, it uses the regional bidirectional reflectance distribution function (BRDF) distribution function (JETHVA; TORRES; YOSHIDA, 2019).

3.4.4.3 MAIAC

DT and DB are standard AOD products developed for the MODIS sensor, and both are applied right after the satellite data (Aqua and Terra) reach the processing center. In other words, the AOD retrieval by these algorithms is done independently, without considering observations made over time. On the other hand, the Multiangle Implementation of Atmospheric Correction (MAIAC) processing algorithm is based on the dynamic analysis of time series (LYAPUSTIN et al., 2011, 2018).

MAIAC assumes that surface reflectance is relatively stable over short time intervals but exhibits high spatial variability. While other atmospheric components, such as aerosols and clouds, change rapidly over time, their spatial variation is limited (LIU et al., 2019a). To achieve this goal, MAIAC uses a time series analysis of images spanning several days (4 days at the poles and 16 days at the equator) and different spatial scales for processing (e.g., 1 km grid cells (pixels), 25 km blocks, and 150 km mesoscale areas) (LYAPUSTIN et al., 2018). In its analyses, MAIAC utilizes 12 spectral bands (B1-B7 in Table 3.4, B8-0.412, B9-0.442, B10-0.487, B11-0.530, and B12-0.547); the 500m and 250m bands are nested within the 1 km grid. Each daily observation is stored in a dedicated memory that accumulates auxiliary information for each 1 km grid cell.

The MODIS measurements' temporal series (4 to 16 days) are used to characterize the surface through the Ross-Thick Li-Sparse BRDF model (LUCHT; SCHAAF; STRAHLER, 2000) using measurements taken in the NIR (2.12 μm). Next, the linear spectral regression coefficient (SRC) between 0.47 and 2.12 μm is calculated for each 1 km grid, providing the separation between atmospheric and surface contributions (LYAPUSTIN et al., 2018). Finally, the AOD parameter at 0.47 μm can be retrieved by analyzing theoretical TOA reflectance in the LUT (LIU et al., 2019a). The AOD is initially retrieved at 0.470 μm , and the AOD parameter at 0.55 μm is calculated based on the spectral properties of the aerosol model used in the retrievals (the model is identified through the LUT) (LYAPUSTIN et al., 2018). More information about the MAIAC algorithm to retrieve AOD from the MODIS sensor can be obtained from Lyapustin et al. (2018).

4 SCOPE OF THE THESIS

This study is organized in the form of chapters. Thus, the first three chapters provide the introduction items, objectives, and theoretical framework, bringing the basic concepts, motivations and justification for the development of this research. The next four chapters are devoted to the results of the procedures performed to meet the research objectives. The methodological sequence is illustrated in Table 4.1, which presents the questions that guide the strategies adopted throughout the research. Each method execution step will be described in the following chapters.

Table 4.1 – Methodological scheme of the strategies adopted to carry out this research.

	Guiding questions	Objective(s)	Phase status
Phase 1	stage 1 Were there mobility restrictions in Brazil?	Assessing the influence of mobility restrictions implemented to contain the transmission of COVID-19 on atmospheric pollutant concentrations in Brazil.	The methodology and results of this phase are available in Chapter 5. This chapter has been published by the Aerosol and Air Quality Research journal and can be accessed through the DOI: 10.4209/aaqr.210351.
	stage 2 Did the restrictions impact air quality?		
	stage 3 Are the variations in air quality similar regionally?		
Phase 2	stage 1 How did mobility restrictions impact the air quality in the state of São Paulo?	Evaluate how mobility restrictions implemented to contain the COVID-19 pandemic affected air quality in the state of São Paulo and investigate the role of atmospheric conditions in this context.	The methodology and results of this phase are available in Chapter 6. This chapter has been published by the Environmental Research journal and can be accessed through the DOI: 10.1016/j.envres.2021.111255.
	stage 2 Are the variations in air quality significant for long periods?		
	stage 3 Do atmospheric conditions have an impact on the observed variations?		
Phase 3	stage 1 Are the data on the tropospheric column density of NO ₂ with the best spatial/temporal resolution correlated with surface pollutant concentrations?	Verify the monitoring capability of PM and NO₂ using remotely sensed data traditionally in use.	The methodology and results of this phase are available in Chapter 7. This chapter has been published by the Remote Sensing of Environment journal and can be accessed through the DOI: 10.1016/j.rse.2023.113514.
	stage 2 Are the data on the AOD with the best spatial/temporal resolution correlated with PM concentrations?		
	stage 3 What factors are related to data availability and accuracy?		
Phase 4	stage 1 Do land cover types influence the retrieval of AOD?	Evaluate the accuracy of the retrieved AOD products using MODIS sensor data.	The methodology and results of this phase are available in Chapter 8.
	stage 2 Does the AOD (solar/sensor) retrieval angle and period of year affect data quality?		
	stage 3 Which AOD dataset best represents the study region?		

5 IMPACTS OF STRATEGIC MOBILITY RESTRICTIONS POLICIES DURING 2020 COVID-19 OUTBREAK ON BRAZIL'S REGIONAL AIR QUALITY¹

Abstract: With the unexpected arrival of the COVID-19 pandemic, countries worldwide were forced to take measures to curb its transmission. Mobility restrictions policies were the primary preventive measures applied around the globe. In addition to reducing the disease spread, they resulted in air quality changes in urbanized areas. In this sense, this study aimed to investigate how the mobility restrictions imposed by public policies impacted the air quality in Brazil during the COVID-19 2020 outbreak. Therefore, air quality (CO, NO₂, PM₁₀, PM_{2.5}, and O₃) and urban mobility datasets available in five populous Brazilian states (São Paulo, Rio de Janeiro, Espírito Santo, Paraná, and Rio Grande do Sul) were analyzed. Variations in air pollutant concentrations were verified by comparing the period during mobility restrictions (2020) and the average concentrations found before the restrictions (2015-2019). In addition, spatial assessment of changes was evaluated using tropospheric NO₂ column densities from the TROPOMI. Although there was no national regulation regarding mobility restrictions, the results show that the reduction in urban mobility was similar for all the analyzed states. Following the mobility behavior, reducing air pollutant concentrations were significant for the first 30 days of restrictions. During this period, the most substantial reductions were observed for CO in the State of Rio Grande do Sul (-53%), NO₂ in Rio de Janeiro (-34%), and PM₁₀ in Espírito Santo (-23%). The exception was observed for O₃, which followed the world trend of increasing concentrations (e.g., 40% increase in Paraná). Spatially, it was possible to evidence that large urban centers (with a large vehicle fleet) were the ones that showed the most significant reduction in NO₂. However, when analyzing longer periods (over 90 days), there is a trend towards an increase in the concentrations of primary pollutants and a consequent reduction in O₃, reflecting the significant increase in mobility rates.

Keywords: Air pollution; Lockdown; Coronavirus; Brazilian policies.

5.1 Introduction

Currently, global society faces the most significant health and humanitarian crisis in its recent history: the COVID-19 pandemic, caused by the SARS-CoV-2 virus (PAK et al., 2020; THE

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LANCET, 2020b). The high transmission potential of this virus through the air and via personal contact soon demanded a global alert, mainly to prevent the spread of the virus (FAN et al., 2020a), since the advance of the disease was accompanied by a concomitant crisis in the health systems (BLUMENTHAL et al., 2020; DE OLIVEIRA ANDRADE, 2020). Thus, most governments worldwide adopted policies such as keeping a safe distance, wearing masks, using alcohol-based products to disinfect hands, staying home, and banning social and public events. In this context of preventive measures, mobility restrictions proved to be very effective in containing the transmission of the virus (MARTINS et al., 2020; WANG et al., 2020b). In addition, they resulted in a reduction in air pollution in several regions (BAUWENS et al., 2020; CHOSSIÈRE et al., 2021; HE; WANG; YIM, 2021; MORALES-SOLÍS et al., 2021; RANA; KERAMAT; GOW, 2021; ZAMBRANO-MONSERRATE; RUANO; SANCHEZ-ALCALDE, 2020).

The reduction of air pollution levels, due to a decrease in mobility, has proven to be an “open-air laboratory” and, as a consequence, has been experimentally investigated by several academic studies (BAO and ZHANG, 2020; COLLIVIGNARELLI et al., 2020; DUTHEIL et al., 2020; LOKHANDWALA and GAUTAM, 2020; SHARMA et al., 2020). These studies have shown that the levels of urban pollution reduction are essentially related to local economic vocation and the degree of restrictions imposed on social distancing (CHOSSIÈRE et al., 2021). Thus, while several locations showed significant improvements in air quality during the pandemic (BAO; ZHANG, 2020; DUTHEIL; BAKER; NAVEL, 2020; LOKHANDWALA; GAUTAM, 2020; MISHRA; KULSHRESTHA, 2021), others presented a positive correlation between the increase in the number of cases of COVID-19 and the high levels of pollution (CONTICINI; FREDIANI; CARO, 2020; TRAVAGLIO et al., 2021).

In Brazil, several studies have been carried out to understand the effects of the pandemic on air quality (BAINY et al., 2021; DANTAS et al., 2020; MANTOVANI et al., 2021; ROCHA et al., 2021; SILVA JÚNIOR et al., 2020). These surveys are mainly concentrated in the Southeast region of Brazil, especially in the metropolitan areas of São Paulo and Rio de Janeiro (DEBONE; DA COSTA; MIRAGLIA, 2020; KRECL et al., 2020; NAKADA; URBAN, 2020). Some of these studies found that, in general, there was an increase in concentrations of pollutants in more industrialized regions. In contrast, there was a reduction in air pollutants in areas with a large fleet of vehicles (NAKADA; URBAN, 2020; RUDKE et al., 2021a).

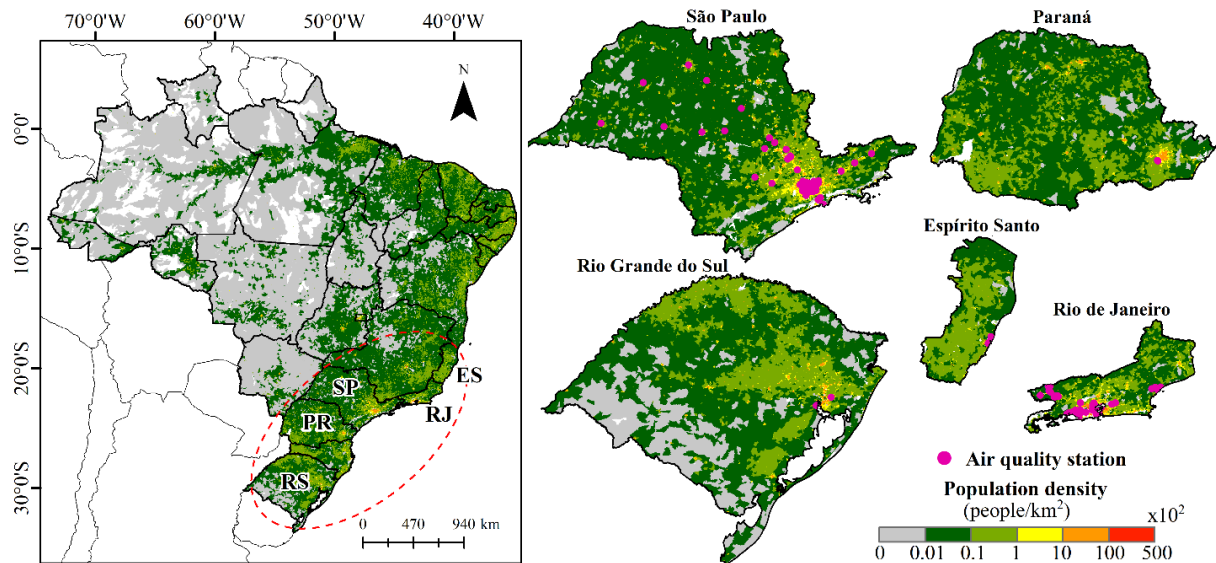
At the moment of writing this text, Brazil is the second country most affected by the pandemic, counting about 25 million cases and more than 625 thousand deaths (MS, 2021). There was no clear position in favor of lockdown or mobility restrictions at the federal level, even with the worsening of the health crisis and the high number of contamination and deaths. The governors of each State were left to arbitrate on local control policies (Barberia e Gómez, 2020; Boschiero et al., 2021; Russo Rafael et al., 2020; Touchton et al., 2021), revealing different scenarios of mobility restrictions. Overall, the adoption of tighter restrictions positively impacted the health system (ALFANO; ERCOLANO, 2020; LAU et al., 2021; MEO et al., 2020; WILDER-SMITH; BAR-YAM; FISHER, 2021). With a very variable range of restrictive measures, quite different from other countries, this scenario offers a unique opportunity to understand the impact of such measures on air pollution. In this sense, this study aimed to evaluate the impact of the mobility restrictions implemented during the 2020 COVID-19 outbreak on the regional air quality of five Brazilian States (São Paulo, Rio de Janeiro, Espírito Santo, Paraná, and Rio Grande do Sul; distributed in the most densely populated regions of the country - Southeast and South). In more specific terms, this study sought to assess how mobility restrictions affected the concentrations of air pollutants at different time intervals and how these changes are spatially distributed.

5.2 Methods

5.2.1 Atmospheric conditions

Brazil is the country with the largest territory in South America, with an area of about 8.5 million km² (IBGE, 2020a). The country borders to the west with most countries of the continent and, to the east, with the Atlantic Ocean (Figure 5.1). The country is geographically divided into 27 federative units (twenty-six States and the Federal District) and has an estimated population of more than 210 million inhabitants (IBGE, 2021a), with a population density of 22.43 inhabitants per km². The Southeast and South Regions are the most densely populated, followed by the Northeast, Central-west, and North (IBGE, 2020b). The South, Southeast, and Central-west Regions have the most extensive agricultural and livestock activities areas. The largest area of forests remnants is concentrated in the northern part of the country, mainly due to the Amazon rainforest. Besides, the Southeast Region is the most industrialized area in the country, with São Paulo being the State with the most prominent industrial production (NAKAZATO; RINALDI; DOMINGOS, 2015; VIEIRA-FILHO; LEHMANN; FORNARO, 2015).

Figure 5.1 - Study area and distribution of the air quality monitoring stations by state. SP: São Paulo, PR: Paraná; RS: Rio Grande do Sul; ES: Espírito Santo; RJ: Rio de Janeiro. The red dashed ellipse highlights the South and Southeast Regions of Brazil.



Stationary sources play a fundamental role in the air pollutants levels in Brazil (KAWASHIMA et al., 2020); however, mobile sources are the primary sources of air pollution in large urban centers (Almeida et al., 2018; Andrade et al., 2012, 2017; Moreira et al., 2018; Rudke et al., 2021). The country has a fleet of more than 106 million vehicles - considering light and heavy-duty vehicles (DENATRAN, 2020a). There are a few air pollution inventories in the country, mostly restricted to vehicular emissions and some urban areas (ANDRADE et al., 2017; CARVALHO et al., 2015; RAFEE et al., 2017; RÉQUIA; KOUTRAKIS; ROIG, 2015; SOUZA et al., 2013; TSAO et al., 2012). In Brazil, most vehicles are powered by diesel, gasoline (gasoline+ethanol), and ethanol. The first use of ethanol as fuel was in the 1920-1930s (adding ethanol to gasoline). In Brazil, it has been used since the 1970s (blended and pure), and later also the use of ethanol in the flex-fuel vehicles resulted in the reduction of some atmospheric pollutants (BRITO et al., 2018; SALVO et al., 2017). However, biofuels can have more significant impacts on regional climate forcings and human health than previously thought (TSAO et al., 2012b).

This study focuses on the South and Southeast Regions of Brazil, more specifically the examined data correspond to São Paulo (SP), Paraná (PR), Espírito Santo (ES), Rio de Janeiro (RJ) and Rio Grande do Sul (RS) States (Figure 5.1). Together, these five States account for approximately 42.8% of the Brazilian population (IBGE, 2021b) and encompass the largest metropolises in the country, such as São Paulo and Rio de Janeiro, which gather the most

significant vehicle fleets (DENATRAN, 2020b). With the increase in vehicular emissions and few works aimed at reducing air pollution for Brazil, the analysis of the air pollutants levels is a significant contribution to the country.

5.2.2 Datasets

6.2.2.1 Ground-Based Data

The air quality data used in this study was obtained through the Institute of Energy and Environment (in Portuguese *Instituto de Energia e Meio Ambiente* – IEMA; <http://energiaeambiente.org.br/qualidadedoar>). The IEMA is a Brazilian Non-profit Organization that integrates, organizes, analyses, and makes available air quality monitoring data carried out in Brazil. The IEMA air quality platform currently provides monitoring data from 2000 onwards for ten States and the Federal District.

Given the need for data available for 2020, only monitoring stations that had measurements carried out for this year were considered. Another criterion used to select stations was the need for at least 50% of the data available between 2015 and 2019. Based on these criteria, 97 automatic air quality stations, available in the same five Brazilian States, were selected for the evaluations carried out in this research (Figure 5.1). The IEMA data for these monitoring stations were obtained from the State Environmental Institute of Espírito Santo (IEMA-ES), Water and Land Institute (IAT) of the State of Paraná, State Environmental Institute (INEA) of Rio de Janeiro, Environmental Protection Foundation (FEPAM) of Rio Grande do Sul State and, Environmental Company of the State of São Paulo (CETESB). The selected stations are capable of monitoring ground-based concentrations of carbon monoxide (CO – 41 stations), ozone (O₃ – 75 stations), particulate matter (PM₁₀ – 73 stations; PM_{2.5} – 17 stations), and nitrogen dioxide (NO₂ – 53 stations). More information about the stations used is available in Table A.1 (Appendix A).

6.2.2.2 Satellite Data

As air pollutant ground-level concentrations are only available for sites with monitoring stations, Tropospheric NO₂ column densities measured by the TROPospheric Monitoring Instrument (TROPOMI) were used to verify the spatial trend variations in air quality. The TROPOMI sensor is coupled to the Sentinel-5 Precursor (S-5P) satellite. The Tropospheric NO₂ column density dataset has about 2600 km of swath width, resulting in almost daily global

coverage (equator crossing time at 13:30 LT) (VEEFKIND et al., 2012). The Tropospheric NO₂ column densities data from TROPOMI showed promising results compared to measurements carried out on the ground (Verhoelst et al., 2021). Furthermore, the data has demonstrated excellent correlations with ground-based NO₂ concentrations acquired from air quality monitoring stations (Bassani et al., 2021; Cersosimo et al., 2020; Kim et al., 2021; Virta et al., 2021).

As Tropospheric NO₂ column densities are not available for the entire study period (data available since the end of April 2018), only data from 2019 and 2020 were used in the analyses. The Level 2 OFFL version product was adopted in this study, with a spatial resolution of 5.5×3.5 km² at the nadir. These data were statistically processed and downloaded from the Google Earth Engine platform. During the analysis, only NO₂ information with a quality assurance value (greater than 0.75) was used, following the guidelines recommended by the producers (ESKES et al., 2019).

6.2.2.3 COVID-19 cases and Mobility Data

Data from the Google Community Mobility Reports were analyzed to assess the mobility restrictions policies adopted by the Brazilian States during the COVID-19 pandemic. The data is freely available (<https://www.google.com/covid19/mobility/>) and contains anonymous information on movement trends obtained from smartphone users during their travels. The platform provides movement trend information for the categories “Grocery & pharmacy”, “Parks”, “Transit stations”, “Retail & recreation”, “Residential” and, “Workplaces”. To demonstrate variations during the analyzed period, we adopted the “Transit stations” class that presents the mobility trends for public transport hubs (e.g., subway, bus, and train stations), as this would be the class most impacted by public policies aimed at social distancing. According to the producers, the daily percentage changes are calculated based on the reference day, which is the median value of the five weeks between 3rd January and 6th February 2020.

Daily data on new confirmed cases of COVID-19 were obtained through the dissemination platform of the Brazilian Ministry of Health (<https://covid.saude.gov.br/>). The platform disseminates the information that is collected at the municipal and State levels. As the analyzed States have very different number and spatial distribution of populations, it was decided to normalize the data of new cases of COVID-19 according to the number of inhabitants in each State. Thus, the information used was daily confirmed new cases per 100 thousand people.

6.2.2.4 Synoptic conditions

It is known that meteorological conditions substantially impact air pollutants levels; precipitation, for example, plays an important role in removing air pollutants from the atmosphere. Therefore, to assess the variation in the behavior of pollutant concentrations caused by mobility restrictions in Brazil during the COVID-19 pandemic, it is essential to check the meteorological conditions, as demonstrated by (Rudke et al., 2021).

The synoptic conditions were verified considering the European Center for Medium-Range Weather Forecasts (ECMWF) Reanalysis version 5 (ERA5) (HERSBACH et al., 2020a) (available in <https://www.ecmwf.int/en/forecasts/dataset/ecmwf-reanalysis-v5>). The ERA5 reanalysis provides hourly data and covers the globe on a 30 km horizontal grid. In this latest release, the atmosphere is resolved using 137 levels from the surface up to a height of 80 km, capturing finer details of atmospheric phenomena (HERSBACH et al., 2020b).

Based on the hourly Convective Available Potential Energy (CAPE) and Precipitable Water (PWater) from ERA5, it was evaluated whether there were significant differences in meteorological conditions for the first semester of 2020 in comparison with the same period for 2019. In this case, the Student T-test was the statistical method used for testing the hypothesis, considering a confidence level of 95% ($\alpha=0.05$).

5.3 Results and discussion

5.3.1 Synoptic conditions

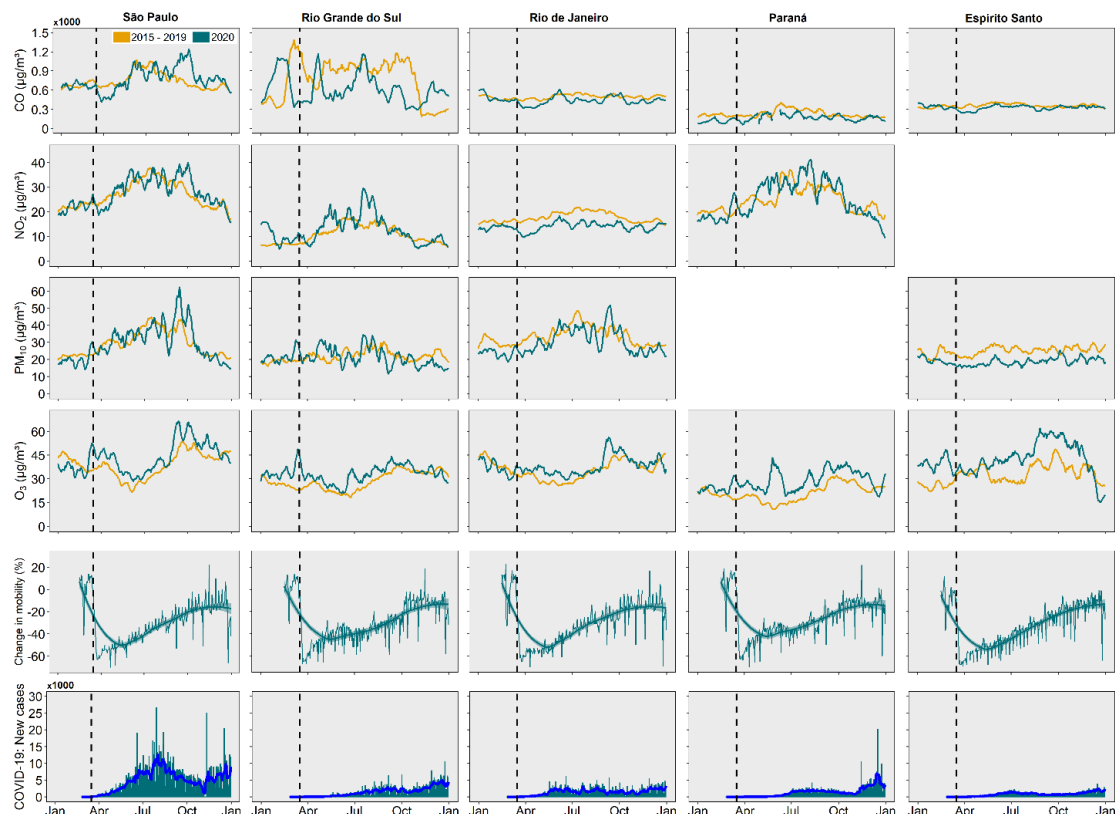
Student's t-test results analysis demonstrates that, in general, the energy available for deep convection and moisture was statistically different between the first semester of 2019 compared to 2020 (CAPE: t-value - 25.0 and p-value - <0.01 ; PWater: t-value - 2.3 and p-value - 0.02). Higher CAPE and Pwater values were observed for 2019 (see Figure A.1, Appendix A). Therefore, it can be inferred that the prevailing atmospheric conditions in 2019 were more prone to mixing the atmospheric mass than in 2020. Thus, it is possible to conclude that during the first semester of 2019, there was a higher dissipation of pollutants caused by meteorological effects when compared to 2020. This fact was also observed by Rudke et al. (2021) in a study carried out for the State of São Paulo. The authors indicated that the greater volume of rainfall in 2019 when compared to 2020, prevented the observation of significant reductions in air pollutant concentrations during the period of restrictions caused by the COVID-19 outbreak.

Consequently, the analysis of expected decreases in the concentration of air pollutants must consider the differences observed for the atmospheric conditions acting in the period before and during the COVID-19 pandemic.

5.3.2 Changes Observed in Air Quality and Mobility

To assess changes in air pollutant concentrations, the comparison between observed values for 2020 and the five years before the pandemic (2015-2019) was considered. The interval between 2015-2019 was supposed to minimize interannual variations, which occur even in years without a pandemic, mainly due to atmospheric conditions and economic factors. Thus, hereafter these periods will be compared to demonstrate the impact of mobility restrictions due to the COVID-19 outbreak on air pollutant concentrations observed for Brazil. In this sense, Figure 5.2 presents 13-day centralized moving averages for the evaluated pollutants (CO, NO₂, PM₁₀, and O₃; PM_{2.5} data for the States of São Paulo and Rio de Janeiro are available in Figure A.2). The figure also shows the mobility trend observed during the pandemic and information regarding new confirmed cases of COVID-19.

Figure 5.2 - 13-day centralized moving averages considering air pollutants concentration before (yellow) and during (green) the COVID-19 pandemic; percentage in mobility change (in blue patterns in-transit movements); and daily trends in the number of new confirmed cases of COVID-19 in Brazil (in blue line are the weekly moving averages), separated by five States. The dashed line indicates the date 16th March 2020, representing the beginning of Brazil's mobility restrictions.



According to Figure 5.2, it is noted that, in general, even before the first mobility restrictions adopted in Brazil (1st January to 15th March), below-average concentrations (2015-2019) were found for 2020. The precipitation regime observed for 2020 can partially explain this fact since it was generally higher than the average from 2015 to 2019 (Figure A.3), except for Paraná and Rio Grande do Sul, which had lower rainfall for 2020. Atypical weather conditions were observed by Martins et al. (2020), with rainfall above average in North and Northeastern Brazil (e.g., 446.3 mm for March in Manaus-MA, which is about 50% higher than the expected rainfall for the month) and drier and warmer atmospheric conditions in Southern Brazil (e.g., 13.8 mm for March in Curitiba-PR, while the expected is 125 mm).

During the suspension of in-person activities decreed by the States, which started on 16th March 2020 for practically all Brazilian States (DANTAS et al., 2020; GERALDI et al., 2021; RUDKE et al., 2021a), there was a sharp reduction in most pollutants for most States, following the mobility behavior. These more accentuated reductions are mainly observed in the States of São Paulo and Rio de Janeiro, where the behavior of air pollutants directly followed the initial changes in mobility. It occurs because most of its monitoring stations are located in their metropolitan areas, where vehicular traffic dominates the source of outdoor air pollution (CARVALHO et al., 2015; DANTAS et al., 2020), differently, for example, from Paraná, where the available stations are located in an industrial area.

In agreement with the state information portals on COVID-19, during 2020, there were still some mobility policies that were even more restrictive, as in the case of the States of São Paulo and Paraná, which during some holidays even closed essential services and stipulated prohibition of displacement during the night period. It is important to emphasize that in 2020 in Brazil, unlike in other countries, there was no measure of strict mobility restrictions as the lockdown. In 2021, the city of Araraquara (in upstate São Paulo) was the first one in Brazil to use such a measure, which proved to be quite effective (FIGUEIREDO FILHO; SILVA, 2021; MALTA et al., 2021), once it reduced the number of deaths in the city up to 80% (FRANCISCO et al., 2021).

In all the five States studied, the concentration of O₃ was higher in 2020 than the average from 2015 to 2019. This increase has also been observed worldwide and may be related to the chemical formation regime of O₃ (COLLIVIGNARELLI et al., 2020; DUTTA; JINSART, 2021; GAO et al., 2021; GRANGE et al., 2021; LU et al., 2013; SHARMA et al., 2020;

SICARD et al., 2020), which in large cities is usually Volatile Organic Compounds (VOC) limited. According to (Siciliano et al., 2020b), who evaluated the increase of O_3 in Rio de Janeiro city, the greatest concentrations result from higher ratios of non-methane hydrocarbons (NMHC) and nitrogen oxides (NO_x), i.e., more significant reduction of NO_x than of NMHC.

In general, during 2020, the measures imposed by the State governments were similar between all of them: restricting social events, regulating the occupation of people in shops, restaurants, gyms, as well as adopting teleworking for administrative services, and remote classes, among several other measures of restriction or total prohibition to avoid or reduce agglomerations of people. However, the intensity of measures and the commitment of the population were variable (Figure 5.2). This occurred due to measures of "relaxation" of the mobility restriction policies adopted by the State governments along the time, in addition to the mismatch between the federal, State, and municipal spheres, which generated uncertainties in the population, favoring the reduction of social distancing. During this period, many closed shops were reopened (with reduced opening hours), which significantly increased the flow of people in large urban centers. This fact induced an increase in mobility and consequently in air pollutants concentrations, generating substantial growth in the number of new cases of COVID-19, which first peaked in mid-July to September (Figure 5.2). Besides that, during the pandemic, some industrial activities were intensified, increasing concentrations of NO_x , SO_2 , CO_2 , and CO (NAKADA; URBAN, 2020; RUDKE et al., 2021a).

Table 5.1 presents the relative differences (%) for air pollutants and mobility in different time intervals (extended version of the table can be accessed in the Appendix A - Table A.2). It is seen that the most significant reductions in primary pollutants (NO_2 and CO) and PM are kept for shorter time intervals, where the mobility restrictions policies were more stringent. On the other hand, when more extended periods are observed, it is possible to perceive a smaller impact of mobility restrictions on the concentrations of these pollutants.

Table 5.1 – Relative differences (%) in air pollutant concentrations and mobility averaged for the five Brazilian States analyzed.

Pollutant		15 days		30 days		90 days		180 days		290 days	
		Conc.	Mobility	Conc.	Mobility	Conc.	Mobility	Conc.	Mobility	Conc.	Mobility
ES	CO	-18.8%		-24.4%		-17.2%		-14.0%		-10.5%	
	PM ₁₀	-21.6%	-52.3%	-23.3%	-56.9%	-28.4%	-54.7%	-28.9%	-33.5%	-26.4%	-35.1%
	O ₃	-9.6%		5.1%		11.3%		27.5%		24.3%	
PR	CO	-41.1%		-45.1%		-29.5%		-30.6%		-27.1%	
	NO ₂	-5.0%	-44.9%	-16.6%	-48.9%	8.1%	-42.5%	7.2%	-37.6%	4.7%	-29.1%
	O ₃	-46.0%		43.9%		74.7%		57.1%		40.1%	
RJ	CO	-16.5%		-22.6%		-12.2%		-14.2%		-12.6%	
	NO ₂	-28.0%		-34.1%		-23.9%		-25.3%		-22.1%	
	PM ₁₀	-19.7%	-51.3%	-23.6%	-54.2%	-11.2%	-53.3%	-10.4%	-42.9%	-9.6%	-32.9%
	PM _{2.5}	-14.7%		8.2%		9.8%		-1.0%		0.6%	
	O ₃	-17.5%		10.7%		19.3%		14.8%		10.7%	
RS	CO	-57.4%		-53.1%		-35.1%		-29.5%		-25.4%	
	NO ₂	-36.1%	-48.8%	-2.4%	-52.0%	-9.0%	-45.6%	21.8%	-41.1%	15.4%	-31.7%
	PM ₁₀	-1.1%		-7.7%		9.1%		-1.3%		-7.0%	
	O ₃	-37.4%		26.3%		18.5%		22.5%		13.1%	
SP	CO	-37.0%		-39.5%		-20.2%		-16.5%		-10.9%	
	NO ₂	-24.2%		-28.8%		-14.5%		-14.9%		-11.4%	
	PM ₁₀	-10.0%	-47.0%	-13.9%	-51.7%	-1.0%	-50.6%	-3.2%	-42.2%	-0.6%	-32.6%
	PM _{2.5}	-12.3%		-20.4%		-4.5%		-10.4%		-7.1%	
	O ₃	-15.9%		15.4%		20.2%		16.5%		22.2%	

Note: The intervals shown correspond to the number of days counted from 16th March 2020, and the corresponding period for 2015-2019: 15 days (16th March to 30th March), 30 days (16th March to 14th April), 90 days (16th March to 13th June), 180 days (16th March to 11th September) and 290 days (16th March to 31st December).

Through Table 5.1, it is observed a sharp reduction in mobility for the first 15 days of mobility restrictions. However, the highest mobility reduction was reached for all the States about 30 days after the first restrictive measure (reduction of approximately 50%); the same behaviour was observed for most of the air pollutants. Looking at the results of restrictions for social distancing applied globally, only in some cases was there a reduction immediately after the measure was enacted, e.g., in India, China, Switzerland, and the Netherlands (VENTER et al., 2020a).

During the first 15 days, the pollutants concentrations in all States decreased, even the secondary pollutants - O₃ and PM_{2.5}. This fact must be associated with the atmospheric conditions during this period that favored the remotion of air pollutants and their precursors (in the case of secondary pollutants) since O₃ precursors (e.g., NO₂), for example, decreased at the same magnitude observed during the first 30 days.

During the first 30 days, the most significant reductions were observed for CO (average reduction of ~ 40%) and NO₂ (~ 20%), pollutants strongly associated with vehicular emissions (BATTERMAN; GANGULY; HARBIN, 2015; FISCHER et al., 2000; GONZÁLEZ et al.,

2017). In this case, attention should be given to the reductions in CO concentrations observed for the States of Rio Grande do Sul, Paraná, and São Paulo, which presented reductions of, respectively, 53% (2015-2019: $208.6 \pm 63.6 \mu\text{g m}^{-3}$; 2020: $114.6 \pm 55.4 \mu\text{g m}^{-3}$), 45% (2015-2019: $890.3 \pm 277.2 \mu\text{g m}^{-3}$; 2020: $417.3 \pm 56.3 \mu\text{g m}^{-3}$), and 39% (2015-2019: $659.7 \pm 45.2 \mu\text{g m}^{-3}$; 2020: $399.1 \pm 130.6 \mu\text{g m}^{-3}$). Additionally, the substantial increase in O₃ concentrations for the same States was also highlighted, which reached 44% (2015-2019: $18.7 \pm 3.4 \mu\text{g m}^{-3}$; 2020: $26.9 \pm 5.5 \mu\text{g m}^{-3}$) in Paraná, 26% (2015-2019: $25.9 \pm 3.4 \mu\text{g m}^{-3}$; 2020: $32.7 \pm 8.1 \mu\text{g m}^{-3}$) in Rio Grande do Sul, and 15% in São Paulo (2015-2019: $38.3 \pm 3.9 \mu\text{g m}^{-3}$; 2020: $44.2 \pm 6.0 \mu\text{g m}^{-3}$).

For more extended periods (from 90 days), less significant reductions are observed for most primary pollutants and analyzed States. On the other hand, an even more notable increase in O₃ concentrations was observed for the Spring season, which, together with Summer, are the seasons that are more favorable conditions to the O₃ formation (SQUIZZATO et al., 2021). A large part of this reduction kept for longer periods is related to the high decreases observed in the initial mobility restrictions since these days are included in the more extended periods. The increase in pollutants is consistent with increased mobility, which although having different characteristics in the States, show a tendency to increase soon after the initial days of restriction (Figure 5.1). During the 90-day period, it is important to mention the increase in PM concentrations, which showed a positive relative difference in the State of Rio Grande do Sul (2015-2019: $20.8 \pm 5.0 \mu\text{g m}^{-3}$; 2020: $22.7 \pm 10.8 \mu\text{g m}^{-3}$). This occurred even with a high rate of reduction in mobility observed for the same period in the State (-45.6%). This can be explained by the high number of fire outbreaks observed for that period, which reached record levels in Brazil (PIVELLO et al., 2021), even during the pandemic. The increase in the number of fire outbreaks was observed throughout the entire national territory (Figure A.4), even in floodable biomes where the impact of fires is generally smaller (PLETSCH et al., 2021). In the specific case of Rio Grande do Sul, from January to May 2020, the State even presented a fourfold increase in the number of fires outbreaks when compared to 2019, surpassing states such as Mato Grosso (increase of 13%) and Mato Grosso do Sul (86%), which are traditionally beset with biomass burning (GZH, 2020). In addition to local fire outbreaks, the region studied is highly impacted by the transport of long-range air masses, which can significantly deteriorate local air quality, even in large urban centers (LOPES et al., 2012; MARTINS et al., 2018; RUDKE et al., 2021a; SANTOS, 2011).

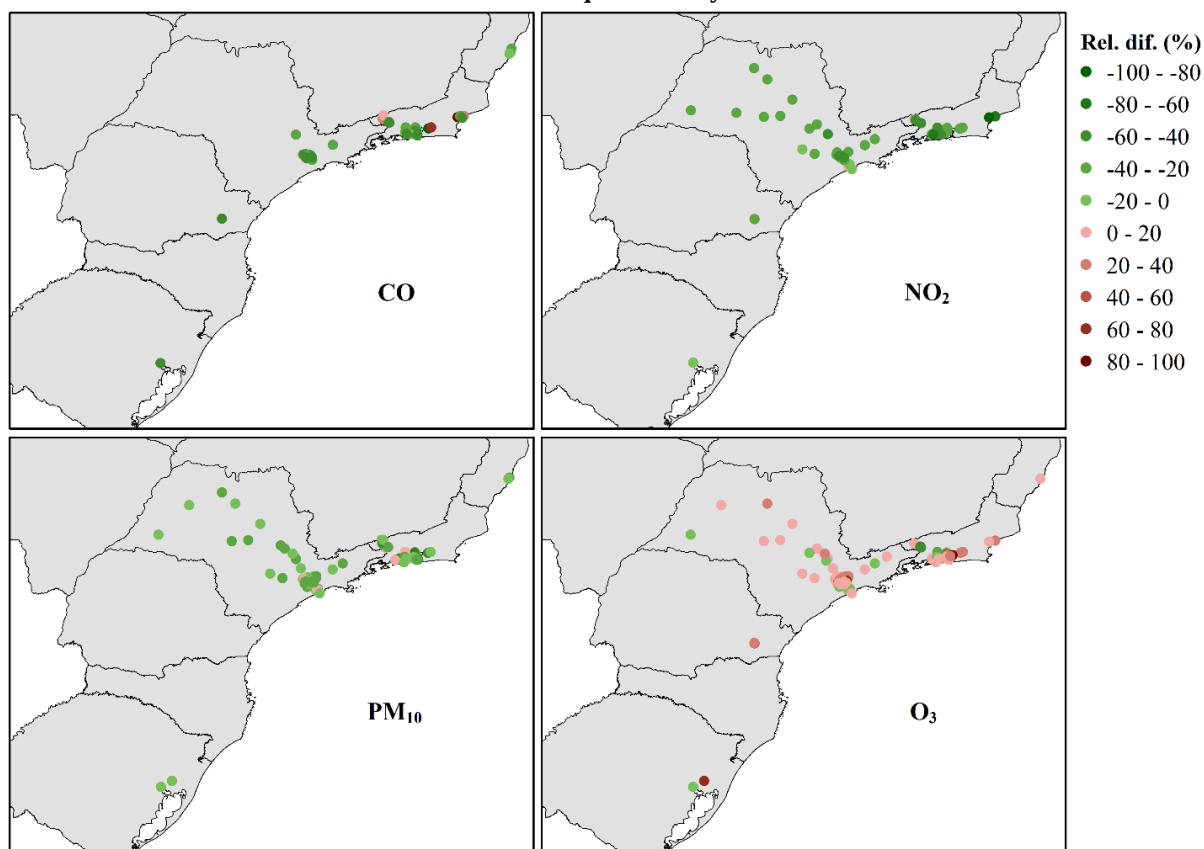
5.3.3 Spatial Variability of Air Quality Changes

As mentioned above, the Brazilian federal government has not adopted a national regulation of social isolation aimed at containing the COVID-19 pandemic. Given the inertia of the Federal government, the Brazilian Supreme Court ruled that, in addition to the federal government, state and municipal governments could also determine isolation, quarantine, and mobility restrictions due to the pandemic (STF, 2020). The first restrictions were adopted by the State of São Paulo and followed by Rio de Janeiro, Rio Grande do Sul, and Paraná states (16th March 2021). On the 21st of the same month, the State of Espírito Santo also decreed the suspension of their activities.

As decision-making was carried out in particular by municipalities (together with the state level, in most States), different types of restrictions were adopted in Brazil. Thus, when studying the average impacts caused by mobility on air pollution by State, there may be some masking of localized effects due to local political and economic characteristics. For instance, increases in air pollutant concentrations in some Chilean cities were detected, even though the general behavior observed for the country was one of reduction (MORALES-SOLÍS et al., 2021). Therefore, increases in air pollutant concentrations in a given city with soft restrictions coupled with reductions seen in other locations can nullify the impact of restrictions at the state level.

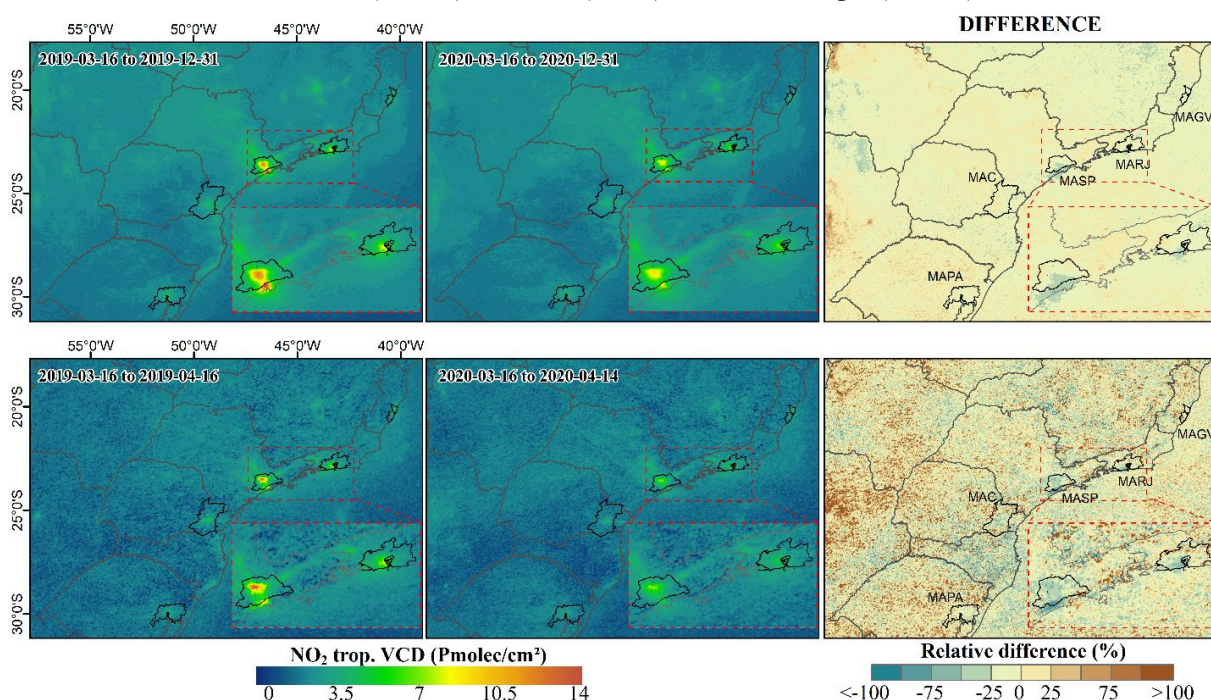
When the impact of mobility restrictions is verified by an air quality station (Figure 5.3), it is noted that the general trend of variation is maintained for the studied pollutants and States, with reductions for the primary pollutants and increases for O₃ (with local variation in intensity). In Paraná, there was a slight increase in NO₂ (2.5%) for a station located in a residential area and a decrease for another station (-26.7%). The reduction in NO₂ occurred because the station is close to a highway (BR 476); thus with the decrease in mobility, there was also a reduction in the pollutant concentration. For the State of Rio de Janeiro, increases in CO concentrations were observed at some monitoring stations, reaching 70.6% (2015-2019: 364.7 $\mu\text{g m}^{-3}$; 2020: 622.1 $\mu\text{g m}^{-3}$) for Itb - Sambaetiba station and 94.4% (2015-2019: 366.4 $\mu\text{g m}^{-3}$; 2020: 712.3 $\mu\text{g m}^{-3}$) for Mc - Fazenda Aires station, which prevented further reductions in pollutant concentrations at the state level. The Itb - Sambaetiba station is located in a sparsely urbanized region, thus suffering less impact from mobility restrictions. Besides, the Mc - Fazenda Aires station is located in a rural area, being more affected by local biomass burning events.

Figure 5.3 - Air pollutants spatial distribution across the study area and relative difference (%) between 16th March with the subsequent 30 days for 2019 and 2020.



Spatially, the greatest reductions in pollutant concentrations were measured in more densely populated areas (Figure 5.1, Figure 5.3, and Figure 5.4). The analyzed States comprise the most urbanized and industrialized region in Brazil (IBGE, 2016), containing important metropolitan areas [e.g., Metropolitan Areas of Greater Vitória (MAGV), São Paulo (MASP), Rio de Janeiro (MARJ), Curitiba (MAC) and Porto Alegre (MAPA)], which showed the greatest reductions observed locally (only data from stations located in the metropolitan area considered). For CO, during the first 30 days of mobility restriction, there was a reduction of 43% (2015-2019: 429.1 $\mu\text{g m}^{-3}$; 2020: 242.8 $\mu\text{g m}^{-3}$) for MAPA, 41% (2015-2019: 102.8 $\mu\text{g m}^{-3}$; 2020: 60.0 $\mu\text{g m}^{-3}$) for MAC, 41% (2015-2019: 279.3 $\mu\text{g m}^{-3}$; 2020: 165.7 $\mu\text{g m}^{-3}$) for MASP, 30% (2015-2019: 217.0 $\mu\text{g m}^{-3}$; 2020: 151.1 $\mu\text{g m}^{-3}$) for MARJ, and 24% (2015-2019: 348.0 $\mu\text{g m}^{-3}$; 2020: 263.3 $\mu\text{g m}^{-3}$) for MAGV.

Figure 5.4 - Mean levels of tropospheric NO₂ density measured by TROPOMI sensor and relative difference (%) for the period from 16th March and the 30 subsequent days for 2019 and 2020. The acronyms presented identify the Metropolitan Areas of Grande Vitória (MAGV), São Paulo (MASP), Rio de Janeiro (MARJ), Curitiba (MAC) and, Porto Alegre (MAPA).



For NO₂, there was a reduction of 33% (2015-2019: 10.5 $\mu\text{g m}^{-3}$; 2020: 7.0 $\mu\text{g m}^{-3}$) for MARJ, 32% (2015-2019: 13.0 $\mu\text{g m}^{-3}$; 2020: 8.8 $\mu\text{g m}^{-3}$) for MASP, 14% (2015-2019: 23.8 $\mu\text{g m}^{-3}$; 2020: 20.4 $\mu\text{g m}^{-3}$) for MAC, and 8% (2015-2019: 4.4 $\mu\text{g m}^{-3}$; 2020: 4.0 $\mu\text{g m}^{-3}$) for MAPA. The expressive NO₂ reductions for MARJ and MASP are also evidenced when evaluating the Tropospheric NO₂ density data measured by the TROPOMI sensor (Figure 5.4).

As CO and NO₂, similar behavior was observed for PM₁₀ that presented reduction of 27% (2015-2019: 25.2 $\mu\text{g m}^{-3}$; 2020: 18.5 $\mu\text{g m}^{-3}$) for MARJ, 23% (2015-2019: 14.1 $\mu\text{g m}^{-3}$; 2020: 10.8 $\mu\text{g m}^{-3}$) for MAGV, 19% (2015-2019: 17.9 $\mu\text{g m}^{-3}$; 2020: 14.5 $\mu\text{g m}^{-3}$) for MASP, and 10% (2015-2019: 21.0 $\mu\text{g m}^{-3}$; 2020: 18.8 $\mu\text{g m}^{-3}$) for MAPA. On the other hand, there were significant increases in the first 30 days of restriction for O₃, which reached 45% in MAC (2015-2019: 18.6 $\mu\text{g m}^{-3}$; 2020: 26.6 $\mu\text{g m}^{-3}$), 36% in MASP (29.6 $\mu\text{g m}^{-3}$; 2020: 35.6 $\mu\text{g m}^{-3}$), 26% in MAPA (25.9 $\mu\text{g m}^{-3}$; 2020: 32.6 $\mu\text{g m}^{-3}$), 11% in MARJ (25.5 $\mu\text{g m}^{-3}$; 2020: 28.2 $\mu\text{g m}^{-3}$), and 5% in MAGV (11.2 $\mu\text{g m}^{-3}$; 2020: 11.7 $\mu\text{g m}^{-3}$).

The results presented here agree with the studies previously carried out for the MASP and MARJ regions (CONNERTON et al., 2020; DANTAS et al., 2020; DEBONE; DA COSTA; MIRAGLIA, 2020; IBARRA-ESPINOSA et al., 2021; NAKADA; URBAN, 2020; NODA et

al., 2021; RUDKE et al., 2021a; SICILIANO et al., 2020a; TADANO et al., 2021), with variations directly related to the study period and how air quality monitoring stations were selected. It should also be noted that even for stations located in the countryside of the States, there is a similar trend in the reduction/increase of air pollutants. This demonstrates the important role of vehicular emission restrictions policies on local/regional air quality, as mobility restrictions carried out mainly in the first 30 days had the potential to affect the average annual concentrations expected for 2020.

5.4 Conclusions

The 2020 COVID-19 outbreak deployed an unique opportunity to assess how the human activities affect the air quality. In this study, we investigated how the restrictive measures imposed in 2020 to reduce the transmission of COVID-19 influenced the air quality of Brazil. Our results demonstrate that, although the federal government had no active participation in developing regulations aimed at social distancing, the Brazilian States used almost the same strategies related to mobility restriction orders. In general, we can affirm that these restrictions adopted by the States had a substantial impact on concentrations of pollutants CO, NO₂ and PM₁₀, and PM_{2.5}, mainly during the first 30 days after the beginning of mobility restriction. The same did not happen with O₃, which reached an increase of 44% in its concentrations at the end of 2020 (Spring and Summer seasons are more favorable to O₃ formation), following a global trend observed in previous studies.

About the spatial analysis, it was possible to demonstrate that in large urban centers, such as the metropolitan areas of the capitals of the five States (MAGV, MASP, MARJ, MAPA, and MAC), they were the ones that showed more effects in the first 30 days of restrictions. A particular emphasis must be given to the MARJ and MASP (largest metropolitan areas in Brazil), which have high vehicle fleet and demonstrated significant reductions in NO₂ concentrations, as observed by the TROPOMI sensor.

Finally, we highlight the importance of mitigating and reducing air pollution indices to reach the new guidelines of the World Health Organization to prevent health and economic impacts. We believe that changes, for example, in the work regime and improvements on public transportation could contribute to it as evidence, especially for primary pollutants. We hope this study will provide subsidies for future policies to control these pollutants, especially when all activities return post COVID-19 pandemic.

It is important to mention that as all studies that require large datasets carried out in Brazil, this one also has limitations. The first limitation is related to the availability of the air quality measurements held in the country; Although some States have measurements for 2020, these datasets are not yet available for public access. In addition to this, a large part of the monitoring stations still relies on manual measurements, and to minimize errors, in this study, we chose to use measures from automatic stations, thus limiting the spatial reach of this study. Besides that, considering the Brazilian territorial area, a small amount of air pollution monitoring stations are available in the country [only 0.03 air quality stations per 1,000 km² (RÉQUIA; KOUTRAKIS; ROIG, 2015)]. Our second limitation is related to a lack of clarity in disseminating information about the restrictions policies carried out by the state health departments, as well as a timetable for their implementation. Most of the data used in the discussions were sourced from media news.

6 HOW MOBILITY RESTRICTIONS POLICY AND ATMOSPHERIC CONDITIONS IMPACTED AIR QUALITY IN THE STATE OF SÃO PAULO DURING THE COVID-19 OUTBREAK²

Abstract: Mobility restrictions are among actions to prevent the spread of the COVID-19 pandemic and have been pointed as reasons for improving air quality, especially in large cities. However, it is crucial to assess the impact of atmospheric conditions on air quality and air pollutant dispersion in the face of the potential variability of all sources. In this study, the impact of mobility restrictions on the air quality was analyzed for the most populous Brazilian State, São Paulo, severely impacted by COVID-19. Ground-based air quality data (PM₁₀, PM_{2.5}, CO, SO₂, NO_x, NO₂, NO, and O₃) were used from 50 automatic air quality monitoring stations to evaluate the changes in concentrations before (January 01 - March 25) and during the partial quarantine (March 16 - June 30). Rainfall, fires, and daily cell phone mobility data were also used as supplementary information to the analyses. The Mann-Whitney U test was used to assess the heterogeneity of the air quality data during and before mobility restrictions. In general, the results demonstrated no substantial improvements in air quality for most of the pollutants when comparing before and during restrictions periods. Besides, when the analyzed period of 2020 is compared with the year 2019, there is no significant air quality improvement in the São Paulo State. However, special attention should be given to the Metropolitan Area of São Paulo (MASP), due to the vast population residing in this area and exposed to air pollution. The region reached an average decrease of 29% in CO, 28% in NO_x, 40% in NO, 19% in SO₂, 15% in PM_{2.5}, and 8% in PM₁₀ concentrations during the mobility restrictions period compared to the same period in 2019. The only pollutant that showed an increase in concentration was ozone, with a 20% increase compared to 2019 during the mobility restrictions period. Before the mobility restrictions period, the region reached an average decrease of 30% in CO, 39% in NO_x, 63% in NO, 12% in SO₂, 23% in PM_{2.5}, 18% in PM₁₀, and 16% in O₃ concentrations when compared to the same period in 2019. On the other hand, Cubatão, a highly industrialized area, showed statistically significant increases above 20% for most monitored pollutants in both periods of 2020 compared to 2019. This study reinforces that the main driving force of pollutant concentration variability is the dynamics of the atmosphere at its various time scales. An

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abnormal rainy season, with above average rainfall before the restrictions and below average after it, generated a scenario in which the probable significant reductions in emissions did not substantially affect the concentration of pollutants.

Keywords: Air Quality; Coronavirus Disease; Air Pollutants; Mobility Restriction.

6.1 Introduction

Air pollution is responsible for more than seven million premature deaths annually worldwide (WHO, 2018). Exposure to air pollutants can lead to health problems, such as cardiovascular diseases (MOKOENA et al., 2019; MOOLGAVKAR, 2000), respiratory diseases (GAUTAM et al., 2016; GUAN et al., 2016; XING et al., 2016), and also mental illnesses and disorders (ALMENDRA et al., 2019; SILVA et al., 2020; SONG et al., 2018). In large urban centers, the major air pollution sources are vehicular emissions and industrial activities (SQUIZZATO et al., 2017; VILAS BOAS et al., 2018; ZHANG; BATTERMAN, 2013). With the Coronavirus Disease 2019 (COVID-19) outbreak, hundreds of articles have sought a nexus between pollution and the pandemic (e.g., AZUMA et al., 2020; CASADO-ARANDA; SÁNCHEZ-FERNÁNDEZ; VIEDMA-DEL-JESÚS, 2020). However, both subjects, pollution and virus, are complexly related to each other through the atmosphere (MARTINS et al., 2020).

In Brazil, São Paulo city and its metropolitan area are known for the pronounced concentrations of air pollutants, mainly due to their numerous vehicular fleets (ANDRADE et al., 2017; KUMAR et al., 2016; LEIRIÃO; MIRAGLIA, 2019). The Metropolitan Area of São Paulo (MASP) comprises 39 cities, including the city of São Paulo, the largest city in South America and its approximately 14 million vehicles have the highest vehicle traffic density of the country, which corresponds to 13% of the Brazilian fleet (DENATRAN, 2020c). According to estimates published in the latest report by the Environmental Company of São Paulo State (in Portuguese Companhia Ambiental do Estado de São Paulo) (CETESB, 2019a), the mobile sources generated about 97% of carbon monoxide (CO), 84% of hydrocarbons (HC), 64% of nitrogen oxides (NO_x), 26% of particulate material (PM) and 17% of sulfur oxides (SO_x) of the total MASP emissions. As a region with such a marked difference between pollutants such as CO and SO_x, it is an important open-air laboratory suitable for assessing how environmental policies impact concentration of pollutants.

Naturally, the emissions in this open-air laboratory, represented by MASP, cannot be controlled for the purpose of study. However, due to the pandemic of COVID-19, there was a sudden forced and marked reduction in productive activities followed by a sharp drop in the mobility of people, as reported in many other major cities in the world (VENTER et al., 2020b; WANG et al., 2020a). Many recent studies carried out across the globe have suggested a significant reduction in air pollutants concentration (BAO; ZHANG, 2020; COLLIVIGNARELLI et al., 2020; COPAT et al., 2020; DUTHEIL; BAKER; NAVEL, 2020; LOKHANDWALA; GAUTAM, 2020; MARQUÈS et al., 2021; SHARMA et al., 2020). In China, the first country affected by the pandemic, an evaluation carried out for 44 cities demonstrated reductions in sulfur dioxide (SO₂) (-6.8%), PM_{2.5} (-5.9%), PM₁₀ (-13.7%), NO₂ (-24.7%), and CO (-4.6%) (BAO; ZHANG, 2020). In India, the study conducted by Sharma et al. (2020) evaluated the concentration for six different pollutants (PM₁₀, PM_{2.5}, CO, NO₂, O₃, and SO₂) in 22 cities, observing significant reductions for PM_{2.5} (-43%), PM₁₀ (-31%), NO₂ (-18%), and CO (-10%). Similar behavior in reducing pollutants due to the pandemic was observed in Europe and the United States, where reductions were observed in the NO_x concentrations (BERMAN; EBISU, 2020; BLUMBERG, 2020; COLLIVIGNARELLI et al., 2020; FILONCHYK; HURYNOVICH; YAN, 2020; SICARD et al., 2020). In Brazil, the partial quarantine, with reductions in road traffic and industrial activities, has also led to a decrease in the levels of pollutants in large cities, mainly for CO and NO_x (CONNERTON et al., 2020; DANTAS et al., 2020; NAKADA; URBAN, 2020; SICILIANO et al., 2020b).

The time-scale variability in pollutant concentrations is linked to the emission sources and the weather conditions that affect the dispersion and the long-range transport from other regions (ANDERSSON; LANGNER; BERGSTROUMM, 2007; HE et al., 2017a; MARTINS et al., 2018). Studies carried out during the pandemic in other regions demonstrate the impact of meteorological variables on air quality, showing that it is impossible to assess changes in pollutant concentrations in a dissociated manner (HÖRMANN et al., 2020; ORDÓÑEZ; GARRIDO-PEREZ; GARCÍA-HERRERA, 2020; PETETIN et al., 2020). Furthermore, in Brazil, an important factor that cannot be neglected are the fires outbreaks, which affect different regions of the country, changing the level of pollutant concentrations, even in large centers (LOPES et al., 2012; MARTINS et al., 2018; PEREIRA et al., 2011; TARGINO et al., 2019). In MASP, favorable conditions for the dispersion of pollutants were found before (February 25 - March 23) and during the partial quarantine (March 24 - April 20), which indicates that the observed reductions in pollutants, during the first weeks of the COVID-19

outbreak, were not determined by changes in the atmospheric conditions (NAKADA; URBAN, 2020). The comparison between the period before and during the partial quarantine demonstrated a decrease in the air pollutant concentrations. In urban centers, substantial emission reductions were observed for NO (-40.4%), CO (-29.8%), NO_x (-25.5%), and NO₂ (-21.5 %) (NAKADA; URBAN, 2020). An exception was observed in the industrial areas, where high air pollutant concentrations occurred even in the quarantine (NAKADA; URBAN, 2020).

Although there have been several studies on the impact of mobility restrictions on the concentration of air pollutants, including those of São Paulo, the analyses were carried out in the wake of the novelty of the pandemic. This means that the studies undergo a lack of a deeper and more detailed analysis that considers temporal and spatial distribution, as well as the atmospheric conditions for the period. Therefore, this work's main objective is to fill this gap and offer an analysis that puts the period under the influence of the pandemic restrictions in a broader context (space and time) and that considers the natural atmospheric dynamics characteristics. To achieve this objective, it was used a dataset from January to June for both years, 2019 and 2020, as well as historical data were used.

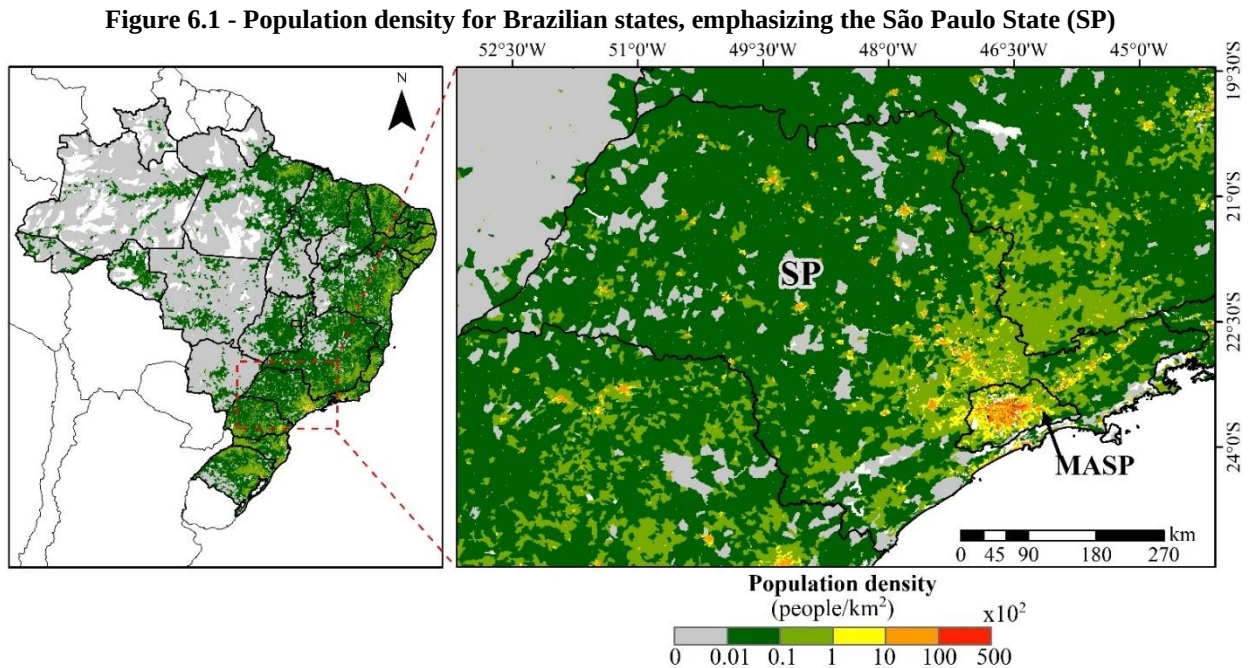
6.2 Methodology

6.2.1 Study area

The study area is the São Paulo State, which is the highest *per capita* GDP and population density in Brazil (IBGE, 2018) (Figure 6.1). São Paulo State has one of the largest urban agglomerations worldwide, the Metropolitan Area of São Paulo (MASP). MASP is home to more than 21 million inhabitants and covers about 8,000 km², 2,000 km² composed of urban areas, making this region the most densely populated in the country (EMPLASA, 2019; IBGE, 2020), shown in Figure 6.1. These characteristics make the area one of the most interesting to assess the impact of social distance measures, especially considering that Brazil is one of the countries most affected by the COVID-19. According to the Köppen climate classification, the State of São Paulo has eight climatic zones, which are divided into two groups, the Humid subtropical zones (67%) and the Tropical zones (33%) (ALVARES et al., 2013). The primary sources of air pollution in São Paulo State are related to its fleet, more than 30.3 million vehicles (DENATRAN, 2020c), and industrial processes. São Paulo State concentrates 36% of Brazilian industrial production, with one of the leading industrial hubs located in Cubatão (composed of

petrochemical/chemical industries, fertilizers, paper, cement, and non-metallic minerals - about 300 sources of pollution) (NAKAZATO; RINALDI; DOMINGOS, 2015; VIEIRA-FILHO; LEHMANN; FORNARO, 2015).

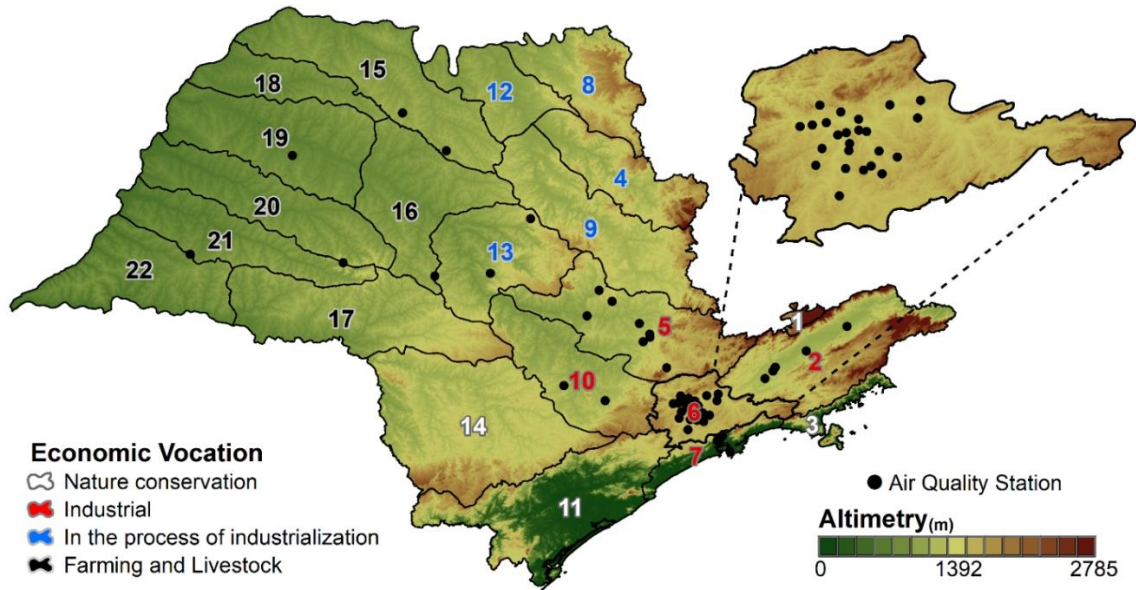
The vast amount of vehicles contributes to air quality problems in this region, making it a local of several studies (ANDRADE et al., 2015, 2017; BRAVO et al., 2016; BRITO et al., 2018; CASTANHO; ARTAXO, 2001; COSTA et al., 2017a; MARTINS et al., 2017a; VILAS BOAS et al., 2018). In the 1930's, the Brazilian government, through a decree, made mandatory the blend of ethanol fuel with gasoline. Also, in the 1970s, the Brazilian government began promoting bioethanol as a fuel, mainly due to the oil crisis. Afterward, this program continued to evolve to use ethanol only in 'flex-fuel' vehicles, which resulted in a significant reduction of some air pollutants (BRITO et al., 2018; SALVO et al., 2017). Programs like PROCONVE (Motor Vehicle Air Pollution Control Program - *Programa de Controle da Poluição do Ar por Veículos Automotores*) and PROMOT (Air Pollution Control Program by Motorcycles and Similar Vehicles - *Programa de Controle da Poluição do Ar por Motociclos e Veículos Similares*) also encourage manufacturers developing cleaner technologies to reduce air pollutants emissions (CETESB, 2018). The creation of these programs with the regulation of stationary sources was essential to reduce some concentrations of pollutants. However, as vehicle fleet increase is a national trend (CETESB, 2019b), air pollution remains an important study object in São Paulo State.



6.2.2 Data Set

The data used in this study are ground-based hourly concentrations of PM₁₀, PM_{2.5}, CO, SO₂, NO_x, NO₂, NO, and O₃ for the historical data series since 1998 (period of the first measurements for some stations) until 2018, for 2019 (year of reference without social distancing restrictions), and for the period from January to June of 2020 (period including days before and during the COVID-19 restrictions in Brazil). The ground-based concentrations were obtained from the CETESB air quality monitoring network (<https://qualar.cetesb.sp.gov.br>). For this study, we considered only automatic air quality monitoring stations. Nowadays, CETESB has 62 automatic stations distributed throughout the São Paulo State, of which 51 contained available data for the analyzed period (Figure 6.2); information about the selected stations is available in Table B.1 (Appendix B). As most stations do not have measures of all pollutants (or present low-quality data), 39 stations for PM₁₀, 19 for PM_{2.5}, 25 for NO_x, 32 for NO₂, 21 for NO, 33 for O₃, 13 for SO₂, and 14 for CO were used. Daily values were calculated based on hourly data for stations with at least 16 hourly data, as recommended by CETESB (2020).

Figure 6.2 - Location of air quality monitoring stations used in this study, São Paulo State. The contours shown are the Hydrographic Units for Water Resources Management – HUWRM. The colors indicated in the numbers refer to the respective economic vocation of each HUWRM



To assess mobility behavior during the period of transition to the pandemic, we used data from the Apple company webpage (<https://covid19.apple.com/mobility>) through a document called "Mobility Trends Reports". The data is obtained from aggregated and information of anonymous smartphone users information during their travels with the navigation function enabled. The reports show the percentage change in visits to places in a geographical area. These places are grouped into three categories: private vehicles, public transport, and walking. The daily percentage changes are calculated based on January 13, 2020, a reference date considered before the mobility effects of the pandemic.

We also evaluated rain and fires in the region to see how much atmospheric conditions may have favored cleaning or accumulating air pollutants. Precipitation values were calculated based on the automatic stations of the National Institute of Meteorology– INMET (<https://portal.inmet.gov.br/>). Data of the total active fires detected by satellites each month were provided by the National Institute for Space Research (INPE). The information can be downloaded in the following online database: http://queimadas.dgi.inpe.br/queimadas/portal-static/estatisticas_estados/. The daily data are available since the year 1998.

6.2.3 Analysis method

The Shapiro-Wilk test was used to assess normality in the data. As air pollutants behave as non-parametric variables, as most environmental variables, the Mann-Whitney U test was used to

determine whether there were differences between the measurements recorded in 2019 and 2020. Two periods were considered for 2019 and 2020: before (75 days from January 1 to March 15) and during (107 days from March 16 to June 30) the mobility restrictions imposed due to the COVID-19 outbreak. Most of the surface data used in this study are concentrated in urban and industrialized areas. The stations located in the countryside are, in general, more recent and do not have the measures of all pollutants. However, even if scarcer, this data's presence was considered relevant for this study (see Table B.1 that provides information on the CETESB air quality monitoring stations). The expansion of the network of stations away from large urban centers is precisely a consequence of some field experiments suggesting the need to monitor pollutants far from the largest urban and industrial areas. Such experiments demonstrated that these regions are not free from urban pollution and can be impacted by the natural biomass burning and the thermoelectric plants of sugarcane bagasse, which are abundant in the countryside of the São Paulo State (KAWASHIMA et al., 2020). These aspects are relevant to have a realistic assessment of how much the mobility restrictions imposed by the COVID-19 impacted the region's air quality. Another aspect of great relevance is the long-range transport of pollutants from fires in central South America and the Amazon region (MARTINS et al., 2018; TARGINO et al., 2019). In this sense, the automatic air quality stations were grouped according to the economic vocation of the regions where they are located, as shown in Figure 6.2. In addition to this grouping, the Hydrographic Units for Water Resources Management (HUWRMs) division was also maintained within each vocational unit. Somehow, HUWRMs are physically separated by their topography (Figure 6.2), a physical barrier that should not be overlooked in air pollution studies.

6.3 Results and discussion

6.3.1 General behavior of ground-based data

13-day centralized moving averages were calculated to evaluate the annual pollutant cycle in all analyses due to the significant variability. In the graphs presented in the sequence of Figure 6.3 to Figure 6.10, the light gray area represents the interval between the minimum and maximum (considering the 13-day centralized moving average) values for the historical series. The dark gray line represents the average value for the historical series, while the blue and red lines represent the average for the year 2019 and the first six months of 2020, respectively. The vertical dashed black line indicates the 76th day of the year 2020 (03/16/2020), when the first

social mobility restrictions began, particularly the suspension of classes at schools and universities (SICILIANO et al., 2020a).

As expected, the highest values in the concentration of pollutants were observed in the region characterized by industrial vocation, where HUWRMs 2, 5, 6, and 7 are located. Discussions will focus on these four regions, which are best represented by the length of the data series and the number of the stations. The results for the other HUWRMs can be found in Table 6.1 and the Figure B.1 to Figure B.6 in the Appendix B. Although these regions have an industrial profile, the values vary both in intensity and in the behavior of the annual cycle.

The topography is an element that isolates these regions, which makes their characteristics the preponderant factor for the concentration of pollutants. The HUWRM 2 is characterized by the Paraíba River valley, which connects the two largest cities in Brazil, São Paulo, and Rio de Janeiro. The region is between two mountain ranges, Serra da Mantiqueira and Serra do Mar. In addition to high population density, it is an important industrial hub. The HUWRMs 5 and 7 are also industrialized, with the former located further inland, on the other side of the Serra da Mantiqueira concerning region 2, and the latter on the coast, between the Atlantic Ocean and Serra do Mar. While the HUWRMs 2 and 5 are more diversified, in HUWRM 7, the petrochemical, steel, chemical, and fertilizer sectors stand out, accounting for most of the national industry's primary products. It is important to note that in the 1970s, means of communication came to consider the city of Cubatão, located in HUWRM 7, as the most polluted city in the world, calling it the "Valley of Death", given the frightening environmental and health impact of resident populations in the area (MONTELEONE-NETO; CASTILLA, 1994). The town became a symbol of the negative consequences of the industrialization patterns in developing countries, which did not consider any pollution control measures (DE MELLO LEMOS, 1998). In HUWRM 6, characterized by the MASP, most air quality stations capture vehicle traffic information, although it is highly industrialized in its surroundings. This region is also located between the Serra da Mantiqueira and Serra do Mar Mountains, but it is not part of the Paraíba Valley, being at a slightly higher altitude and with the somewhat lower adjacent mountain ranges. The individual behaviors of pollutants PM_{10} , $PM_{2.5}$, CO, SO₂, NO_x, NO₂, NO, and O₃ will be discussed below.

The information contained in Table 6.1 is strategic for understanding the impact of pandemic restrictions and assessing what is observed in Figure 6.3 to Figure 6.10. Table 6.1 presents the general behavior of the comparison between average pollutant concentrations for 2019 and 2020, considering the periods before and during the pandemic restrictions in 2020. It indicates (symbols) whether, in 2020, there was an

increase or decrease compared to 2019 for the periods before (January 1 to March 15) and during (March 16 to June 30). The specific results of the Mann-Whitney U test can be seen in Table B.2 – Mann-Whitney U test results compare average daily concentrations between the years 2019 and 2020, considering the period before (January 1 to March 15) and during (March 16 to June 30) the COVID-19 outbreak.

Table 6.1 – Mann-Whitney U test results compare average daily concentrations between the years 2019 and 2020, considering the period before (January 1 to March 15) and during (March 16 to June 30) the COVID-19 outbreak

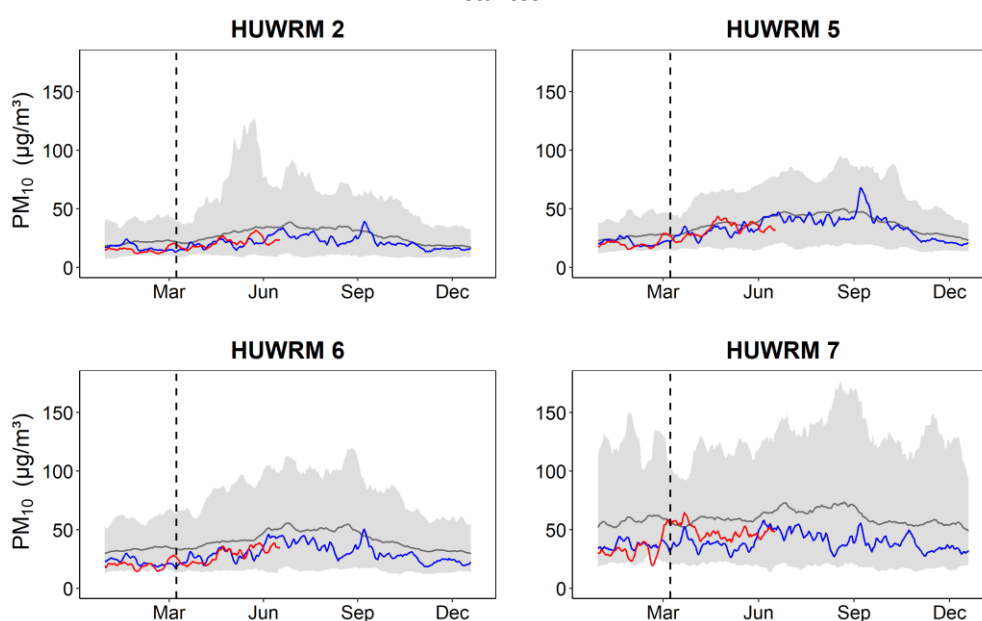
HUWRM	Station name	PM ₁₀		PM _{2.5}		NO _x		NO ₂		NO		SO ₂		CO		O ₃	
		Bef.	Dur.	Bef.	Dur.	Bef.	Dur.	Bef.	Dur.	Bef.	Dur.	Bef.	Dur.	Bef.	Dur.	Bef.	Dur.
2	Guaratinguetá	↓	=	NA	NA	↓	↓	↓	↓	↓	↓	NA	NA	NA	NA	=	=
	Jacareí	=	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	↓	↓
	S.José Campos	↓	=	NA	NA	↓	↓	↓	↓	=	↓	=	=	NA	NA	↓	=
	S.José Campos-Jd.Satelite Taubaté	=	=	↓	=	-	↓	↓	↓	-	=	NA	NA	↑	↑	=	↑
5	Campinas-Centro	=	↓	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	↓	↓	NA	NA
	Campinas-Taquaral	NA	NA	NA	NA	↓	↓	↓	↓	NA	NA	NA	NA	NA	NA	=	=
	Campinas-V.União	NA	NA	=	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	=	↓
	Jundiaí	↓	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	=	↑
	Limeira	↑	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Paulínia	↓	=	NA	NA	NA	NA	NA	NA	NA	NA	↓	-	NA	NA	=	=
	Piracicaba	↑	↑	=	=	-	↓	↓	=	=	↓	NA	NA	NA	NA	=	=
	Santa Gertrudes	=	↓	=	↓	↑	↑	=	=	↑	↑	NA	NA	NA	NA	NA	NA
6	Capão Redondo	-	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	↓	↑
	Carapicuíba	=	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	↓	↓	=	↑
	Cerqueira César	↓	=	NA	NA	↓	↓	↓	↓	NA	NA	-	=	↓	↓	NA	NA
	Congonhas	=	=	=	↓	=	↓	=	↓	NA	NA	=	=	↓	↓	NA	NA
	Diadema	↓	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	↓	↑
	Grajaú-Parelheiros	=	=	↓	↓	NA	NA	NA	NA	NA	NA	NA	NA	↓	↓	NA	NA
	Guarulhos-Paço Municipal	=	=	=	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	=	↑
	Guarulhos-Pimentas	NA	NA	NA	NA	↓	↓	↓	↓	↓	↓	↓	=	↓	↓	=	↑
	Ibirapuera	NA	NA	↓	↓	↓	↓	↓	↓	NA	NA	NA	NA	↓	↓	=	↑
	Interlagos	=	=	NA	NA	NA	NA	NA	NA	NA	NA	-	↓	NA	NA	-	↑
	Itaim Paulista	NA	NA	↓	=	↓	↓	↓	↓	=	↓	NA	NA	NA	NA	=	=
	Marg.Tietê-Pte Remédios	↓	↓	↓	=	↓	↓	↓	↓	↓	↓	=	=	↓	↓	NA	NA
	Mooca	NA	NA	=	↓	NA	NA	NA	NA	NA	NA	NA	NA	↓	↓	NA	NA
	N.Senhora do Ó	=	=	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Osasco	=	=	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	NA	NA
	Parque D.Pedro II	NA	NA	-	=	↓	↓	↓	↓	↓	↓	NA	NA	↓	↓	=	↑
	Pico do Jaraguá	NA	NA	=	↓	NA	NA	=	↓	NA	NA	NA	NA	NA	NA	NA	NA
	Pinheiros	NA	NA	NA	NA	↓	↓	↓	↓	↓	↓	NA	NA	↓	↓	=	↑
	S.André-Capuava	↓	↓	NA	NA	NA	NA	NA	NA	NA	NA	=	=	NA	NA	↓	↑
	S.Bernardo-Centro	NA	NA	=	=	-	↓	↓	↓	-	↓	NA	NA	↓	↓	-	↑
	S.Bernardo-Paulicéia	=	↓	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
	Santana	NA	NA	↓	↓	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	-	↑
	São Caetano do Sul	=	=	-	=	-	↓	↓	↓	-	↓	-	=	NA	NA	-	↑
	Taboão da Serra	=	↓	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
7	Cubatão-Centro	↓	=	NA	NA	=	↓	↓	↓	NA	NA	↓	=	NA	NA	NA	NA
	Cubatão-V.Parisi	=	↑	NA	NA	=	↑	=	↑	↑	↑	=	=	NA	NA	NA	NA
	Santos-Ponta da Praia	=	↑	↓	↓	NA	NA	=	=	NA	NA	↓	↓	NA	NA	=	↑
10	Sorocaba	-	=	NA	NA	NA	NA	-	↓	↑	↓	NA	NA	NA	NA	-	↑
	Tatuí	=	=	NA	NA	NA	NA	=	↓	=	↓	NA	NA	NA	NA	↓	=
13	Araraquara	=	↑	NA	NA	NA	NA	↓	=	NA	NA	NA	NA	NA	NA	NA	NA
	Bauru	=	=	NA	NA	↓	↓	↓	↓	NA	NA	NA	NA	NA	NA	=	↑
	Jaú	↓	=	NA	NA	↓	↓	=	↓	↓	↓	NA	NA	NA	NA	=	↑
15	Catanduva	=	=	NA	NA	=	↓	=	↓	NA	NA	NA	NA	NA	NA	↓	↑
	São José do Rio Preto	=	=	=	=	=	=	=	=	=	=	NA	NA	NA	NA	NA	NA
19	Araçatuba	=	↑	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	↓	↑
21	Marília	=	=	NA	NA	↓	↓	↓	↓	=	↓	NA	NA	NA	NA	=	↑
22	Presidente Prudente	↓	=	NA	NA	=	↓	=	↓	↓	↓	NA	NA	NA	NA	↓	=

Legend: Bef. Before the period of the pandemic restrictions; Dur. During the period of the pandemic restriction; ↓ Statistically different and lower concentration at a monitoring station in 2020 concerning 2019; ↑ Statistically different and higher concentration at a monitoring station in 2020 concerning 2019; = Monitoring station without statistically significant differences in the concentration for 2019 and 2020; NA, not available data.

5.3.1.1 PM₁₀

PM₁₀ is the pollutant with the largest number of measurement stations in the State of São Paulo. Thus, it can provide a more complete spatialized view of the behavior of pollution in the study period, including information outside the industrialized region. The average annual concentrations of historical series for PM₁₀ in HUWRMs 2, 5, 6, and 7, were 25.7 [15.5 - 31.9], 34.8 [23.1 - 43.3], 38.8 [25.0 - 48.2] and 58.3 [36.6 - 74.1] $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The bracketed intervals represent the interquartile range. Notably, HUWRM 7 has a history of the highest average concentration of this pollutant and does not show seasonality in concentrations (Figure 6.3). It is precisely the region highly impacted by industry, especially petrochemicals, as described above. For the other regions, the annual cycle is very well defined, with the minimum values being recorded in the summer months (rainy season) and maximum values in the winter months (drier), as already observed in previous studies (e.g., MARTINS et al. (2017a)). The high recurrence of thermal inversion events in São Paulo city in the winter months plays an important role in episodes of high particulate matter concentration (FREITAS et al., 2007). Besides, the resuspension of soil dust is responsible for most of the coarse particle mass (ANDRADE et al., 2017; CASTANHO; ARTAXO, 2001). This characteristic is also observed for HUWRM 7, but in this case, the fertilizer factories and the soil resuspension represent about 70% of the coarse particle mass (KERR et al., 2001).

Figure 6.3 – PM₁₀ concentration ($\mu\text{g}\cdot\text{m}^{-3}$) for HUWRMs in the State of São Paulo. The light gray area represents the interval between the minimum and maximum. The dark gray line represents the average for the historical series; blue and red lines represent the average for the years 2019 and 2020, respectively. The vertical dashed black line indicates the 76th day of the year 2020 when the first restrictions were started



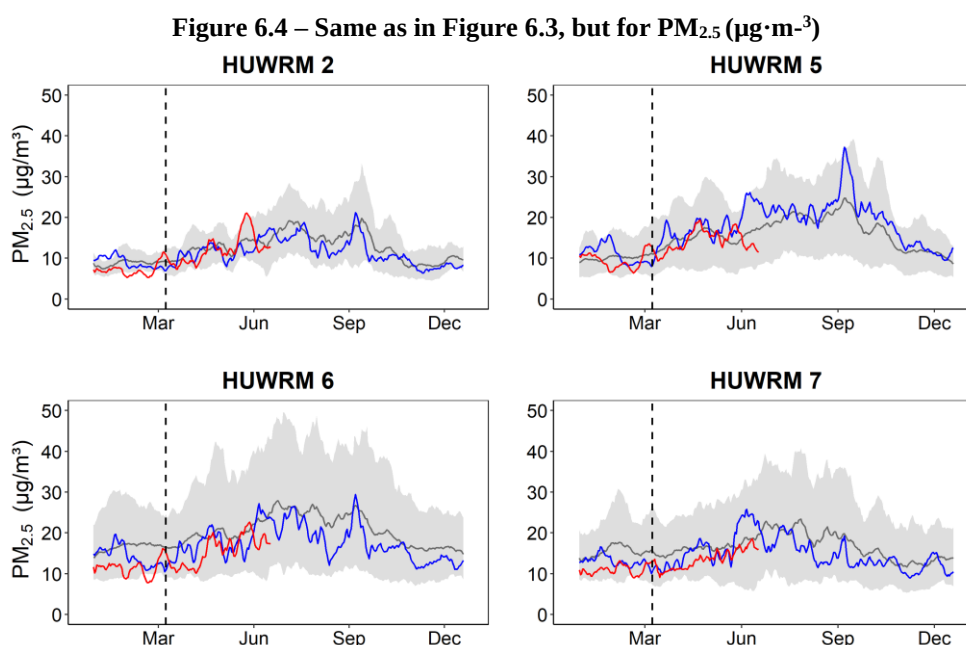
Concerning the first six months of the year 2019 and 2020, it was observed that the PM₁₀ concentrations are significantly below the average values of the historical series. According to recent studies, all regions analyzed in this study have shown reduction trends for most pollutants in 2020, mainly due to the adoption of more strict policies to control emissions (ANDRADE et al., 2017; CARVALHO et al., 2015). No significant differences (p -value > 0.05) were observed between the years 2019 and 2020 for the first period (Bef.) for most of the stations (Table 6.1). However, the year 2020 presents slightly lower concentrations (Figure 6.3). Average concentrations in 2019 were 16.9, 21.2, 22.6, and 37.0 $\mu\text{g}\cdot\text{m}^{-3}$ for HUWRMs 2, 5, 6, and 7, respectively, and 15.1, 20.4, 20.1, and 33.8 $\mu\text{g}\cdot\text{m}^{-3}$ for the same regions in 2020. In the period following the beginning of restrictions due to the pandemic (Dur.), there is no significant difference for most monitoring stations (Table 6.1). The average values in 2019 were 21.6, 32.9, 30.9, and 40.4 $\mu\text{g}\cdot\text{m}^{-3}$ for HUWRMs 2, 5, 6, and 7, respectively, and 21.4, 32.0, 28.7, and 47.8 $\mu\text{g}\cdot\text{m}^{-3}$ for the same regions in 2020. An exception to this behavior was HUWRM 7, where the difference was statistically significant with higher average concentrations in 2020 (Dur. period), as already mentioned in the study of Nakada and Urban (2020). As the concentrations increase from autumn to winter, following the seasonality, the period before the restrictions do not seem reasonable for comparison.

5.3.1.2 PM_{2.5}

The annual average and the interquartile range (in brackets) concentrations for historical series of PM_{2.5} in HUWRMs 2, 5, 6 and 7 were 11.7 [7.3 - 14.9], 15.0 [9.2 - 19.4], 19.1 [12.2 - 23.9], and 15.8 [10.1 - 20.3] $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The HUWRM 6 (MASP) stands out with the highest average concentrations, possibly due to the numerous vehicle fleet (largest in the country), which is responsible for most of the concentration of this pollutant (ANDRADE et al., 2017). In sequence, HUWRM 7 was the one with the highest average concentration. It is worth mentioning that, unlike PM₁₀, there is no PM_{2.5} measurement in Cubatão; the city of Santos being the only one that contains a monitoring station for this pollutant in region 7. The Santos monitoring station is located close to the Port area, one of largest port complexes in the Latin America. Thus, the main sources of pollutant must be linked to the activities at port (mainly those related to circulation of heavy vehicles) and emissions by ships in the port (SALANI, 2018).

The annual cycle of PM_{2.5} has the same behavior as PM₁₀ since both are influenced by precipitation (Figure 6.4); however, it does not present the reduction trend as seen for the PM₁₀

since the measurements of this pollutant started in 2010 (Table B.1) at Congonhas station (MASP) and it has an expressive contribution of secondary sources (ANDRADE et al., 2012, 2017; DE A. ALBUQUERQUE et al., 2019). In 2019, a peak with high concentrations of $PM_{2.5}$ was observed between August and September, particularly between 09/16 and 09/22, for most HUWRMs. In general, this period is characterized by an intense number of fire outbreaks. A consultation carried out with the Burning Program from the National Institute for Space Research (INPE) [<http://queimadas.dgi.inpe.br/queimadas/bdqueimadas/>] confirms that the peak in the number of fire outbreaks coincides with the concentration of $PM_{2.5}$. These peaks were more evident in HUWRMs localized in the São Paulo State interior, where the daily averages of $PM_{2.5}$ reached 73.5 (HUWRM 15) and 50.9 (HUWRM 5) $\mu g \cdot m^{-3}$.



There was a statistically significant difference (p -value < 0.05) between concentrations in the period before 2019 and 2020. In at least half of the stations assessed, concentrations in 2020 were considerably lower than in 2019. For this period in 2019, average concentrations of 9.0, 10.9, 14.3, and 13.0 $\mu g \cdot m^{-3}$ were observed for the HUWRMs 2, 5, 6, and 7, respectively, and 7.2, 9.7, 11.5, and 11.0 $\mu g \cdot m^{-3}$ for the same regions in 2020. During the restrictions (the period during), there are statistically significant differences (p -value < 0.05), especially for HUWRM 6 (MASP). For 2019 the average values found were 12.0, 17.9, 18.1, and 16.5 $\mu g \cdot m^{-3}$ for the HUWRMs 2, 5, 6, and 7, respectively, and 12.0, 13.9, 15.6, and 13.6 $\mu g \cdot m^{-3}$ for the same regions in 2020. It is observed that this evident reduction in the concentration of $PM_{2.5}$ for 2020, which was not observed for PM_{10} , is consistent with the observations of Sharma et al. (2020),

which attributes this behavior to a more significant contribution of anthropogenic sources to the concentrations of PM_{2.5}, mainly industrial and vehicular emissions (burning of fuels) (ALVES et al., 2015; PEREIRA et al., 2017a).

In this context, it is important to mention that until November of 2018, Brazil had no legislation establishing maximum limits for PM_{2.5} (currently legislated by CONAMA n° 491/2018) (CONAMA, 2018). Thus, there are few air quality stations in the country capable of carrying out PM_{2.5} measurements. Besides, when available, these stations bring only recent measures, which prevents an adequate assessment of the behavior of pollutant during the pandemic period concerning previous years.

Another relevant aspect for the context of this study is the use of in natura biomass as a fuel. According to some studies, the domestic use of wood for cooking or heating is usually reported as an important source of pollutants. Unlike other countries, especially in the northern hemisphere, the use of heating is almost non-existent in Brazil, mainly because there are only a few days in the year when the temperature gets closer to 0 °C (ALVARES et al., 2013). On the other hand, the use of firewood and charcoal for pizzerias, restaurants, and steakhouses is considerable in São Paulo (KUMAR et al., 2016; LIMA et al., 2020; VIEIRA-FILHO; PEDROTTI; FORNARO, 2013). Vieira-Filho et al. (2013) indicate that approximately 7.5 ha of eucalyptus forest in the State of São Paulo are used as firewood per month by pizzerias and steakhouses. Besides, Kumar et al. (2016) suggest that 3% of atmospheric emissions at MASP are related to fuelwood burning by pizzerias. Another important factor is associated with land use and land cover in the region and the management of cultivated areas, especially in the interior of the São Paulo State (RUDKE et al., 2021b, 2019). Mostly in sugar cane plantations, the pre-harvest burning practiced has demonstrated a significant impact in increasing concentrations of pollutants associated with fires (e.g., smoke, inhalable particulate matter, and total suspended particulates) (EL CHAMI; DACCACHE; EL MOUJABBER, 2020; TSAO et al., 2012; URIARTE et al., 2009). Over the years, there has been a reduction in this agricultural practice, mainly due to sanctions from regional governments seeking to gradually encourage farmers to reduce fires (TSAO et al., 2012). Even so, more than half of the sugar cane cultivation areas continue to be burned in São Paulo State (RUDORFF et al., 2010).

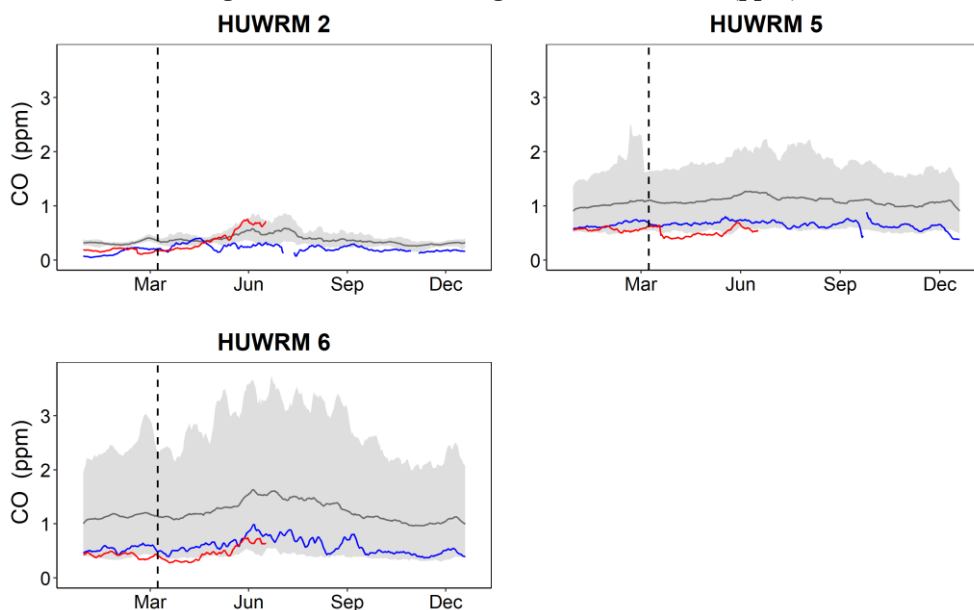
5.3.1.3 CO

CO is the most restrictive pollutant for a spatialized approach to behavior during the pandemic since from 14 stations that measure CO, most of them (12) is located in the MASP (HUWRM 6) and only two outside. The annual average and the interquartile range of CO concentrations in HUWRMs 2, 5 and 6 were 0.34 [0.22 - 1.3], 1.0 [0.7 - 5.6], and 1.2 [0.7 - 5.2] ppm, respectively. The highest concentrations are observed in HUWRM 6, in which mobile sources are responsible for 97% of their emission (CETESB, 2019a). According to CETESB, light vehicles are the main source of carbon monoxide emissions (63%) in the region. For the other HUWRMs, there is also a greater contribution from mobile sources (93% for HUWRM 5 and 72% for HUWRM 2) (CETESB, 2019a).

Figure 6.5 shows that the CO concentrations for the years 2019 and 2020 are below the average concentrations in the historical series, as observed for HUWRMs 5 and 6, which have long-time series. This behavior is directly related to reducing emissions from new light vehicles, in compliance with the PROCONVE and PROMOT phases (ANDRADE et al., 2017). However, this trend could not be seen in recent data as in HUWRM 2, where the measurements started in 2015. The efficiency of vehicular emissions control policies seems to have reached its maximum.

In all regions, statistically significant differences were observed between the average values for the period before 2019 and 2020 (p -value <0.05). 2019 observed average concentrations were 0.15, 0.65, and 0.54 ppm for HUWRMs 2, 5, and 6, respectively, and 0.17, 0.56, and 0.42 ppm for the same regions in 2020, and them with higher concentrations in 2019 at 5 and 6. HUWRM 5 was the one with the highest average concentrations of CO, which can be attributed to the fact that the measurements come from a single monitoring station that is located in the central region of Campinas, the largest city in the State of SP outside MASP, with more than one million inhabitants. Thus, although some stations located in a high vehicle traffic area at HUWRM 6 have higher concentrations than the measurements in Campinas (D'AMELIO; CAMPOS; ALVIM, 2017), the average for HUWRM 6 is lower than the average for HUWRM 5.

Figure 6.5 – Same as in Figure 6.3, but for CO (ppm)



Statistically significant differences (p -value <0.05) were also observed during the period with mobility restrictions. HUWRM 2 showed an increase of 4.1% compared to the average concentrations of previous years for the same pandemic period. HUWRMs 5 and 6 showed reductions of 27.0% and 25.3%, respectively. The CO measurements of HUWRM 2 come from a single station and may be representing the particular behavior of a specific location, which for some reason, increased during the period of mobility restrictions.

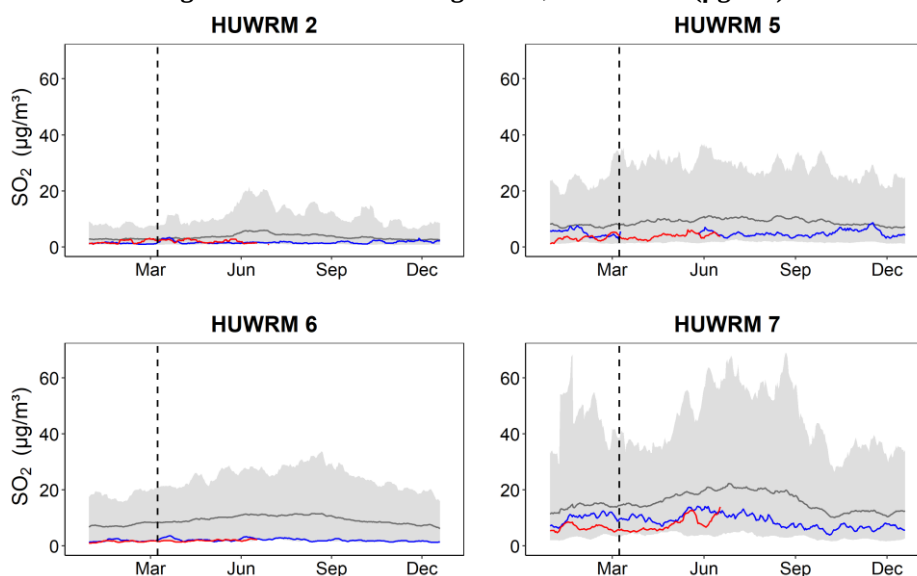
5.3.1.4 SO₂

The annual average and the interquartile range of SO₂ concentrations in HUWRMs 2, 5, 6 and 7 were 3.4 [1.4 - 4.2], 8.5 [3.4 - 10.8], 8.6 [3.3 - 12.5], and 15.1 [7.6 - 19.1] $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The highest average concentrations were observed for HUWRM 7, mainly due to the industrial complex in the Cubatão region and the maritime transport in the port of Santos, which is associated with the use of fuel with a high sulfur content (LEDOUX et al., 2018; YU-PENG; LIN; JONG, 2005).

The SO₂ concentrations in all HUWRMs are significantly lower than the historical average concentrations recorded (Figure 6.6) considering the study periods. This behavior is due to the regulation of emissions, which has promoted a gradual reduction in fuel sulfur content in recent decades (both industrial and automotive (ANDRADE et al., 2017; CETESB, 2019a). Most stations do not indicate a statistically significant difference between 2019 and 2020 for the first period (p -value <0.05), except for the stations located in HUWRM 7. Average concentrations

in 2019 of 1.3 , 5.2 , 1.8 , and $9.8 \mu\text{g}\cdot\text{m}^{-3}$ were observed for HUWRMs 2, 5, 6, and 7, respectively, and 1.9 , 3.2 , 1.5 , and $6.5 \mu\text{g}\cdot\text{m}^{-3}$ for the same regions in 2020. If we consider only the port station measurements in HUWRM 7, there is an 80% reduction in SO_2 concentrations for 2020 compared to 2019 (from $13.9 \mu\text{g}\cdot\text{m}^{-3}$ in 2019 to $2.7 \mu\text{g}\cdot\text{m}^{-3}$ in 2020). Therefore, the decrease observed from 2019 to 2020 at HUWRM 7 is essentially due to the considerable reduction at the station located in port of Santos. The decline in SO_2 concentrations in port of Santos port is most likely related to the International Maritime Organization (IMO) regulations applied to marine fuels, which became effective in January 2020. Based on this regulation, the maximum SO_2 in the fuel content used onboard ships went from 3.50% to 0.50% (IMO, 2019).

Figure 6.6 – Same as in Figure 6.3, but for SO_2 ($\mu\text{g}\cdot\text{m}^{-3}$)



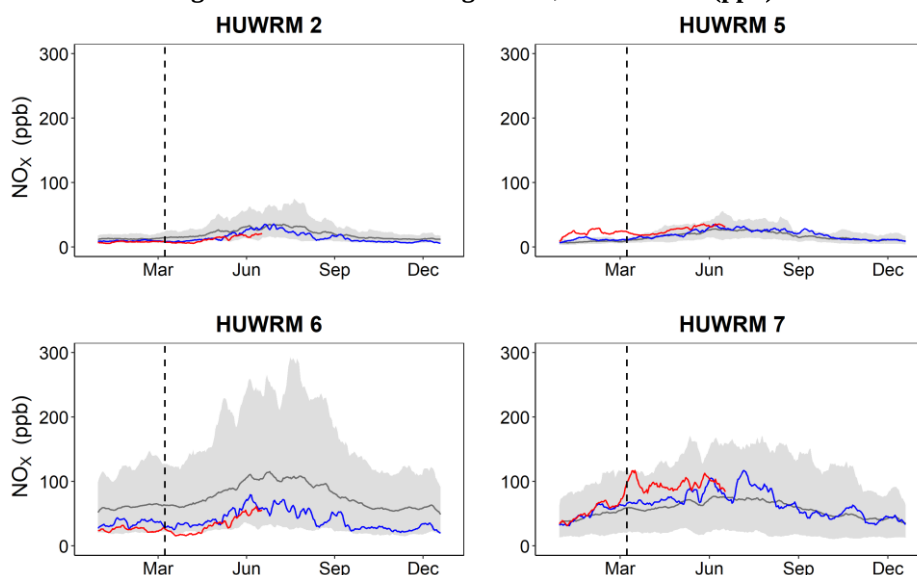
For the period with mobility restrictions, the significant differences remained for HUWRM 7 and not significant for the other regions (HUWRM 5 did not present data for this period in 2019). The average concentrations found in 2019 were 1.7 , 2.4 , and $10.8 \mu\text{g}\cdot\text{m}^{-3}$ for HUWRMs 2, 6, and 7, respectively, and 2.0 , 1.9 , and $8.1 \mu\text{g}\cdot\text{m}^{-3}$ in the same regions in 2020. Therefore, the results suggest no evident reduction in SO_2 concentrations due to restrictions in the pandemic period.

5.3.1.5 NO_x

The annual average and the interquartile range of NO_x concentrations observed for HUWRMs 2, 5, 6, and 7 were 17.7 [$9.4 - 21.2$], 16.6 [$8.6 - 21.7$], 71.6 [$40.6 - 90.6$], and 56.7 [$33.4 - 73.4$] $\mu\text{g}\cdot\text{m}^{-3}$, respectively. Figure 6.7 shows the annual cycle of average NO_x concentrations, and the

highest concentrations are observed in HUWRM 6 that are mainly related to vehicle emissions since this sector contributes to 67% of NO_x emissions (CETESB, 2019a), mostly by heavy vehicles. Estimates indicate that heavy vehicles (diesel) are responsible for about three-quarters of this pollutant's vehicle emissions (CETESB, 2019b, 2019a).

Figure 6.7 – Same as in Figure 6.3, but for NO_x (ppb)



As observed for other pollutants, in the figures for 2019 and 2020, the concentrations are substantially below the historical average for HUWRM 6, a region with long data series. This reduction is related to the PROCONVE phases, which significantly restricted vehicles' NO_x emission limits (ANDRADE et al., 2017). As for the first period of 2019 and 2020, concentrations were remarkably close for HUWRMs 2 and 7, higher values for 2020 in HUWRM 5, and lower values for 2020 in HUWRM 6. The significant increase for the first period of 2020 compared to the same period in 2019 was observed only for the Santa Gertrudes station at HUWRM 5, located in an industrial hub of the ceramic sector. It should be noted that the last months of 2019 were favorable for the ceramic industry since the civil construction sector reached the highest level of economic activity in comparison to previous years (CNI, 2019). In the first months of 2020, the civil construction sector performance maintained the high growth standards observed for the end of 2019.

During the restricted period, significant reductions in NO_x concentrations were observed for most stations. On the other hand, increases in concentrations were observed, only in Santa Gertrude (HUWRM 5) and Cubatão-Vila Parisi (HUWRM 7) stations. The concentrations were

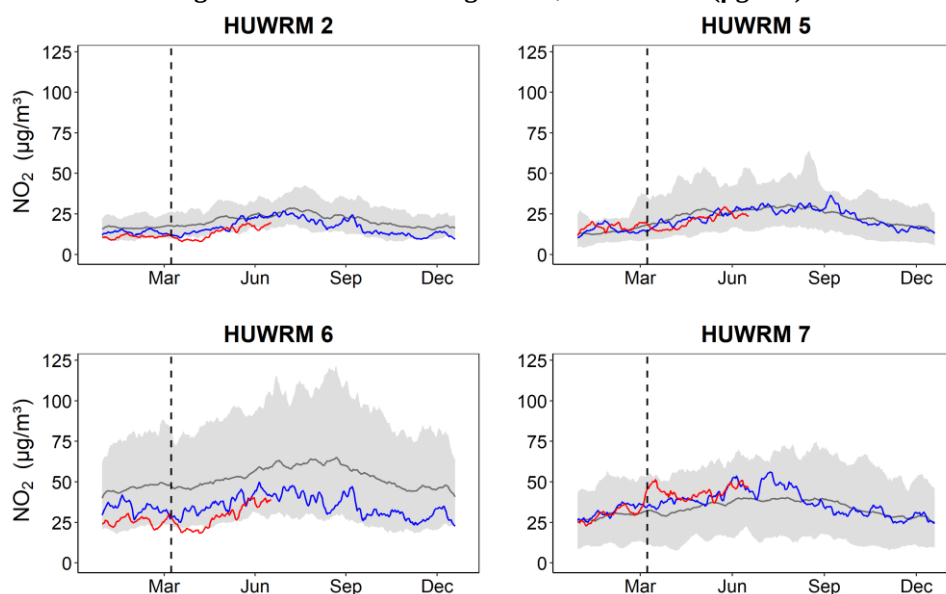
sufficiently high to change the average value trend in these regions. The 2019 average concentrations were 16.6, 21.1, 45.0, and 75.8 $\mu\text{g}\cdot\text{m}^{-3}$ for HUWRMs 2, 5, 6, and 7, respectively, and 12.6, 26.6, 34.0, and 95.5 $\mu\text{g}\cdot\text{m}^{-3}$ for the same regions in 2020. According to Krecl et al. (2020), reductions in NO_x concentrations have been observed over the last few years in the MASP due to the limits adopted for vehicle emissions (HUWRM 6). However, the 2020 decrease may be related to the mobility restrictions imposed by COVID-19 since the reductions observed for the period are greater than those expected due to the implementation of a control policy. In this case, a decrease in the traffic was evident after the restrictions started due to the pandemic.

It should also be noted here that some international factors, but mainly a favorable condition for the appreciation of soy exports, may have stimulated the demand for fertilizers, whose sales peak in April and May (IEA, 2015). In this case, orders for the fertilizer industry will likely be placed sometime earlier, thus coinciding with the beginning of the restrictions due to the pandemic and directly impacting the NO_x emissions in the Vila Parisi station, next to the petrochemical and fertilizer production. It is worth remembering that 100% of the ammonium nitrate consumed in Brazil is produced in this industrial hub.

For NO_2 the annual average and their interquartile range concentrations in HUWRMs 2, 5, 6, and 7 were 19.2 [13.4 - 23.9], 21.6 [14.4 - 26.8], 50.1 [36.3 - 61.4], and 33.4 [24.3 - 41.1] $\mu\text{g}\cdot\text{m}^{-3}$, respectively. HUWRM 6 has the highest average concentrations recorded for the historic period. Like CO, NO_2 is considered an important traffic marker (COSTA et al., 2017b).

Except for HUWRM 7, in other HUWRMs, it is observed that the concentrations for 2019 and 2020 are below the average values of the historical series, demonstrating a reduction in NO_2 . There was a statistically significant difference for the first period of 2019 and 2020, especially in HUWRMs 2 and 6 (Figure 6.8 and Table 6.1). In the period following the beginning of the restrictions, a significant difference was observed in almost all regions. In 2019, the averages found were 17.8, 22.9, 36.8, and 40.9 $\mu\text{g}\cdot\text{m}^{-3}$ for HUWRMs 2, 5, 6, and 7, respectively, and 14.0, 20.7, 29.1, and 44.0 $\mu\text{g}\cdot\text{m}^{-3}$ for the same regions in 2020. An increase was observed for the Cubatão region, mainly due to high concentrations measured at the Cubatão V.Parisi station. Even during the period with mobility restrictions, the Cubatão station registered a daily average of 85.6 $\mu\text{g}\cdot\text{m}^{-3}$ (March 19, with a maximum hourly average of 124 $\mu\text{g}\cdot\text{m}^{-3}$), probably due to its intense industrial activity, as discussed previously for the NO_x behavior.

Figure 6.8 – Same as in Figure 6.3, but for NO₂ (µg·m⁻³)

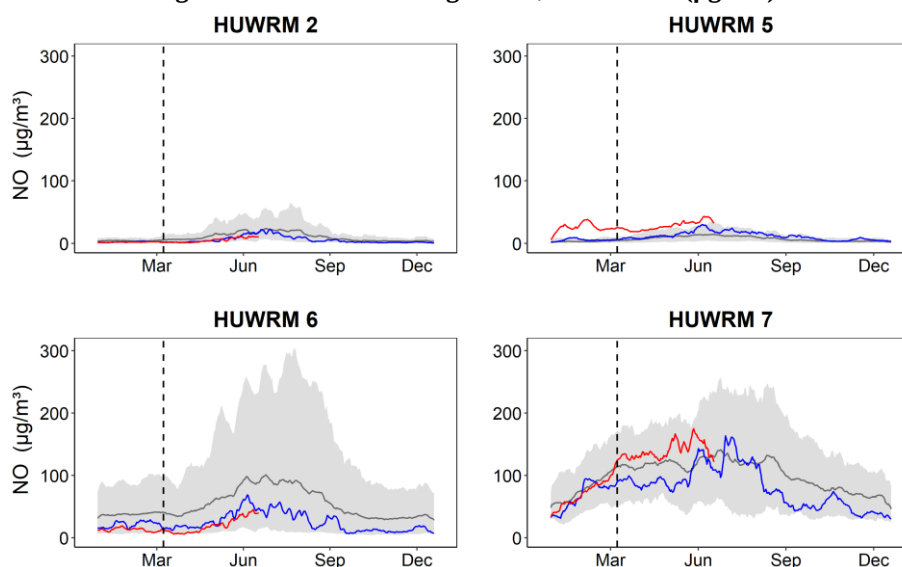


For NO the annual average and their interquartile range concentrations in HUWRMs 2, 5, 6 and 7, were 8.4 [1.9 - 9.5], 7.9 [2.3 - 10.3], 49.9 [19.2 - 63.6], and 96.7 [59.0 - 126.4] µg·m⁻³, respectively. The highest concentrations in the historical series were observed for HUWRM7, specifically at Cubatão V.Parisi station, and for HUWRM 6, as observed for NO_x and NO₂.

In the case of NO concentrations, most stations showed significant reductions in the first period of 2020 compared to the previous year (

Figure 6.9 and Table 6.1). On the other hand, significant increases in NO concentrations were observed at Cubatão V.Parisi, Sorocaba, and Santa Gertrudes stations. Average concentrations of 2.3, 5.3, 21.1, and 64.8 µg·m⁻³ were observed in 2019 for HUWRMs 2, 5, 6, and 7, respectively, and 1.9 23.8, 12.8, and 70.9 µg·m⁻³ for the same regions in 2020. During the restricted period, a significant reduction was also observed in most stations. Significant increases were observed only for Cubatão V.Parisi and Santa Gertrudes stations, as already mentioned for NO_x and NO₂ pollutants. The concentrations measured in 2019 for HUWRMs 2, 5, 6, and 7 were 7.2, 14.5, 31.0, and 96.4 µg·m⁻³, respectively, and 5.34, 28.2, 20.9, and 138.7 µg·m⁻³ in 2020. Therefore, when comparing the pandemic period with previous years, the concentrations suggest significant variations, increasing or decreasing, depending on the region of study and location of the stations.

Figure 6.9 – Same as in Figure 6.3, but for NO ($\mu\text{g}\cdot\text{m}^{-3}$)



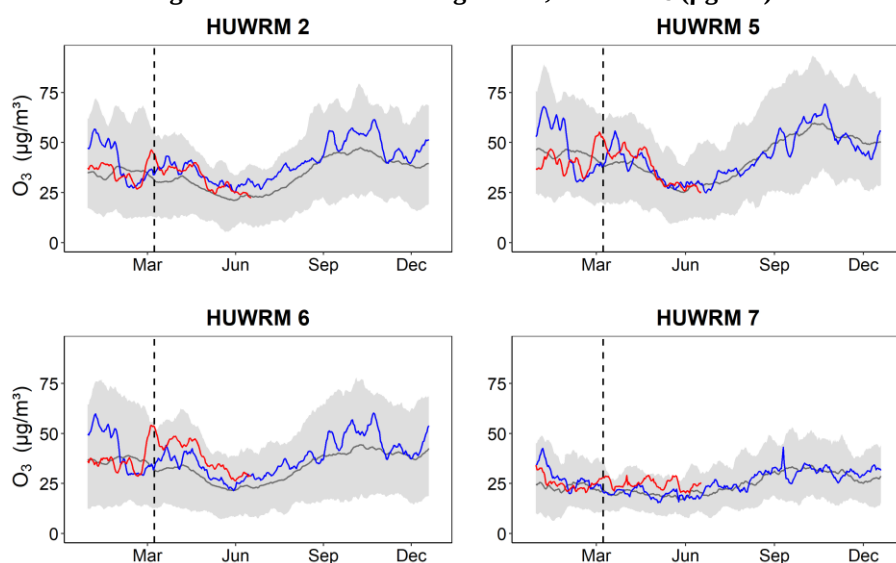
5.3.1.6 O₃

The historical average and the interquartile range of O₃ concentrations in HUWRMs 2, 5, 6, and 7 were 34.2 [24.6 - 42.3], 42.8 [31.0 - 53.1], 34.4 [24.3 - 43.2], and 25.1 [17.3 - 31.8] $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The highest concentrations of O₃ were observed in HUWRM 5, where high concentrations are associated with emissions of ozone precursors from fixed and mobile sources and the chemical regime (VOCs-limited or NO_x-limited). Modeling studies carried out in the metropolitan region of Campinas, the location with the highest average concentrations, indicate that the meteorological conditions in the region, associated with the topographic characteristic, are related to the low dispersion of ozone and precursors and favor transportation from the metropolitan region of São Paulo (BOIAN; ANDRADE, 2012). HUWRM 6 also has high concentrations, mainly related to vehicular emissions of NO_x and VOCs, impacted by the type of fuel used (high ethanol content) and the age of their fleet (ALVIM et al., 2018).

In contrast to the other pollutants, O₃ concentrations for 2019 and 2020 are above the average values of the historical series, showing a trend of increasing (Figure 6.10). When comparing the first period of the years 2019 and 2020, lower average concentrations are observed for 2020, but with a non-significant difference ($p\text{-value} > 0.05$) for most stations. Average concentrations observed for the first period of 2019 were 40.8, 47.3, 42.1, and 29.3 $\mu\text{g}\cdot\text{m}^{-3}$ for HUWRMs 2, 5, 6, and 7, respectively, and 36.1, 41.4, 37.3, and 25.8 $\mu\text{g}\cdot\text{m}^{-3}$ for the same regions in 2020. During the restricted period, significant increases in O₃ concentrations were observed in most stations, especially those at MASP (HUWRM 6). HUWRMs 2, 5, 6, and 7 showed average concentrations during the restrictions period, respectively, of 33.5, 36.1, 32.1, and 19.8 $\mu\text{g}\cdot\text{m}^{-3}$.

in 2019, and 31.3, 36.5, 37.8, and 25.0 $\mu\text{g}\cdot\text{m}^{-3}$ for the same period in 2020. The increases in O_3 concentrations in HUWRM 6 are strictly related to reductions in NO_x concentrations (NAKADA; URBAN, 2020; ORLANDO et al., 2010) since in MASP, as in several other large urban areas in the world, the chemical regime is usually VOC-limited (ANDRADE et al., 2017; MARTINS; ANDRADE, 2008). This behavior has already been evaluated through simulations carried out for the region (ALVIM et al., 2018; ORLANDO et al., 2010), as well as it was observed in other areas during the outbreak of COVID-19 (COLLIVIGNARELLI et al., 2020; SHARMA et al., 2020; SICARD et al., 2020; SICILIANO et al., 2020b; WANG et al., 2020a).

Figure 6.10 – Same as in Figure 6.3, but for O_3 ($\mu\text{g}\cdot\text{m}^{-3}$)



6.3.2 Mobility and social distancing

Figure 6.11 shows the evolution of the trend of displacement of cell phone users during the first half of 2020, whose movements were grouped into travel by private vehicles, public transport, and walking. The percentage shown in Figure 6.11 is relative to the volume of displacement on January 13, 2020, when the effects of pandemic were non-existent in the State of São Paulo. Although they are a non-representative sample of society, since it considers users of a specific device, it is robust and capable of identifying trends in changing displacement patterns of people.

Figure 6.11 – Evolution in the displacement of cell phone users during the first half of 2020. The vertical dashed line indicates the 76th day of the year 2020 when the first restrictions were started

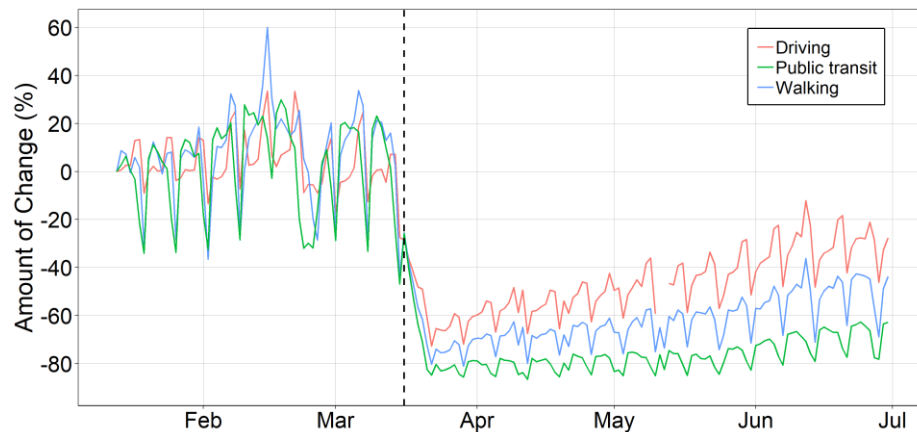


Figure 6.11 shows an increasing trend in displacement as of February, which coincides with Brazil's summer school holidays ends. From 14 to February 16, there is also a marked increase in the circulation of people on foot, which coincides with a date called "pre-carnival", an occasion when crowds go to the streets to dance. The graph clearly shows that it is a weekend completely different from the others and that, instead of a normal decreasing, there is an increase in the number of people walking. The increase in the number of vehicles is also evident, just as the decrease in public transport is not so marked. On the weekend following the "pre-carnival", which is a little longer, the traditional carnival party occurs from February 21 to 24. It is interesting to note the opposite behavior for the "pre-carnival" time, where there are no festive activities on the streets of the cities of the State of São Paulo, so different of what happens in coastal towns in northeastern Brazil, where many people go to streets during the carnival. In São Paulo, during this festival, the people prefer to stay at home or travel to other locations, including those with the street carnival. From March 16 to 22, there is a sharp drop in mobility of the people due to some restrictions to contain the pandemic, such as closing schools and universities and canceling public events (SICILIANO et al., 2020a). During this period, there was a constant reduction in displacement, which reached an average decrease of more than 50% in relation to the reference date (January 13). From March 23 to April 03, a Partial lockdown occurred. Some other restrictions have been applied, such as closing shopping centers, restaurants, bars, beaches, and all the non-essential business and substantially reducing public transport. There was an average reduction of more than 60% during those days, reaching a decrease of more than 70% in driving and 80% in public transport displacements. The period from April 4 to June 30 is considered a "relaxed" partial lockdown; because, during those days, many of the closed shops have reopened, but with reduced opening hours. Even with relaxation

in restrictions, the reduction in the displacements remained below those registered for the reference date, with an average decrease of 60% for driving and 76% for public transport.

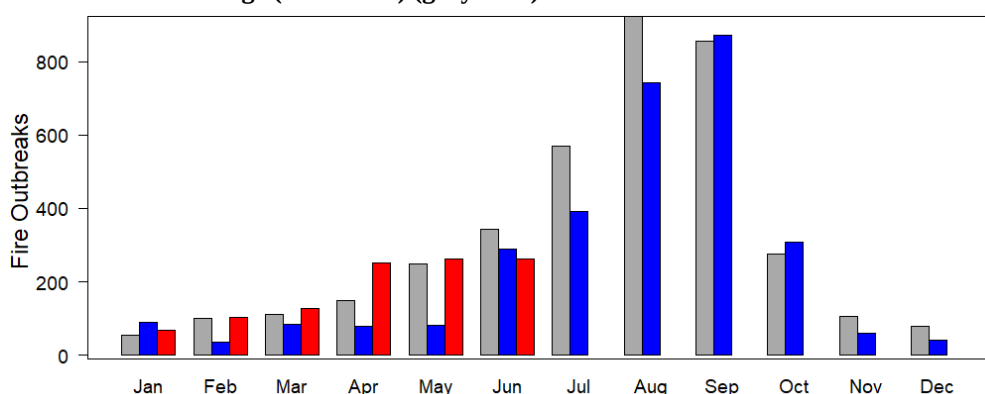
6.3.3 Atmospheric conditions

The general conditions of atmospheric circulation play an important role in the concentration of pollutants (MARTINS et al., 2018; PETETIN et al., 2020; TARGINO et al., 2019). For the study region, precipitation and long-distance transport conditions undergo considerable variations from one year to another. They can amplify or even completely cancel the effect of the pandemic restriction on air quality. This is especially true for precipitation, which has a high capacity for cleaning the atmosphere. The wet deposition is considered an important pathway for removing several air pollutants (GUO et al., 2014; PAN et al., 2017; YOO et al., 2014). Yoo et al. (2014) indicate that scavenging of air pollutants is directly related to rain intensity. The authors also suggest that precipitation is especially associated with PM removal, followed by SO₂, NO₂, CO, and O₃.

By comparing the variation of the atmospheric pollutant concentrations (Figure 6.3 to Figure 6.10) and the precipitation data (Figure B.7), the importance of the rain scavenging in the region is clear. The rainy season (October to March) is the period with the lowest concentrations of pollutants. In contrast, the dry season (April to September) is the period with the highest concentration of pollutants. For the first 45 days of 2020, the period with the most significant differences in the average concentrations for PM_{2.5}, the accumulated rainfall was significantly higher than the previous year (266.2 mm in 2019 and 415.5 mm in 2020) for the State of São Paulo. On the other hand, in March, coinciding with the beginning of the restrictions resulting from the pandemic, there was a significant decrease in precipitation, which lasted through April and May, potentially contributing significantly to the increase in the concentration of pollutants. A more detailed analysis can be found in the Appendix B (Figure B.7 and Figure B.8). For all stations with available data, it is confirmed that the absence of rain contributes significantly to the increase in the concentration of pollutants. The likely reduction in pollutants due to the restrictions of the pandemic may have been prevented as the rains decreased from April. However, as we moved away from the start date of the pandemic restrictions, the concentration increased substantially, almost following the expected pattern of high concentrations of pollutants for the dry season.

In addition, the absence of rain is directly related to the increase in the number of fire outbreaks. Although the maximum number of fires in the region occurs in the second half of each year, which is outside the study period, a few persistent fires are enough to disturb the pollutants concentration measured by an air quality monitoring network. There were more than 250 fire outbreaks in each month during the months of April and May in 2020, about three times the values observed for the same period in 2019 (Figure 6.12), which are expected to have a measurable impact on the region.

Figure 6.12 – Monthly number of fire outbreaks for 2019 (blue bars), 2020 (red bars), and historical average (2003-2018) (gray bars) for the State of São Paulo



Fires play an important role in changing the level of pollutant concentrations in the region (LOPES et al., 2012; MARTINS et al., 2018; PEREIRA et al., 2011; TARGINO et al., 2019). In 2020, this became evident by comparing the period before and during the pandemic, considering only days without rainfall events (Table B.3). The results show that even in a period with reduced vehicle traffic (Figure 6.11), there is a significant increase in pollutant concentrations, especially in the interior of the State (Table B.3). Comparing the period during the pandemic and before for HUWRM 6, no significant changes are observed in most stations, especially for CO and NO₂ pollutants (pollutants related to vehicular transport), demonstrating a low impact of mobility restrictions when the entire period is analyzed.

The importance of fires in the air quality of the region is evident when we analyze the results in Figure 6.12; there is a substantial increase in fires in the region, whose emissions could be transported to the areas where the monitoring stations are located (Figure B.9). An example of such an assertion can be seen when analyzing particulate matter concentrations for the Itaim Paulista station, the station with the largest increases in PM_{2.5} concentration during the pandemic. The highest concentrations of 2020 recorded in the station were observed from May 30 to June 2. In the same period, approximately 10% of the fire outbreaks of 2020 were

registered in the São Paulo State, playing an important role in worsening the air quality of the region (Figure B.9 shows the direction of the trajectories and the location of the fire outbreaks). This demonstrates the impact that pollutant transport has on the region, especially that related to the biomass burning emissions, which reached record levels in 2020 (TOMAZELA, 2020), even during the pandemic period.

6.4 Conclusions

This study evaluated the behavior of pollutant concentrations for the Metropolitan Area of São Paulo and its neighboring under the effects of the COVID-19 pandemic restrictions. In addition to historical averages, we compared the behavior of 2019 with the before and during mobility restrictions periods of 2020 for the main pollutants of interest for air quality studies. The region of the study was divided into sub-regions to facilitate the understanding of the behavior of pollutants. The area of interest can be understood as a nucleus influenced mainly by vehicular emissions but surrounded by predominantly industrial emissions and influenced by long-range transport. The main findings of this study are:

- 1) For most of the analyzed pollutants, there was no substantial reduction in the pollutant concentration for the period of COVID-19 restrictions compared to the before period months of 2020;
- 2) The few stations where significant reductions were observed are in the central core of the study region, which is the most impacted by vehicular emissions, with a decrease in primary pollutants CO and NO_x, and an increase in O₃;
- 3) Compared to historical averages, 2019 and the months analyzed for 2020 showed significant reductions in pollutant concentration for most stations, mainly resulting from public policies to control emissions. However, this generalized reduction was not reflected in O₃;
- 4) Some stations located in peripheral industrial areas showed an increase in the concentration of pollutants for 2020 compared to 2019, which may be associated with the specific industrial activities around those stations. The beginning of the pandemic may have coincided with the time of increase in some specific industrial activities; the pandemic may also have impacted the demand from some industrial sectors;

5) Despite being the rainy season, it was observed that precipitation was much above average in the period January to March 2020 and below average after the COVID-19 outbreak. During the dry season (fall and wintertime) the precipitation decreased, and as a tendency, the concentrations increased following the seasonality behavior. The rainfall above (below) the average before (after) the beginning of the pandemic restrictions promoted a reduction (increase) in the concentrations for the months before (after) the beginning of the pandemic in 2020 in comparison to the same period of 2019;

6) Above the average number of fire outbreaks in the months following the start of the pandemic restrictions may have impacted the study area and, together with abnormal rainfall distribution, prevent obvious evidence of improved air quality due to reduced human activities from the restrictions.

Finally, we suggest that any study focusing on analyzing the effects of the pandemic on air quality should consider the atmospheric conditions for the period. Neglecting such atmospheric contribution may produce misinterpretation of the results, especially when evaluating the effectiveness of emission control measures. Weeks with highly variable pollutant concentration alternated throughout the study period, suggesting that short analyses without considering seasonality can lead to misleading conclusions. The regionalization of the analysis is also fundamental for the adequate treatment of the different pollution sources, especially the vehicular and industrial, which are the dominant sources in the study region.

7 EVALUATING TROPOMI AND MODIS PERFORMANCE TO CAPTURE THE DYNAMIC OF AIR POLLUTION IN SÃO PAULO STATE: A CASE STUDY DURING THE COVID-19 OUTBREAK³

Abstract: Atmospheric pollutant data retrieved through satellite sensors are continually used to assess changes in air quality in the lower atmosphere. During the COVID-19 pandemic, several studies started to use satellite measurements to evaluate changes in air quality in many different regions worldwide. However, although satellite data is continuously validated, it is known that its accuracy may vary between monitored areas, requiring regionalized quality assessments. Thus, this study aimed to evaluate whether satellites could measure changes in the air quality of the state of São Paulo, Brazil, during the COVID-19 outbreak; and to verify the relationship between satellite-based data [Tropospheric NO₂ column density and Aerosol Optical Depth (AOD)] and ground-based concentrations [NO₂ and particulate material (PM; coarse: PM₁₀ and fine: PM_{2.5})]. For this purpose, tropospheric NO₂ obtained from the TROPOMI sensor and AOD retrieved from MODIS sensor data by using the Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm were compared with concentrations obtained from 50 automatic ground monitoring stations. The results showed low correlations between PM and AOD. For PM₁₀, most stations showed correlations lower than 0.2, which were not significant. The results for PM_{2.5} were similar, but some stations showed good correlations for specific periods (before or during the COVID-19 outbreak). Satellite-based Tropospheric NO₂ proved to be a good predictor for NO₂ concentrations at ground level. Considering all stations with NO₂ measurements, correlations greater than 0.6 were observed, reaching 0.8 for specific stations and periods. In general, it was observed that regions with a more industrialized profile had the best correlations, in contrast with rural areas. In addition, it was observed about 57% reductions in tropospheric NO₂ throughout the state of São Paulo during the COVID-19 outbreak. Variations in air pollutants were linked to the region economic vocation, since there were reductions in industrialized areas (at least 50% of the industrialized areas showed more than 20% decrease in NO₂) and increases in areas with farming and livestock characteristics (about 70% of those areas showed increase in NO₂). Our results demonstrate that Tropospheric NO₂ column densities can serve as good predictors of NO₂ concentrations at ground level. For MAIAC-AOD, a weak relationship was observed, requiring the evaluation of other possible predictors to describe the relationship with PM. Thus, it is

³ Rudke, A.P., Martins, J.A., Hallak, R., Martins, L.D., de Almeida, D.S., Beal, A., Freitas, E.D., Andrade, M.F., Koutrakis, P., Albuquerque, T.T.A. Evaluating TROPOMI and MODIS performance to capture the dynamic of air pollution in São Paulo state: A case study during the COVID-19 outbreak. *Remote Sensing of Environment*, v. 289, n. October 2022, p. 113514, 2023. DOI: 10.1016/j.rse.2023.113514.

concluded that regionalized assessment of satellite data accuracy is essential for assertive estimates on a regional/local level. Good quality information retrieved at specific polluted areas does not assure a worldwide use of remote sensor data.

Keywords: AOD; MAIAC; Air Quality; NO₂ Monitoring; Particulate Matter Monitoring; Sentinel-5P.

7.1 Introduction

The global pandemic of coronavirus disease 2019 (COVID-19) has been causing an overload on health care systems worldwide due to the high rate of hospitalized people (DRIGGIN et al., 2020; ORTIZ-PRADO et al., 2020). Thus, measures to prevent the transmission of the virus have been adopted around the world. Among these measures, mobility restriction was proposed by several governments, including the imposition of a hard lockdown in regions most affected by the COVID-19 outbreak (ENGLE; STROMME; ZHOU, 2020; TANG, 2020; THE LANCET, 2020a; ZHANG et al., 2021; ZHOU et al., 2020). These restrictions suggested by the World Health Organization (WHO, 2020) have proven helpful and were replicated in different parts of the world. In addition, they were effective not only in reducing the transmission of the virus, but also resulted in the reduction in the concentration of various air pollutants associated with vehicular and industrial emissions (BAO; ZHANG, 2020; BAUWENS et al., 2020; DANTAS et al., 2020; FAN et al., 2020b; KRECL et al., 2020; VENTER et al., 2020b; WANG et al., 2021).

Mobility restrictions were mainly imposed in large urban centers, where transmission rates were higher (LI; DAI, 2020). Studies carried out during lockdown periods have shown significant improvements in air quality in several parts of the globe, especially in Europe, Asia, and North America (e.g., Bao and Zhang, 2020; Berman and Ebisu, 2020; Cicala et al., 2020; Collivignarelli et al., 2020; Dutheil et al., 2020; Gama et al., 2021; Gkatzelis et al., 2021; Lokhandwala and Gautam, 2020; Sharma et al., 2020; Tian et al., 2021). The main conclusions of the studies based on surface measurements are that, in the short term, reductions occurred in the concentrations of particulate matter (PM_{2.5} and PM₁₀), carbon monoxide (CO), nitrogen oxides (NO_x), and sulfur dioxide (SO₂). On the other hand, increases in tropospheric ozone concentrations (O₃) were observed, especially in large urban centers where Volatile Organic Compounds (VOC) are commonly limiting in the atmospheric chemical regime of ozone

(COLLIVIGNARELLI et al., 2020; SHARMA et al., 2020; SICARD et al., 2020; SICILIANO et al., 2020b; WANG et al., 2020a).

Globally, most urban areas do not have robust air quality monitoring networks, making it difficult to assess the continuous spatio-temporal profile of air pollutants. However, most of the pandemic-related discussions about the drop in air pollution were developed based on remote sensing information (BAUWENS et al., 2020; DEVARA et al., 2020; DIAMOND; WOOD, 2020b; FAN et al., 2020a; NAKADA; URBAN, 2020). In this case, the main evaluated pollutants have been tropospheric nitrogen dioxide (NO₂) and columnar aerosol/PM loading. NO₂ data have been largely observed through the sensors Tropospheric Monitoring Instrument (TROPOMI) onboard Sentinel-5 Precursor and Ozone Monitoring Instrument (OMI) onboard Aura (e.g., Bauwens et al., 2020; Diamond and Wood, 2020; Fan et al., 2020a; Nakada and Urban, 2020; Tobías et al., 2020; Zambrano-Monserrate et al., 2020). Aerosol/PM information has been estimated through the Aerosol Optical Depth (AOD), retrieved predominantly from MODIS sensor data (DIAMOND; WOOD, 2020b; FAN et al., 2020a; ZAMBRANO-MONSERRATE; RUANO; SANCHEZ-ALCALDE, 2020).

Remote sensing-based measurements are essential in areas not covered by air quality monitoring networks, especially in developing countries of Africa, Asia, and Latin America. For instance, until 2014, air quality measures were carried out in a few major Brazilian cities (Réquia et al., 2015), corresponding to only 0.03 air quality monitoring stations per 1,000 km², a number much smaller than in the USA (0.5), Japan (4.9), and Germany (5.18) (RÉQUIA; KOUTRAKIS; ROIG, 2015). Satellite remote sensing is a tool with enormous potential to fill this gap. However, it is known that, despite the retrieved information being useful for a given region, it could present several limitations when applied in a different region, restricting its use for decision-making purposes. Retrieval uncertainties are regionally dependent on several environmental factors, such as those related to particle emission from different geo-climatic and/or degrees of urbanization, particle composition and size range, prevailing synoptic conditions, and local meteorological conditions (e.g., humidity, wind, temperature), affecting both satellite and ground-level measurements (FALAH et al., 2021). In the case of Brazilian urban areas, for example, the transport of biomass burning emissions from the Amazon and Cerrado produces very high concentrations of pollutants, frequently exceeding air quality standards (Martins et al., 2018; Pereira et al., 2011; Rudke et al., 2021; Targino et al., 2019). Therefore, although many studies have already conducted quality assessments for multiple

satellite data in China, the USA, and some European countries (HE et al., 2017b; IALONGO et al., 2020; LIU et al., 2019a; NICHOL; BILAL, 2016; TACK et al., 2021; VERHOELST et al., 2021), there is still a need for regionalized evaluations in areas with distinctive local characteristics. In this context, the sharp drops in air pollutant levels observed during the COVID-19 outbreak (COLLIVIGNARELLI et al., 2020; GKATZELIS et al., 2021; NAKADA; URBAN, 2020; RUDKE et al., 2021a, 2022) can configure an ideal scenario to evaluate the performance of satellite remote sensing in capturing surface air quality variability.

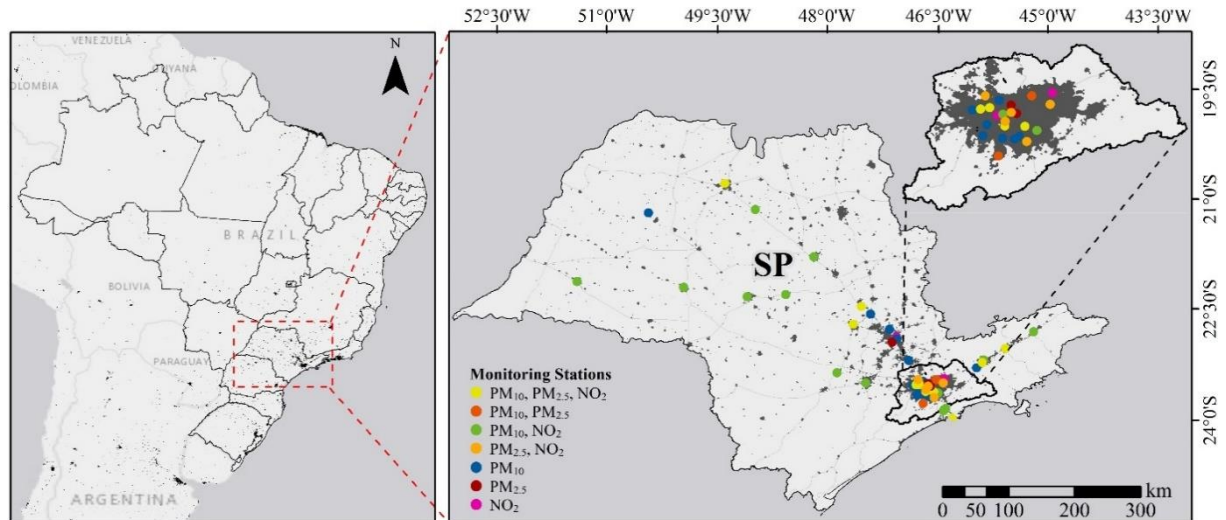
The COVID-19 pandemic is a rare event, which has shown significant variations in the concentrations of air pollutants over several locations worldwide. Therefore, it serves as an open-air laboratory for assessing the applicability of remote sensing products to map the regional dynamic of air pollutants, especially sudden changes in pollution levels. In this direction, this study aims to evaluate the performance of two satellite sensors, Moderate Resolution Imaging Spectroradiometer (MODIS) (JUSTICE et al., 1998) and Tropospheric Monitoring Instrument (TROPOMI) (VEEFKIND et al., 2012), designed to monitor the variability of AOD and tropospheric NO₂, respectively. For this purpose, the variability of AOD and tropospheric NO₂ column density data were evaluated against ground-based data during the partial mobility restriction imposed by the COVID-19 outbreak over the state of São Paulo, Brazil. This area was selected, as it is the Brazilian state with the most robust air quality monitoring network.

7.2 Methodology

7.2.1 Description of the Study Area

The study was conducted in the state of São Paulo (Figure 7.1), the most populous Brazilian state - more than 46 million inhabitants living in 645 cities spread over approximately 250,000 km² (CETESB, 2020; IBGE, 2020d). The state is located in an essentially humid climate zone, with tropical climate areas in the northwest and on the coast (ALVARES et al., 2013). São Paulo presents different economic vocations and is considered the most economically developed state in Brazil, primarily due to its industrial, farming, and livestock activities (CETESB, 2020). Together with industries and biomass burning, the large vehicle fleet makes the region the primary source of air pollutants in Brazil (ANDRADE et al., 2017; CETESB, 2020; PEREIRA et al., 2017b).

Figure 7.1 - Location of the study area. The dots over the State of São Paulo (SP) represent the locations of the CETESB monitoring stations used in this study, and the area emphasized with the largest number of stations represents the Metropolitan Area of São Paulo (MASP). The urban areas were based on Visible Infrared Imaging Radiometer Suite (VIIRS) Nighttime Light data from 2020 (<https://eogdata.mines.edu/products/vnl/>).



Although it is considered the state that produces the most pollutants in the country, São Paulo has experienced a significant reduction in the concentrations of various pollutants over the years (ANDRADE et al., 2017; CETESB, 2020; RUDKE et al., 2021a). It is currently known that a large amount of this reduction is related to Brazilian air pollution control policies, namely motor vehicle air pollution control programs (PROCONVE) and air pollution control programs for motorcycles and similar vehicles (PROMOT), in addition to local mobile and fixed emission control programs (ANDRADE et al., 2017; CARVALHO et al., 2015; GÓMEZ PELÁEZ et al., 2020). Besides local sources, long-range transport of air pollutants from biomass burning (both forest and agricultural) plays a significant role in the study area, as suggested by recent research (e.g., Martins et al., 2018; Pereira et al., 2011; Squizzato et al., 2021; Targino et al., 2019). Therefore, during the biomass burning season (August to November), the real pollution levels in the region are highly dependent on the national policy to control fires in the Amazon region, Cerrado and, more recently, the Pantanal.

The Metropolitan Area of São Paulo (MASP, Figure 7.1) is the largest urban area in Brazil. Approximately half of the state of São Paulo's population lives in this region (21.6 million inhabitants). The main city of MASP is São Paulo (capital of the State), which is among the most populated cities in the world (≈ 12.3 million inhabitants) and is the foremost industrial center in Latin America. Similarly to other major urban centers in the State, this region has the most significant air quality degradation related to mobile sources (CETESB, 2020). Mobile

sources in the MASP are responsible for around 97% of the emissions of CO, 84% of hydrocarbons (HC), 64 % of NO_x, 26% of PM, and 17% of sulfur oxides (SO_x) (CETESB, 2019a). Other critical air pollution hotspots in the State of São Paulo are the industrial centers of Cubatão and Santa Gertrudes (CETESB, 2020; RUDKE et al., 2021a). Cubatão is known for its essential petrochemical, steel, chemical, and fertilizer sectors, which account for most of the national industry primary products (VIEIRA-FILHO; LEHMANN; FORNARO, 2015). The industrial sector is the main source of pollutants in Cubatão, a city known worldwide in the past as one of the most polluted places in the world. The rugged topography and unfavorable weather conditions have also hindered the dispersion of pollutants (CETESB, 2020; VIEIRA-FILHO; LEHMANN; FORNARO, 2015). The industrial hub of Santa Gertrudes is also a significant source of air pollutants for the region, mainly particulate matter, since this is the largest producer of ceramics in the country (CETESB, 2020).

7.2.2 Data Sets

The data used in this study are ground-based concentrations of PM₁₀, PM_{2.5}, and NO₂, besides satellite-based Tropospheric NO₂ column density and AOD. The ground-based concentrations were obtained from the Environmental Company of São Paulo State (CETESB) air quality monitoring network (<https://qualar.cetesb.sp.gov.br>). NO₂ satellite data are from the TROPOspheric Monitoring Instrument (TROPOMI), accessed through the Copernicus Open Access Hub (<https://scihub.copernicus.eu>). AOD data from the Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm were obtained from the National Aeronautics and Space Administration (NASA) webpage (<https://ladsweb.modaps.eosdis.nasa.gov>). Among the algorithms used to retrieve AOD from MODIS sensor (Deep Blue, Dark Target, and MAIAC), MAIAC was used because it has the best spatial resolution (1 km) and is widely used to estimate PM in epidemiological studies (e.g., Cohen et al., 2017; Just et al., 2015; Kloog et al., 2014; Li et al., 2021; Maheshwarkar and Sunder Raman, 2021).

The dataset adopted for this assessment considers the first semester of 2019 and 2020. To represent the first half of 2020 the period from December 2019 to June 2020 was considered and to represent the first half of 2019 the period from December 2018 to June 2019. This practice was followed due to lower data availability before the mobility restrictions (March 16), since this is the rainy season in the study region, when clouds significantly reduce data retrieval. The characteristics of the data used in this study are briefly described below.

7.2.2.1 Ground-Based Data

According to CETESB, in 2019, São Paulo had 62 fixed automatic stations, two automatic mobile stations, and 23 manual monitoring points (CETESB, 2020). Thus, São Paulo has a density of 0.35 stations per 1,000 km², while Brazil has 0.03. The State of São Paulo is, therefore, the best choice of location for assessing the quality of remote sensing data in Brazil.

In this study, we considered only 50 automatic air quality monitoring stations providing measurements of at least one pollutant of interest (PM₁₀, PM_{2.5}, and/or NO₂), and with data available for the analysis period (first semesters of 2019 and 2020). Of these, 39 stations were used to measure PM₁₀, 20 for PM_{2.5}, and 32 for NO₂ (Figure 7.1). The information of the selected stations is available in Table C.1 (Appendix C). The data provided by the CETESB automatic stations correspond to data constantly taken from the atmosphere at intervals of five seconds, automatically accumulated in the form of an hourly average and made available for access.

7.2.2.2 Satellite Data

TROPOMI

TROPOMI was launched in October 2017 and is a passive hyperspectral nadir-viewing imager aboard the ESA's Copernicus Sentinel-5 Precursor (S-5P) satellite. The swath width is approximately 2600 km, resulting in almost daily global coverage (equator crossing time at 13:30 LT) (Veefkind et al., 2012). This Instrument has received particular attention during the pandemic, which ended up impacting the concentration of pollutants (Faridi et al., 2021; Gkatzelis et al., 2021).

For the obtention of the NO₂ column density, TROPOMI measures the Earth radiance at 0.2-0.4 nm resolution in the visible absorption band (VAN GEFFEN et al., 2019). The procedure used to retrieve NO₂ vertical column density (VCD) is based on three steps: (i) the amount of NO₂ along the slant path (Slant Column Density - SCD) is derived using the Differential Optical Absorption Spectroscopy (DOAS) technique (PLATT; PERNER, 1983) (VERHOELST et al., 2021); (ii) the SCDs are incorporated by the TM5-MP Chemical Transport Model (CTM) that allocates the SCDs to a vertical profile of the NO₂ concentration to separate the stratospheric and tropospheric amounts (VERHOELST et al., 2021); and finally (iii) the SCDs are transformed to vertical column densities using Air Mass Factors (AMFs) (VAN GEFFEN et

al., 2019). Depending on the CTM mode used, forecast mode (1-day forecast meteorological) or delayed processing mode (12-hour forecast meteorological data), the near-real-time (NRTI) or offline (OFFL) TROPOMI measurements are generated (VERHOELST et al., 2021).

Although the S-5P satellite was launched in October 2017, NO₂ data are only available from the end of April 2018, after the end of the commissioning phase (the period used to verify the data chain, i.e., the instruments and ground-processing algorithms, before releasing the data to the users) (ESA, 2020). This study used tropospheric NO₂ column density data from Level 2 OFFL (processor version 1.3.2), which had a spatial resolution at the nadir of 7.0×3.5 km² until August 6, 2019 and a resolution of 5.5×3.5 km² afterward. The data are made available together with detailed quality flags. We followed the guidelines recommended by the producers for tropospheric applications that indicate to use only NO₂ information with a quality assurance value greater than 0.75, which removes very cloudy scenes, snow- or ice-covered scenes, and problematic retrievals (ESKES et al., 2019).

MODIS

Onboard the Terra and Aqua satellites, MODIS acquires data at the top of the atmosphere (TOA) since 1999 and 2002, respectively. The MODIS sensor has 36 spectral bands, which vary in wavelength from 0.4 µm to 14.4 µm in both satellites. The temporal resolution of the sensor is 1 to 2 days and moderate spatial resolutions of 250 m, 500 m, and 1000 m. Of the 36 spectral bands available on the sensor, 7 have primary use aimed at aerosols, with wavelengths ranging from 0.47 to 2.13 µm (referring to the visible spectrum region, near-infrared, and short-wave infrared) (NICHOL; BILAL, 2016). Due to the almost daily data and the long data series, the sensor has been used in numerous aerosol-related studies, mainly by using AOD data at 550 nm (HSU et al., 2013, 2004; LEVY et al., 2013; LEVY; REMER; DUBOVİK, 2007; LYAPUSTIN et al., 2011, 2018; REMER et al., 2005; TANRÉ et al., 1997).

AOD represents how much the aerosol layer can prevent the sunlight direct transmission through the atmosphere. Therefore, it is not possible to directly determine the concentration of the particles from satellite (or remote sensing), such as the traditional PM_{2.5} and PM₁₀ monitored by air quality monitoring stations. However, the relationship between AOD and surface level particulate matter has been evaluated for PM_{2.5} and PM₁₀, and the results indicate a good correlation (e.g., Just et al., 2015; Kloog et al., 2015, 2011; Li et al., 2021; Wang and Christopher, 2003). Therefore, although the relationship between the concentration of particles

and AOD may depend on several environmental factors, there is sufficient evidence for the use of AOD as an indicator of the greater or lesser presence of particles near the surface. Thus, in this study, we evaluated AOD estimates at a wavelength of 550 nm, as a predictor for particulate matter.

In this research, the MODIS MCD19A2 product was used for obtaining AOD data. MCD19A2 is a MODIS-AOD data retrieved using the MAIAC algorithm that combines information from the Aqua and Terra satellites (LYAPUSTIN et al., 2011, 2018). The MAIAC algorithm uses a multi-day time series analysis (4 days at the poles and 16 days at the equator) and different spatial scales for processing [e.g., 1 km grid cells (pixels), 25-km blocks, and 150 mesoscale areas km] (LYAPUSTIN et al., 2018). In the analysis, MAIAC uses 12 spectral bands that are nested in a 1-km grid. The MAIAC AOD retrieval has been evaluated over South America, demonstrating robust performance over heterogeneous surfaces (MARTINS et al., 2017b).

7.2.3 Comparison between satellite- and ground-based air quality measures

Satellite images have extensive spatial coverage at a short time interval, while ground-based measurements provide a high sampling rate at a local point. Thus, a direct comparison using the location of the air quality monitoring station or using only the satellite overpass time could restrict the probability of matchup data due to cloud cover or the time delay between datasets. For comparison, only the nearest pixels from each air quality monitoring station were used, considering extraction radii of 3, 5, and 10 km. Extraction radii smaller than 10 km were chosen not to excessively degrade the satellite data, allowing a more significant number of collocated data that appropriately represent air quality stations. Besides, the closest hourly ground-based measurement from the satellite overpass time was selected, considering a maximum temporal window of ± 1 hour. Data from December of the previous year of 2019 and 2020 were used to increase the amount of data in the period before the COVID-19 outbreak in Brazil (the first case was confirmed on February 26, 2020, and the beginning of mobility restrictions was on March 16), which is a rainy period in the region, reducing data collection probability by satellite. Therefore, to represent the year 2019, data from December 2018 to June 2019 were used, and the same strategy was used for 2020. Besides, March 16 was used as the framework for mobility restrictions, so the period before this date was called “before” the COVID-19 outbreak (December 1 to March 15), and the following period (March 16 to June 30) was considered as “during” COVID-19. Based on the extracted data, comparisons were made using Spearman’s

correlation coefficient, since the data do not have the same units, making it impossible to use other statistical approaches.

7.2.4 Change in air quality assessed by satellite observations

Changes in air quality over the State of São Paulo during the mobility restrictions were evaluated for Tropospheric NO₂ column density data. This analysis considered a short data series, 30 days before and after the beginning of the restrictions imposed by the COVID-19 (day of year 76), seeking to emphasize only the period with the most restrictive control practices (RUDKE et al., 2021a). Thus, satellite data from 2020 (February 15th to April 14th) was used in comparison with data from 2019 (February 15th to April 15th).

In addition to the comparison made for periods before and after the restrictions, the effect of the measures adopted to contain the COVID-19 outbreak was quantitatively estimated. In this sense, the equation, previously applied by Fan et al. (2020a), was used to estimate the effect of the mobility restrictions. Thus, based on data for pollutants before and after the restrictions equivalent period, but in 2019, together with data for the period before restrictions in 2020, the expected value for the period after the beginning of mobility restrictions in 2020 was estimated (Equation 7.1).

$$X_{2020after,est} = X_{2020before} \frac{X_{2019after}}{X_{2019before}} \quad (7.1)$$

Where X represents the evaluated pollutant. This equation assumes that the effect of emissions and the impact of meteorological factors are similar in the two periods compared.

Finally, based on the observed and expected values for the period after the beginning of the restrictions, the relative difference was calculated.

7.2.5 Atmospheric conditions

The role of meteorology on air quality variability is well known (ELMINIR, 2005; PEARCE et al., 2011) and must be considered to avoid misinterpretations regarding air quality changes. In some cases, the expected effects of the mobility restrictions on air pollutants may be reduced or intensified due to unconsidered variations in meteorological variables. In their review work, (GKATZELIS et al., 2021) present several methodologies that apply different techniques to estimate the variability induced by atmospheric conditions to better quantify the impact of

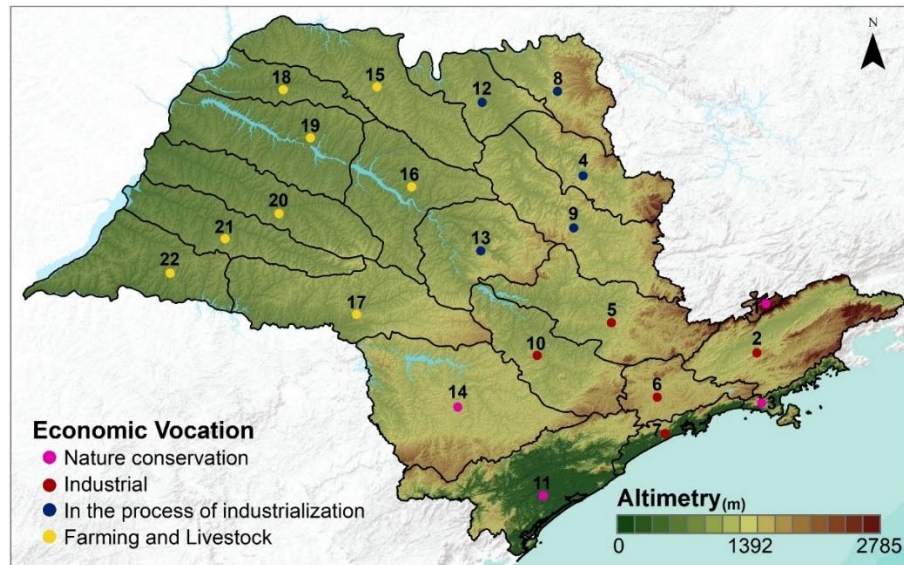
mobility restrictions. The authors mention that the most recurrent analyses are performed by normalizing pollutants using long-lived species such as CO, normalizing by dilution corrections using meteorological variables, benchmarking periods of similar meteorology, analyzing synoptic meteorological conditions, and performing back trajectory analysis using models as the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT).

Considering the importance of atmospheric circulation for the concentration of pollutants, we analyzed some meteorological fields and parameters associated with them [Convective Available Potential Energy (CAPE) and Precipitable Water (PWater), temperature, wind speed, total precipitation, Planetary Boundary Layer Height (PBLH), and Relative Humidity (RH)]. Weather condition is particularly important to consider possible factors related to the degradation of satellite data (AOD and Tropospheric NO₂ data retrievals), and/or affect the variation in the concentrations of atmospheric pollutants (weather variables have a great impact on the intra- and interannual variability of atmospheric pollutants). The analysis of the synoptic conditions for the State of São Paulo during the study period was based on the ECMWF Reanalysis v5 (ERA5), the fifth generation of the ECMWF atmospheric reanalysis (HERSBACH et al., 2020a). The global reanalysis is provided by Copernicus Climate Change Service (C3S) and covers the period from January 1950 to the present. The ECMWF model provides hourly estimates of atmospheric variables at a 30 km grid resolution, 137 levels (from surface up to 80 km). The data can be freely accessed on the following website: <https://www.ecmwf.int/en/forecasts/dataset/ecmwf-reanalysis-v5>.

7.3 Results and discussion

The State of São Paulo was divided into Hydrographic Units for Water Resources Management (HUWRMs) for better presentation and discussion of results. The convenience of HUWRMs considers the topography and climatic types/subtypes, which directly or indirectly affect the concentration of pollutants, and should be considered in air quality studies. Furthermore, each of the HUWRMs was previously identified according to their economic vocation by CETESB, which contributes to the discussion of the main factors related to local air pollution. The division by HUWRM and its assigned identification number are shown in Figure 7.2, which will be used to discuss the results. More information about the characteristics of HUWRMs can be found in CETESB (2020) and Rudke et al. (2021).

Figure 7.2 - Map of the 22 Hydrographic Units for Water Resources Management (HUWRMs) along with the State of São Paulo. The dot colors indicate the economic vocation of each HUWRM.



7.3.1 Mobility restrictions

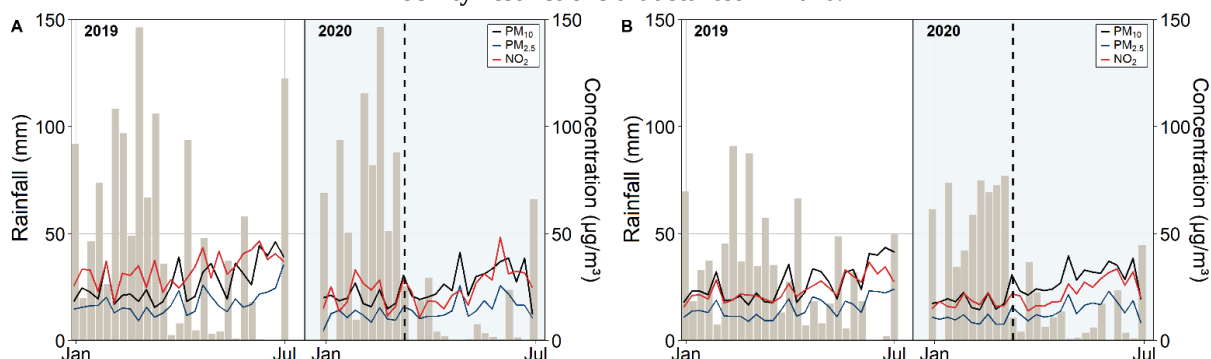
In mid-February 2020, confirmed cases of COVID-19 began to emerge in Brazil. The first death caused by COVID-19 occurred on March 12 in São Paulo State and, due to its vast population and the significant movement of people, São Paulo quickly became one of the main epicenters of COVID-19 in the country. To curb the transmission of the virus, the first mobility restrictions were applied on March 16. From March 16 to April 3, the first partial lockdown took place, where there was the closure of commerce in general, suspension of non-essential services, and the establishment of home office work (RUDKE et al., 2021a, 2022). In this period, there was a significant reduction in public transport displacements (80%) in the State and less vehicle circulation (about 60% reduction). Consequently, a decrease in air pollutant concentrations was observed in many cities across São Paulo for the first 30 days of restrictions (RUDKE et al., 2021a, 2022). After this period with the more significant restrictions, governments began to “relax” their sanctions, seeking to partially resume economic activities in the region. Thus, in mid-April, commerce and other non-essential activities started to work on shorter hours than those adopted before the pandemic. This fact caused the number of vehicles on the road to increase and, consequently, the concentrations of air pollutants began to rise (RUDKE et al., 2022). Figure C.1 shows how mobility restrictions reduced the number of displacements in the State of São Paulo. More information on how the sanctions adopted by the São Paulo government to reduce the transmission of COVID-19 impacted mobility and, consequently, the concentrations of pollutants can be obtained in the studies of (RUDKE et al., 2021a, 2022).

7.3.2 Atmospheric conditions

This section summarizes atmospheric conditions for the first semesters of 2019 and 2020 and their correlations with the studied air pollutants (PM_{10} , $PM_{2.5}$, and NO_2); Complete details are given in Section C1 - Atmospheric conditions and impact of fire emissions on measurements of the Appendix C. The results of the t-Student test using CAPE and PWater indicated that, generally, the energy available for deep convection was statistically different between January and June of 2019 in comparison to the same period of 2020. This is valid both for both São Paulo city and state. However, São Paulo city and state show statistically similar conditions regarding moisture for the first six months of the two years. These two variables indicate that different atmospheric conditions may have predominated in the region during the two years analyzed in this study. In addition, a visual estimate of the number of synoptic cyclones indicates 24 synoptic cyclonic vortices and associated cold fronts passing over São Paulo in 2019, against 31 such systems passing in the same region in 2020. This significant difference shows the predominance of different atmospheric conditions for the two years.

Although the first half of 2020 presents a greater activity of atmospheric systems crossing over the study region, such events were concentrated in the first months of the year, i.e., before the restrictions due to the pandemic. This differential temporal distribution between the two years significantly impacted the concentration of pollutants. As we can easily see in Figure 7.3, while in 2019, rainfall events remained reasonably distributed throughout the first half of the year, in 2020, there was a greater concentration in the first weeks of the year, cleaning the atmosphere in a period that precedes the mobility restrictions due to the pandemic. As a result, the atmosphere was relatively clean when the lockdown took effect. In the weeks following the lockdown, there was a significant reduction in convective activity, thus reducing the chance of pollutant dispersion/removal. Consequently, the expected decreases in the concentration of pollutants due to decreased human activities did not manifest as intensely as observed in other urban regions worldwide. There was even an increase in some of the pollutants in some stations (RUDKE et al., 2021a). This change in the behavior of the atmosphere, which occasionally coincided with the lockdown period, occurred throughout the State of São Paulo, but was much more pronounced for MASP.

Figure 7.3 - Observed weekly rainfall and air pollutant levels for (a) MASP and (b) State of São Paulo during the study period. Rainfall data were obtained from the National Institute of Meteorology (INMET) automatic stations (<https://portal.inmet.gov.br/>). The vertical dashed black line indicates the beginning of mobility restrictions that started in 2020.



The correlations between NO_2 , PM_{10} , and $\text{PM}_{2.5}$ measured at ground level and temperature, wind speed, precipitation, PBLH, and RH show that precipitation and relative humidity significantly impact air pollutant levels, especially for particulate matter. Comparisons between the period studied for 2019 and 2020 do not show significant differences in the correlations of meteorological variables and pollutant concentrations measured at ground level. However, it is possible to observe that, in general, the 2020 correlations were more significant than those of 2019. This fact may be related to reducing very localized sources due to mobility restrictions, especially on high-traffic roads.

Biomass burning is an important source of air pollutant emissions in Brazil. Analyzing the first semester of 2019 and 2020, it is observed that fire outbreaks have the same behavior for both years, but with a more expressive density mapped for 2020. The evaluation of the primary sources of air pollutants demonstrates that the places with an intense density of fires tend to impact the air quality of São Paulo. However, it is observed that the impact of long-range transport of aerosols and gases is lower when the entire period is evaluated than the more localized sources (which include local fire emissions). It should be noted that the period analyzed here (January to June) is not the typical period of biomass burning in Brazil, which extends from August to November.

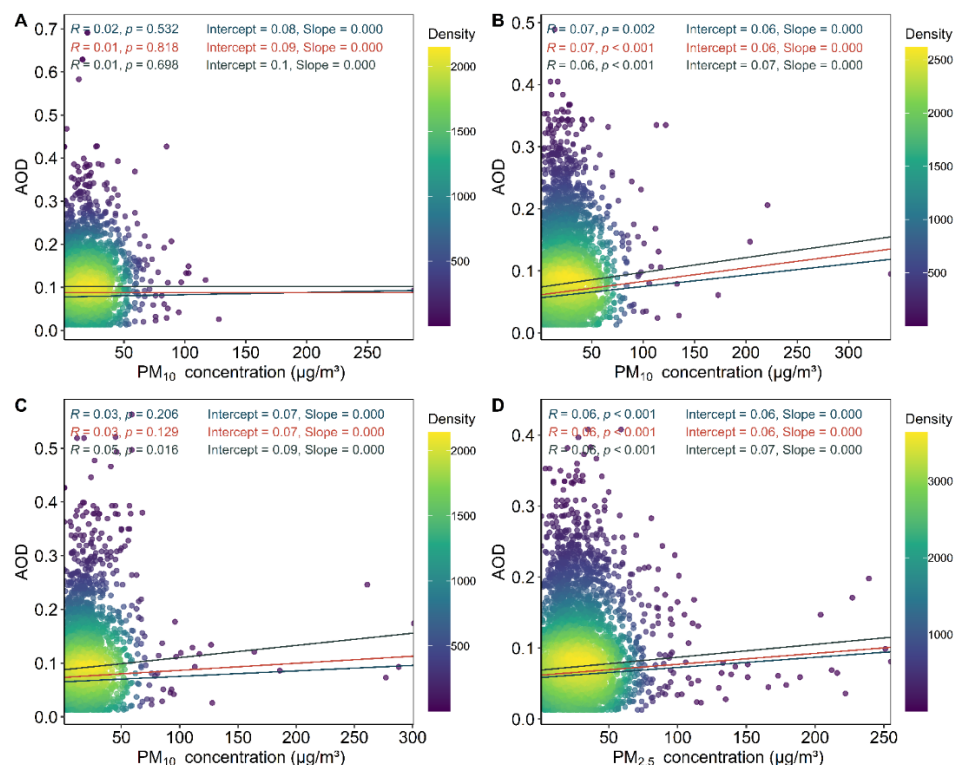
7.3.3 Quantitative Spatiotemporal Evaluation of Satellite Estimates

Due to uncertainties related to satellite data retrieval, in this section, we will evaluate tropospheric NO_2 and AOD data by comparing them to ground-based observations.

7.3.3.1– Satellite estimates - Aerosols (AOD)

The correlations between MAIAC-AOD and concentrations of PM₁₀ were calculated considering all available CETESB stations (Table C.1) and are presented in Figure 7.4 and Table C.2 and Table C.3 of the Appendix C. The AOD data were extracted considering the nearest satellite data (pixel centroid) into three extractions radii (10 km, 5 km, and 3 km from the CETESB station). In general, the correlations were largely non-significant ($\alpha > 0.05$) and, when significant, only weak or moderate correlations were found. The highest correlation values are associated with estimates based on the smallest area around the station (3 km). This relationship with the extraction radius reinforces the fact that, although distance increases the chance of obtaining samples for a given day, it also increases the inclusion of pixels that do not represent the station measurements. It is also observed that the correlations for 2020 are slightly higher than for 2019. In addition, the correlation results are consistent with previous research carried out in the region (e.g., Damascena et al., 2021; Natali, 2008).

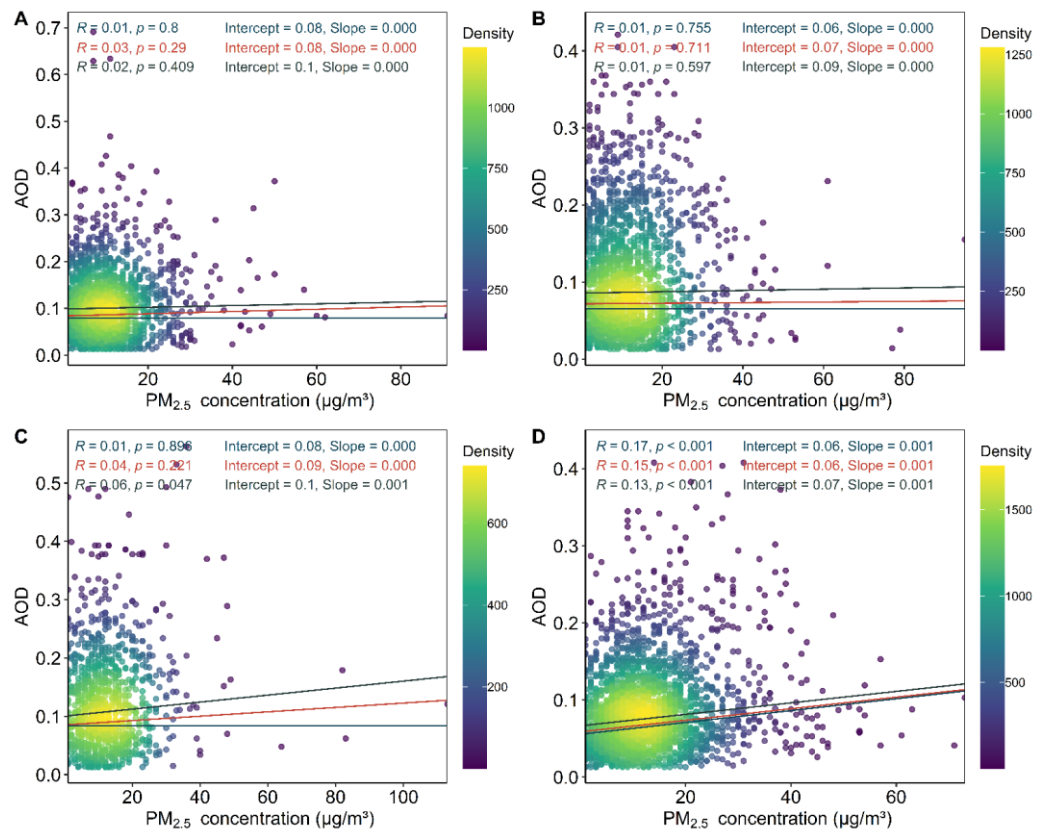
Figure 7.4 - Scatterplot of MODIS-AOD at 550 nm (Aqua and Terra satellites) and ground-based measurements of PM₁₀ for the periods: (A) before March 16, 2019; (B) after March 16, 2019; (C) before March 16, 2020; and (D) after March 16, 2020 (mobility restrictions). The blue, orange, and green lines indicate the regression line (Theil–Sen estimator) for AOD data extracted at 3, 5, and 10 km. The same colors indicate the correlation coefficient (R), p-value (p), intercept, and slope for each extraction radius.



Concerning PM_{2.5}, it is observed that the correlations with AOD were higher and more significant (Figure 7.5), when compared to PM₁₀ data, reaching 0.61 at Santa Gertrudes station (3 km radius) (Table C.4 and Table C.5). Although the correlations are relatively higher, it should be considered that the number of samples is lower; of the 213 daily data for the year

2020 (from 12-01-2019 to 06-30-2020), less than a quarter was used to calculate the correlations. One factor that should be mentioned is that, in general, the correlations were more significant for 2020 than for 2019. Furthermore, as observed for PM_{10} , the correlations considering data from the MODIS sensor coupled to the Terra satellite are smaller (and less significant) when compared to the sensor coupled to the Aqua satellite. This fact was also observed by Damascena et al. (2021, 2019) and Natali (2008) and may be related to satellite overpass times. The Terra satellite crosses the study region in mid-morning when the Planetary Boundary Layer (PBL) starts to evolve, and the aerosols are still not well mixed in the atmospheric column. In contrast, the Aqua crosses in the early afternoon, when the aerosols are well integrated into the atmospheric column (DAMASCENA et al., 2021).

Figure 7.5 - Same as in Figure 7.4, but for $PM_{2.5}$.



PBLH is considered a key factor with a direct influence on the relationship between satellite-derived AOD and ground-level PM since AOD is a measure of the total vertical column atmosphere, while PM is a measure performed at the surface. Thus, assuming that aerosols are homogeneously distributed under PBL, some researchers suggested that AOD values normalized by PBLH can be considered extinction at the surface level (GONG et al., 2017). In China, using MAIAC-AOD and $PM_{2.5}$, low correlations were observed for several urban

agglomerations ($R < 0.4$) (HE; WANG; YIM, 2021). The authors corrected the AOD data based on RH and PBLH, leading the correlations to more than 0.6 (HE; WANG; YIM, 2021). Damascena et al. (2021) corrected the AOD data considering these variables (RH and PBLH) for MASP, but the correlations remained weak (< 0.4). To assess the influence of PBL in the region and period studied, the AOD data were corrected considering the PBLH (AOD/PBLH). Table 1 shows the correlations between PM and AOD (observed and corrected). Overall, little change in correlations was observed for PBLH-corrected AOD. Slight improvements in correlations were kept mainly for the period before March 16 (beginning of mobility restrictions) and correlations with PM_{10} . When the correlations are observed for stations and clustering classes (Table C.2 to Table C.6), it is verified that the correction by PBLH has different effects throughout the state of São Paulo, with improvements in correlations in some regions and reductions in others. The low improvement or even decrease in correlations when AOD was corrected may demonstrate that the PBL is not well-mixed, as suggested by Damascena et al. (2021) for MASP. Furthermore, it is important to mention that the PBLH data used to correct AOD come from reanalysis (ERA5) and have a resolution of about 30 km, which may impact the accuracy of the data used for the correction.

Table 7.1 – Correlation between particulate matter (PM_{10} and $PM_{2.5}$) measured at ground level by CETESB stations and AOD data (observed and corrected) retrieved using MAIAC algorithm.

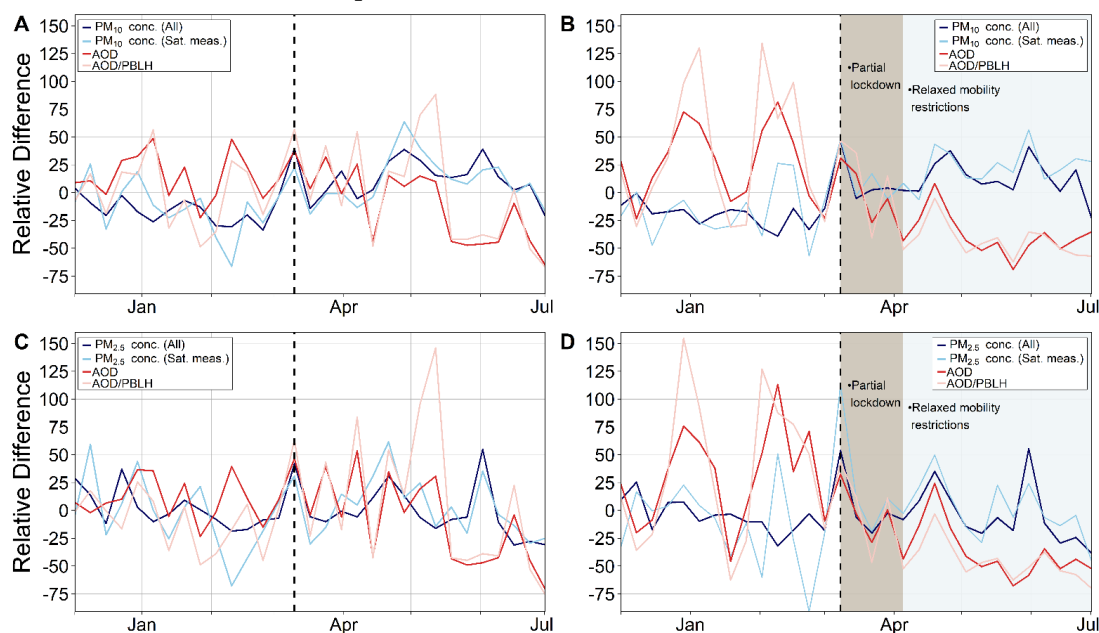
		2019-2020		2019 - Bef.		2019 - Aft.		2020 - Bef.		2020 - Aft.	
	ER	AOD	AOD/PBLH	AOD	AOD/PBLH	AOD	AOD/PBLH	AOD	AOD/PBLH	AOD	AOD/PBLH
$PM_{2.5}$	AQUA	3km	0.21***	0.19***	0.13	0.07	0.08	0.06	0.24*	0.28***	0.27***
		5km	0.2***	0.17***	0.12	0.08	0.07	0.06	0.35***	0.27***	0.24***
		10km	0.17***	0.15***	0.01	0.01	0.07	0.06	0.24***	0.28***	0.27***
	TERRA	3km	0.02	0.02	-0.07	-0.11*	-0.02	-0.03	-0.04	-0.03	0.13***
		5km	0.01	0.01	-0.04	-0.09*	-0.03	-0.03	-0.04	-0.04	0.12***
		10km	0	0.01	-0.03	-0.07*	-0.01	-0.01	-0.03	-0.01	0.08**
PM_{10}	AQUA	3km	0.08***	0.09***	0.2***	0.22***	0.13***	0.05	0.19**	0.21**	0.14***
		5km	0.05**	0.07***	0.12*	0.17***	0.14***	0.07*	0.18**	0.18**	0.13***
		10km	0.01	0.05**	0.06	0.14***	0.12***	0.06*	0.17***	0.2***	0.11***
	TERRA	3km	0.03*	0.03*	0	-0.01	0.04	0.01	0	0.02	0.09***
		5km	0.03*	0.03**	0.01	-0.01	0.04	0.02	0.03	0.05	0.08***
		10km	0.01	0.02*	0	0	0.04	0.03	0	0.03	0.06**

Note: ER – Extraction radius; AOD – correlation between PM and observed AOD; AOD/PBLH - correlation between PM and corrected AOD (AOD/PBLH). Before (Bef.) and after (Aft.) indicate the correlation for the period before and after March 16 - 2019 and 2020.

By disaggregating the concentrations of PM_{10} and $PM_{2.5}$ into class intervals (Figure C.2 and Figure C.3), it is observed that there is no great variation in the AOD values, even with high variations in the concentrations of particulate matter (PM_{10} and $PM_{2.5}$). Temporally, there is a low relationship between AOD at 550 nm and $PM_{10}/PM_{2.5}$ data. Considering the average of AOD and $PM_{10}/PM_{2.5}$ data for 2019 and the difference related to this period, it is observed that there is no great similarity between data behavior (Figure 7.6). Furthermore, it is difficult to assertively assess whether the AOD data were influenced by the mobility restrictions

implemented during the COVID-19 outbreak or whether the behavior is related to some other behavior inherent in the data. Spatially, there was no significant pattern regarding the variation in correlations, both for PM_{10} and $PM_{2.5}$ (Table C.6). However, regions with industrial economic vocation were the only ones that presented significant correlations ($\alpha > 0.05$), specifically HUWRMs 2, 5, and 7.

Figure 7.6 - Weekly dynamics of MODIS-AOD at 550 nm (Aqua) and ground-based PM_{10} for 2019 (A) and 2020 (B); and MODIS-AOD and ground-based $PM_{2.5}$ for 2019 (C) and 2020 (D). The vertical dashed black line indicates the 76th day of 2019 and 2020, when the first restrictions started in 2020. The averages represent data obtained through the highest extraction radius (10 km). The relative difference was calculated using variable averages for 2019 as a comparison. The light blue lines indicate weekly concentrations of particulate matter (PM_{10} and $PM_{2.5}$) that have matching satellite data. The dark blue lines indicate the weekly concentrations of particulate matter for all the data available in the satellite overpass, even without data for the satellite.



In general, the direct relationship between PM and AOD showed low correlations for the state of Sao Paulo (in many cases, not statistically significant), diverging from studies that demonstrated good correlations for these variables (GONG et al., 2017; HE; WANG; YIM, 2021; KONG et al., 2016; ZHENG et al., 2017). Damascena et al. (2021) indicate that the low sensitivity of MAIAC-AOD for MASP may be related to the background aerosol model used to characterize South America. The background aerosol model used to characterize South America is also used in AOD data retrieval for Northern North America and the Eastern United States (LYAPUSTIN et al., 2018). In these regions, several studies have shown good results in converting MAIAC-AOD to $PM_{2.5}$ (e.g., Chudnovsky et al., 2014; Di et al., 2016; Goldberg et al., 2019; Hu et al., 2014; Pu and Yoo, 2021). These differences between the state of São Paulo and the North American region may be related to the different chemical composition of aerosols

for the areas, as shown for MASP by Damascena et al. (2021). The model used for these regions is based on the climatic characteristics of the Goddard Space Flight Center (GSFC), an Aerosol Robotic Network (AERONET) site in eastern USA (DAMASCENA et al., 2021; LYAPUSTIN et al., 2018).

Evaluating the aerosol classification based on the AERONET data proposed by Logothetis et al. (2020) (Figure C.4), there are very different characteristics between the AERONET sites in GSFC and São Paulo (HUWRM 6) and Cachoeira Paulista (HUWRM 2). For GSFC, there is a homogeneous characteristic of aerosols, which are largely classified as “fine non-absorbing”. However, for São Paulo (HUWRM 6) and Cachoeira Paulista (HUWRM 2), there is great heterogeneity in the types of aerosols, ranging from “fine highly absorbing”, “fine moderately absorbing”, and “fine slightly absorbing”, which demonstrates a large discrepancy between the types of aerosols defined for GSFC. Furthermore, through aerosol volume size distribution (Figure C.4), it is observed that fine particles are dominant at GSFC, mainly in summer, while in São Paulo (HUWRM 6) and Cachoeira Paulista (HUWRM 2), the fine and coarse fractions are similar in almost all seasons. For the state of São Paulo, the peak volume concentrations are observed in winter and spring, a period with a large presence of fire outbreaks that have a wide range of particle size distribution (MARTIN et al., 2010; RUDKE et al., 2021a; UMO et al., 2015).

The fine particulates present in the atmosphere of eastern US have a large contribution of Sulfate and Nitrate [$(\text{SO}_4^{-2} + \text{NO}_3^-)/\text{PM}_{2.5}$ (%): Boston $\approx 40\%$, New York (NH_4^+ , SO_4^{-2} , and NO_3^-) $\approx 50\%$], non-absorbing fine-mode aerosols, and lower participation of carbonaceous such as black carbon (BC), absorbing fine-mode aerosols [$\text{BC}/\text{PM}_{2.5}$ (%): Boston $\approx 7\%$, New York $\approx 8\text{-}11\%$] (ITO; XUE; THURSTON, 2004; LEE et al., 2014; VENKATACHARI et al., 2006). The fine particulate from the state of São Paulo has variable concentrations, with different sources and chemical compositions but, in general, it also has a large contribution of Sulfate and Nitrate [São Paulo city (HUWRM 6) ($\text{SO}_4^{-2} + \text{NO}_3^- + \text{NH}_4^+$) $\approx 50\%$; Araraquara (HUWRM 13): water-soluble ions accounted for 12% of $\text{PM}_{2.5}$; Cubatão (HUWRM 7): $\text{SO}_4^{-2} + \text{NO}_3^-$ accounted for $\approx 60\%$ of the ions evaluated by Valarini (2011)], but with a greater contribution of carbonaceous [$\text{BC}/\text{PM}_{2.5}$ (%): São Paulo city (HUWRM 6)= 38%, Cubatão (HUWRM 7)=15-20%; Araraquara (HUWRM 13)= organic carbon (OC) accounted for 18% of $\text{PM}_{2.5}$] (GONÇALVES et al., 2017; MIRANDA et al., 2012; PEREIRA et al., 2017b; VALARINI, 2011). Thus, part of the divergences between the types of aerosols found for GSFC

and in São Paulo (Figure C.4) may be related to the greater presence of carbonaceous (elemental and organic carbon) in the atmosphere of São Paulo, since BC is the most absorbing component of the atmospheric aerosol and a primary pollutant (FIERCE et al., 2016). This fact may explain the low correlation between AOD data and particulate matter for the state of São Paulo. In addition, this fact may also explain the higher correlations for 2020, compared to 2019, since the main source of pollutants in São Paulo is vehicular, which has a large presence of carbonaceous in its combustion (BRITO et al., 2013). Thus, with the reduction in mobility due to the COVID-19 outbreak, there was a decrease in the amount of carbonaceous present in the atmosphere.

It is essential to mention that the low correlation between AOD and PM does not necessarily indicate the low accuracy of MAIAC but rather the low ability to represent this relationship directly. Comparisons made with AOD data from MAIAC and AERONET demonstrate good agreement globally (QIN et al., 2021) and over South America (MARTINS et al., 2017b). However, in urban areas (bright surfaces), the MAIAC algorithm presents poorer results than those in vegetated areas (MARTINS et al., 2017b; QIN et al., 2021). For MASP, the comparisons between MAIAC and AERONET showed correlations of 0.74 and 0.64 and root mean square error (RMSE) of 0.087 and 0.079 for Aqua and Terra (Figure C.5), respectively, like those found by Martins et al. (2017) for urban areas. As observed by Martins et al. (2017), the retrievals presented dispersion points scattered in low AOD values and with a slight tendency to underestimate the values (Bias Terra: -0.024 and Bias Aqua: -0.044). The retrieval fraction within the expected error (EE) envelope of Aqua (72.28%) was better than that of Terra (69.49%), suggesting that Aqua product is more suitable for urban retrievals. According to Martins et al. (2017), the multiple pollutants sources contribute to aerosol microphysics, which represents a difficulty for aerosol models used in satellite retrievals.

Finally, it should be noted that different environmental factors can influence the AOD-PM relationship. Among these factors, it is known that the long-range transport of biomass-burning emissions has a substantial impact. Although the burning season in Brazil does not occur during the observed period, it is possible that isolated events were included in the study, generating different impacts for AOD and PM, depending on the height at which emissions are injected into the atmosphere (see Section C2 - Impact of fire emissions on satellite measurements, Appendix C). In addition, as there is variability in the various geographic (e.g., land cover, population density, altitude, traffic density) and meteorological factors (e.g., rainfall,

temperature, PBLH, wind speed, RH), a variable spatial relationship between AOD and MP is expected, requiring a multivariate evaluation. As an example of such an assertive, (LI et al., 2018) found low correlations between AOD and $PM_{2.5}$ (R : 0.19) and were still able to build a model to estimate $PM_{2.5}$ with an R^2 of 0.87 by using mixed-effect models. In this case, the authors used not only MAIAC-AOD to estimate $PM_{2.5}$ but also meteorological parameters, proxy variables for emission sources, Normalized difference vegetation index (NDVI), and Land cover.

7.3.3.2 Satellite estimates – tropospheric NO_2

The correlations between the tropospheric NO_2 vertical column density (TROPOMI) and NO_2 concentration measured by CETESB stations are shown in Figure 7.7 and Table C.7. The results suggest that the two distinct ways of monitoring NO_2 show good correlation in most stations. As with AOD, NO_2 data showed, in general, a higher correlation when considering the smallest spatial window (3km) (Table C.7). Although there was good agreement between values calculated for 2019 and 2020, there was an exception to this behavior for some stations caused by low correlation and disagreements from the different years.

Figure 7.7 - Scatterplot of satellite-based tropospheric NO_2 vertical column density (VCD) derived from TROPOMI (S-5P) and ground-based measurements of NO_2 for the periods: (A) before March 16, 2019; (B) after March 16, 2019; (C) before March 16, 2020; and (D) after March 16, 2020 (mobility restrictions). The blue, orange, and green lines indicate the regression line (Theil–Sen estimator) for NO_2 data extracted at 3, 5, and 10 km. The same colors indicate the correlation coefficient (R), p-value (p), intercept, and slope for each extraction radius.

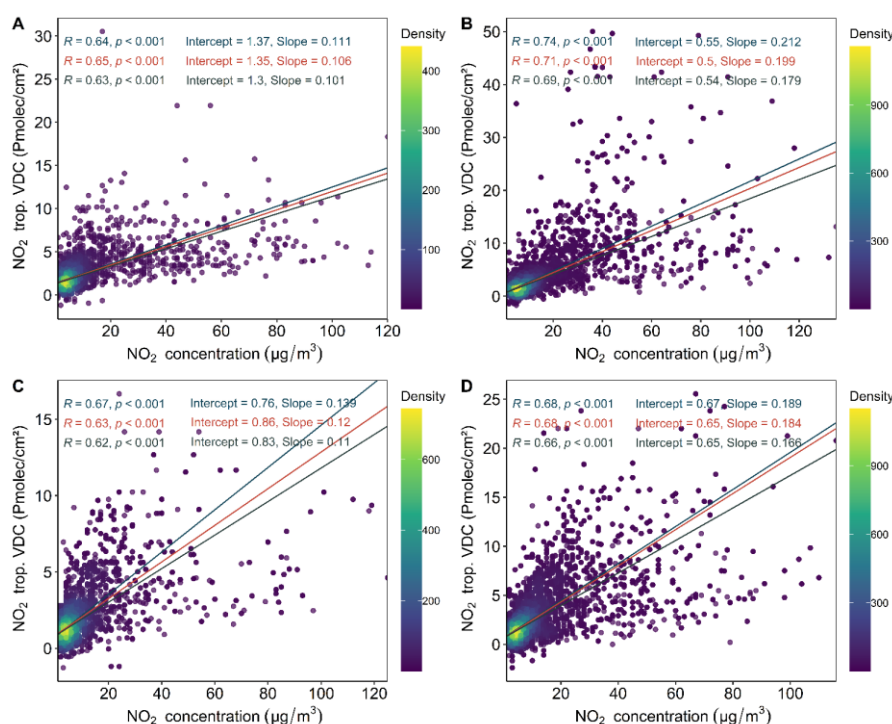


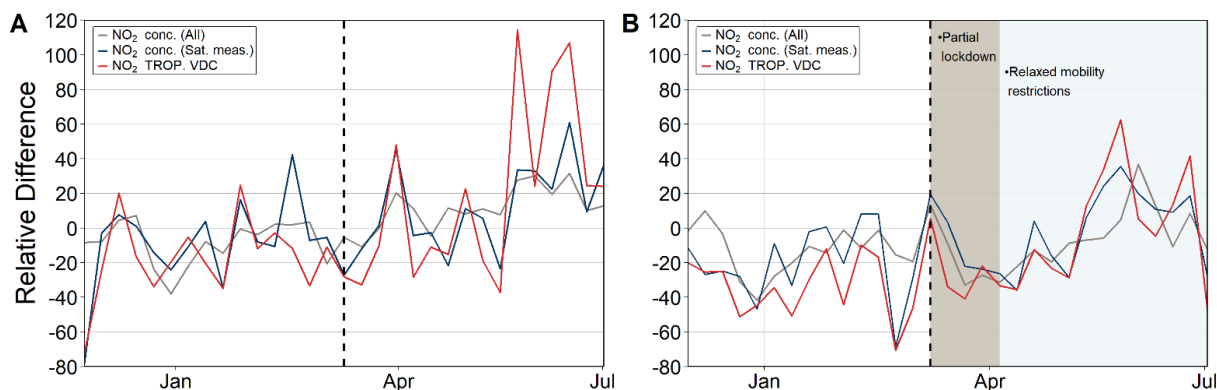
Table 2 presents the averages, standard deviations (SD), and correlation results (3 km spatial window) between the ground based NO₂ and tropospheric NO₂ density. Through Table 2, it is possible to verify that, in general, stations with the highest mean concentrations and the highest standard deviations were the ones with the highest correlations [e.g., S.Bernardo-Centro, Parque D.Pedro II, Ibirapuera, and São Caetano do Sul (HUWRM 6)]. Furthermore, it is observed that, with the decrease in average and standard deviations in 2020, there were also reductions in correlations. The relationship with standard deviations can be verified when observing the 2019 correlations for Marília (HUWRM 21) and Pico do Jaraguá (HUWRM 6) stations, which have an average of around 6 to 7 $\mu\text{g}\cdot\text{m}^{-3}$, but different standard deviation values. Marília presented a smaller data variation when compared to Pico do Jaraguá, showing that the concentrations at Marília are more homogeneous, while those of Pico do Jaraguá station are more heterogeneous. Similarly to Marília, the Araraquara station (HUWRM 10) presented more homogeneous concentrations (2019 and 2020), making the variability in tropospheric NO₂ low and generating lower correlations. The Ibirapuera station had more heterogeneous concentrations, which made the correlations higher. This demonstrates that the sensor has difficulty capturing small variations at the ground level. This fact is also demonstrated through the disaggregation of NO₂ concentrations into class intervals (Figure C.6), since there is a clear distinction between values up to 20 $\mu\text{g}\cdot\text{m}^{-3}$ and higher, but a great overlap between the other categories. This fact demonstrates that good correlations will be obtained when there is a greater variation range in NO₂ concentrations.

Table 7.2 – Mean, standard deviation (SD), and correlation between tropospheric NO₂ density (TROPOMI) (Pmolec/cm²) and NO₂ concentration measured by CETESB stations (µg·m⁻³). The non-significant correlations (p-value > 0.05) are shown in bold. Data represents only the smallest extraction radius (3 km).

HUWRM	Station name	TROPOMI-2019		Air Quality Station-2019		R	TROPOMI-2020		Air Quality Station-2020		R
		mean	SD	mean	SD		mean	SD	mean	SD	
2	Guaratinguetá	1.82	0.80	5.0	3.4	0.19	1.08	0.68	4.3	2.0	0.11
	S.José Campos	3.13	1.73	11.8	6.5	0.5	1.13	0.68	4.8	2.3	0.36
	S.José Campos-Jd.Satelite	3.23	1.78	10.0	6.1	0.46	2.65	1.43	3.5	2.6	0.18
	Taubaté	2.03	1.03	8.1	6.3	0.5	1.23	0.75	4.6	2.9	0.22
5	Campinas-Taquaral	3.15	1.83	6.0	3.5	0.44	6.45	4.40	22.6	14.5	0.58
	Piracicaba	2.24	1.34	6.1	3.3	0.38	6.35	3.91	52.2	21.3	0.23
	Santa Gertrudes	1.85	0.99	13.3	6.4	0.38	6.65	5.13	18.1	11.5	0.81
6	Cerqueira César	7.76	4.23	36.0	16.0	0.52	6.15	3.63	47.1	20.1	0.56
	Congonhas	8.39	4.30	64.6	19.1	0.68	1.39	0.91	3.9	2.8	0.46
	Guarulhos-Pimentas	5.02	6.92	9.3	8.2	0.45	4.01	2.92	9.8	4.9	0.47
	Ibirapuera	9.91	7.82	15.3	10.2	0.81	7.10	3.88	15.0	11.0	0.7
	Itaim Paulista	6.08	3.85	12.1	6.2	0.63	4.56	2.33	10.5	4.9	0.71
	Marg.Tietê-Pte Remédios	6.15	3.88	69.9	24.6	0.4	0.96	0.80	5.6	2.1	-
	Osasco	6.11	4.55	37.3	16.8	0.59	5.62	3.77	56.0	25.9	0.41
	Parque D.Pedro II	8.73	4.55	25.4	10.6	0.86	0.92	0.87	4.2	2.2	0.27
	Pico do Jaraguá	4.87	5.03	6.9	7.0	0.71	5.24	3.93	31.6	16.3	0.81
	Pinheiros	7.29	3.14	33.0	12.4	0.56	6.53	4.35	19.4	10.7	0.71
	S.André-Capuava	12.03	10.55	21.7	9.8	0.63	4.17	2.46	7.5	6.4	0.74
	S.Bernardo-Centro	8.57	7.47	20.4	10.1	0.86	6.18	4.14	20.3	11.1	0.73
	São Caetano do Sul	11.09	10.80	22.3	7.9	0.74	1.85	0.94	6.0	3.4	0.31
	Cubatão-Centro	11.82	10.03	26.7	18.1	0.73	1.03	0.64	3.4	1.9	0.24
	Cubatão-V.Parisi	11.51	9.25	54.1	23.2	0.38	5.49	2.77	15.0	7.5	0.59
7	Santos-Ponta da Praia	6.85	9.35	14.9	12.0	0.66	4.90	2.46	14.9	5.4	0.64
	Sorocaba	2.90	1.86	9.4	3.6	0.71	2.99	2.66	9.6	5.7	0.41
10	Tatuí	1.52	0.90	6.3	3.7	0.28	3.08	1.72	9.5	6.3	0.56
13	Araraquara	1.39	0.71	5.5	2.4	0.3	1.62	1.15	18.6	6.1	0.45
	Bauru	1.33	0.84	5.0	2.6	-0.04	3.97	4.32	10.3	10.3	0.67
	Jaú	1.18	0.81	4.6	2.0	-0.04	6.06	2.84	17.3	6.9	0.56
15	Catanduva	1.38	0.82	4.4	2.4	0.26	1.32	0.85	6.1	3.6	0.28
	São José do Rio Preto	1.43	0.77	5.6	2.9	0.06	2.27	1.68	8.8	4.0	0.54
21	Marília	1.16	0.73	6.3	3.0	-0.04	1.18	0.85	4.9	2.6	0.44
22	Presidente Prudente	1.09	0.73	3.6	1.7	-0.18	1.43	0.80	5.3	3.2	0.38

Temporally, the tropospheric NO₂ vertical column density showed similar behavior to that observed for the NO₂ measured at ground level (Figure 7.8). It is observed that, for 2020, the behavior of concentrations at ground level is better represented by tropospheric NO₂ vertical column density. In addition, looking at the weekly NO₂ concentrations for all the data available in the satellite overpass, even without data for the satellite (grey line in Figure 7.8), it is clear how the lack of data affects the average calculations.

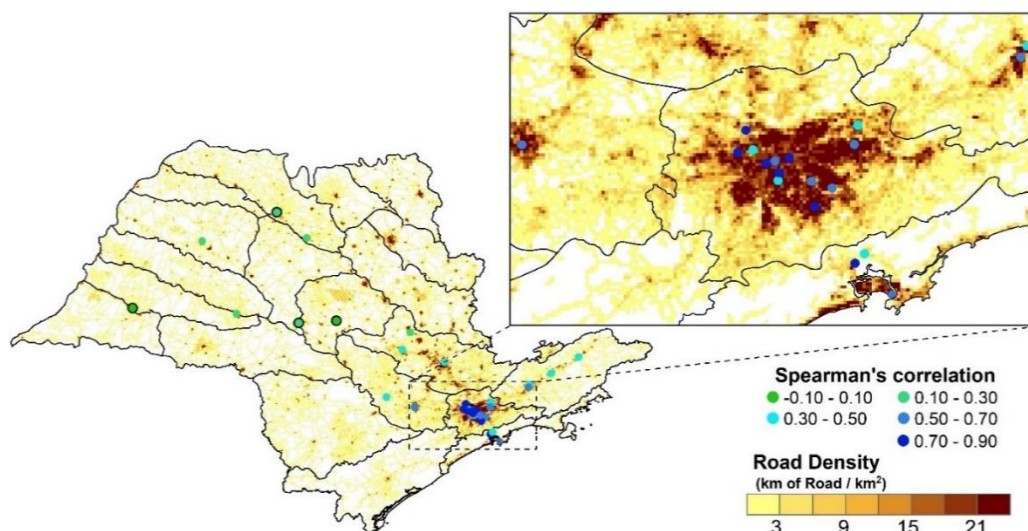
Figure 7.8 - Weekly dynamics of tropospheric NO₂ VCD derived from TROPOMI (S-5P) and ground-based measurements of NO₂ for 2019 (A) and 2020 (B). The vertical dashed black line indicates the 76th day of 2019 and 2020, when the first restrictions started in 2020. The averages represent data obtained through the smallest extraction radius (3 km). The relative difference was calculated using variable averages for 2019 as a comparison. The blue lines indicate weekly concentrations of NO₂ that have matching satellite data. The red lines indicate the weekly concentrations of NO₂ for all data available in the satellite overpass, even without data for the satellite.



Spatially, the highest correlations were observed for HUWRM 2 (R: 0.57), 6 (R: 0.51), 7 (R: 0.68), and 10 (R: 0.55), all with characteristics of industrial economic vocation (Table C.8). Although it has an industrial economic vocation, HUWRM 5 was the only one in this category to show low correlations (R: 0.14). For this region, low correlations were observed especially for the Campinas-Taquaral and Piracicaba stations, which did not present significant ($\alpha > 0.05$) correlations. Both stations are in the transitions between urban and rural areas (peri-urban stations, Table C.7), which presented the worst correlation because they are far from urban centers and, therefore, receive less localized pollution than stations close to major traffic routes. Only HUWRM 13 and 22 did not present statistically significant correlations ($\alpha > 0.05$). HUWRM 22 has an economic vocation for Farming and Livestock, with a smaller contribution from the pollutant when compared to other regions (RUDKE et al., 2021a). The HUWRM 13 has an economic vocation characterized as "in the process of industrialization", but it presents NO₂ concentrations similar to those observed for regions with Farming and Livestock economic vocation (Table 2), which may be related to the type of industry present in the local, mainly agro-industrial (GONÇALVES et al., 2017). Figure 7.9 shows the distribution of correlations obtained by aggregating all the data evaluated (2019 and 2020) and may shed some light on the observed behavior. The figure shows that the highest correlations were obtained in stations located in intensely inhabited regions (MASP), which present a greater flow of vehicles. In addition, a good correlation was obtained close to the industrial hub of Cubatão (UGRHI 7). This region showed increases in the concentrations of several pollutants, even during the mobility restrictions imposed by the COVID-19 outbreak (RUDKE et al., 2021a). In these

regions, due to the greater smudge of pollution, NO₂ can be constricted and become trapped within the Planetary Boundary Layer, as mentioned by Cersosimo et al. (2020).

Figure 7.9 - Spatial distribution of correlations between tropospheric NO₂ VCD and ground-based measurements obtained for the first semester of 2019 and 2020. Dots with a black border indicate non-significant correlations ($\alpha > 0.05$).



Overall, the tropospheric NO₂ data obtained through the TROPOMI have shown good correlations with the ground-based NO₂ acquired from air quality monitoring stations (BASSANI et al., 2021; CERSOSIMO; SERIO; MASIELLO, 2020; FIOLETOV et al., 2022; GOLDBERG et al., 2021; KIM; BRUNNER; KUHLMANN, 2021; STRATOULIAS; NUTHAMMACHOT, 2020; VÎRGHILEANU et al., 2020; VIRTÀ et al., 2021; ZHENG et al., 2019). However, some studies also found lower correlations between these measurements for rural areas (BASSANI et al., 2021; JEONG; HONG, 2021; VIRTÀ et al., 2021). These studies indicate that, in more remote regions, due to their very low NO₂ concentrations, the minimum detectable limit of the sensor may be reached (BASSANI et al., 2021), generating unrealistic data. Assessing tropospheric NO₂ variations over remote locations is a challenge, since these areas are generally far away from the primary sources of NO₂ and thus present concentrations much lower than those over urban areas. In São Paulo, the stations with the lowest correlation located in regions with a rural vocation also showed lower NO₂ levels (mean: 5.1 $\mu\text{g}\cdot\text{m}^{-3}$, SD: 2.7 $\mu\text{g}\cdot\text{m}^{-3}$, and maximum: 16 $\mu\text{g}\cdot\text{m}^{-3}$), when compared to the urban stations with the highest correlations (mean: 15.5 $\mu\text{g}\cdot\text{m}^{-3}$, SD: 10.4 $\mu\text{g}\cdot\text{m}^{-3}$, and maximum: 108 $\mu\text{g}\cdot\text{m}^{-3}$). From comparisons to ground-based total and tropospheric column measurements, validation studies demonstrate that TROPOMI tends to overestimate data at rural and remote locations due to lower concentrations of NO₂ (IALONGO et al., 2020; MARAIS et al., 2021; VERHOELST et al., 2021; ZHAO et al., 2020). Due to the much lower NO₂ levels over rural areas, the estimates

of tropospheric NO₂ may not follow the slight daily variations observed by the ground-based stations. This behavior may be related to a possible overestimation of the stratospheric column fraction. An example of such assertion is the Bauru region (HUWRM 13), which exhibited non-significant correlations in our estimates and showed a negative Bias between stratospheric NO₂ measured by the monitoring station (Zenith-Scattered-Light DOAS, ZSL-DOAS) and by TROPOMI (-1.2695 ± 0.00726 Pmolec/cm², data available at: <https://mpc-vdaf-server.tropomi.eu/no2/no2-offl-zsl-doas-sunset/bauru#Baseline>) (VERHOELST et al., 2021). In this case, the underestimation of stratospheric NO₂ is possibly followed by an overestimation of the tropospheric column, since separating these columns in areas with low NO₂ levels remains challenging. (VAN GEFFEN et al., 2019) indicate that lower NO₂ levels can lead to greater difficulty in separating the stratospheric and tropospheric NO₂ columns, due to uncertainties in the NO₂ profile. In addition to these uncertainties, a more significant impact of the instrument noise is expected to affect the estimates over rural areas, as NO₂ levels are much lower.

Thus, the results found for the state of São Paulo demonstrate that the TROPOMI sensor is promising for mapping the variability of tropospheric NO₂ in densely populated regions, which present a source of expressive emission of the pollutant. However, this accuracy was not maintained for less populated areas, with economic characteristics focused on agriculture and agribusiness. It is also worth noting that the measurements of air quality stations (punctual data) were compared with 5.5×3.5 or 7.0×3.5 km² areas (size of TROPOMI pixel), which can reduce the correlations in locations that have very localized sources, once this effect will be diluted along the pixel on the satellite.

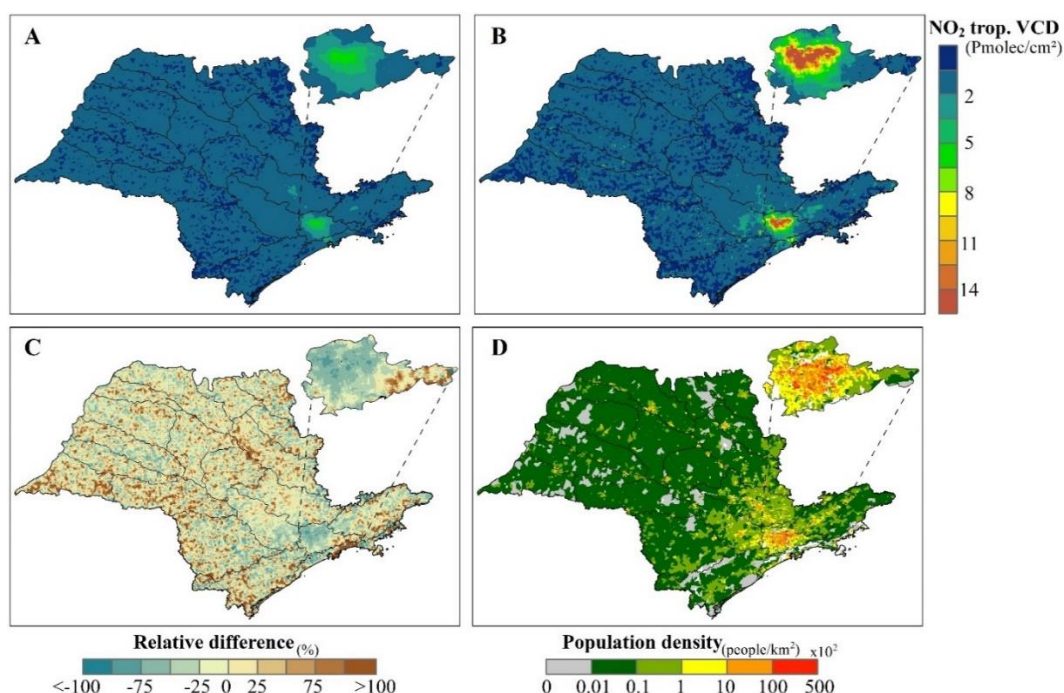
Finally, the findings of this study demonstrate the potential of the TROPOMI data and indicate the need for further studies regarding the relationship between AOD and PM. In the second case, studies that seek to identify PM_{2.5} from the satellite are important as a function of its high potential for damage to human health, especially in regions with a scarce monitoring network, such as Brazil. As NO_x is an important precursor of secondary PM_{2.5}, high correlations between NO₂ and PM_{2.5} are expected in places where pollutant sources are strongly associated (WU et al., 2016). In the State of São Paulo, low to moderate correlations were observed (R:0.34-0.44), depending on the period analyzed (Figure C.7). This behavior was also maintained when comparing concentrations of PM_{2.5} with tropospheric NO₂ densities from the TROPOMI sensor (R:0.28 to 0.38). This demonstrates that TROPOMI data can also capture this relationship

between NO_2 and $\text{PM}_{2.5}$ at ground level. In addition, the correlations between concentrations of $\text{PM}_{2.5}$ and tropospheric NO_2 densities were greater than those observed for AOD data.

7.3.4 Spatiotemporal Distribution of the Air Pollution Monitored by Satellite

To demonstrate how mobility restrictions during the COVID-19 outbreak have affected air quality over the state of São Paulo, the spatial pattern of reduction/increase in tropospheric NO_2 was evaluated. In this section, only tropospheric NO_2 was considered, since these data presented better correlations with the data measured at ground level. Figure 7.10 shows the average for the 30 days after the onset of mobility restrictions, considering tropospheric NO_2 observed by satellite and estimated by Eq. 7.1. Satellite data considered for the estimates used averages of 30 days before and after the beginning of the restrictions adopted in 2020 in comparison to the same period of 2019 (this dataset can be seen in the Appendix C - Figure C.8). As mentioned in section 2.4, using Eq. 7.1 to estimate tropospheric NO_2 for 2020 assumes that the meteorological scenario has a similar average impact as 2019. On the other hand, slightly different atmospheric conditions were observed, with rainfall having less effect on wet deposition and a higher number of fire outbreaks during the mobility restrictions. With the prevailing atmospheric conditions in 2020, air pollutant concentrations were expected to be higher than those observed for the same period in 2019. Thus, considering this scenario, it is possible to infer that the values estimated by Eq. 7.1 are possibly underestimated; the density of tropospheric NO_2 expected for 2020 would be undoubtedly higher than those shown in Figure 7.10-B. This scenario observed for 2020 also prevented higher air quality improvements during mobility restrictions, as mentioned by (RUDKE et al., 2021a, 2022).

Figure 7.10 - Change in air quality over the state of São Paulo for 30 days after the beginning of mobility restrictions (March 16, 2020). Observed values for tropospheric NO₂ VCD (A), expected values estimated by Eq.7.1 (B), relative difference between A and B (C), and population density (D) for the state of São Paulo.



The satellite data presented in Figure 7.10 shows that the areas with the highest amount of pollutants are closely related to the places with the highest population density, as shown in (RUDKE et al., 2021a, 2022). This is essentially due to the larger vehicle fleet associated with population size and the industrial hub already established in these regions. In addition, it is observed that there is no homogeneous behavior in the state, with increases registered in the interior, especially in places with an economic vocation for Farming and Livestock, and reductions observed mainly in Industrial areas or in the process of industrialization. Assessing the entire state of São Paulo, 57% of the area showed some level of reduction in Tropospheric NO₂. Reductions in industrial areas or in the process of industrialization reflect the decrease in economic activities and the adoption of mobility control practices in the 30 days. The reduction in mobility is the main driver of the decrease in MASP (HUWRM 6). This area has the highest density of vehicles per inhabitant in Brazil and many other places in the world. For MASP, the results presented in Figure 7.10 show a substantial reduction in Tropospheric NO₂, in which the average drops reached 51%.

Unlike industrial or in industrialization areas, there is an increase in Tropospheric NO₂ in several cities in the interior of the State, where the main economic vocation is Farming and Livestock. The most substantial increases were observed for HUWRMs 9 (16.6%), 17 (15.3%),

and 22 (87.0%). The increases in Tropospheric NO₂ occurred in areas with lower concentrations of pollutants (Figure 7.10); thus, the rises may be related to seasonal pollutants variability or lower data quality for these areas, as shown in the previous section.

Studies developed based on data acquired through air quality monitoring stations demonstrate similar behavior for MASP, with greater or lesser reductions according to the evaluated period (CONNERTON et al., 2020; DEBONE; DA COSTA; MIRAGLIA, 2020; NAKADA; URBAN, 2020; NODA et al., 2021; RUDKE et al., 2021a; SICILIANO et al., 2020a; TADANO et al., 2021). Regarding the interior of the state, Rudke et al. (2021) demonstrated great spatial variability in the concentrations of pollutants when analyzing one semester. However, the study shows that, even in interior cities, most of the monitored stations showed significant reductions in NO₂ concentrations compared to the same period in 2019. For particulate matter, an inverse behavior was observed; most stations located in the interior did not show significant variation and, when significant, they showed an increasing tendency. It is important to point out that air quality monitoring stations in the state are located, in general, in medium or large urban areas. Increases in Tropospheric NO₂ values (Figure 7.10), on the other hand, were mainly observed in areas with lower population density.

It is important to consider that, by October 2020, one-third of the research published on the effects of mobility restriction policies (adopted during the COVID-19 outbreak) on air quality used data monitored by satellite (GKATZELIS et al., 2021). Although this type of information is very relevant for spatial assessments, especially in places with little or no ground-level monitoring network, some considerations should be made regarding its monitoring capacity.

As for the monitoring carried out through remote sensing, one of the main problems in data retrieval by passive sensors is related to the presence of clouds (CERSOSIMO; SERIO; MASIELLO, 2020; SANO et al., 2007). Furthermore, regarding the properties of atmospheric aerosols, an additional challenge concerns the distinction between scattered light by aerosol particles and by atmospheric gases and some types of complex surfaces (e.g., snow/ice, desert, etc.) (DUBOVİK et al., 2021; MHAWISH et al., 2019). For NO₂ retrievals, another issue is related to the presence of aerosols in the atmosphere, which significantly impacts scattering properties (VASILKOV et al., 2020). This type of interference, along with errors and problematic retrievals, substantially reduces the availability of continuous and uniform sampling and coverage for a given region.

Figure C.9 shows the availability of Tropospheric NO₂ data for the state of São Paulo, considering data observed for the 30 days before and after the beginning of the restrictions adopted in 2020, compared to the same period for 2019. The percentage of valid days during the 30 days before the mobility restrictions is approximately 35% for São Paulo [average quantity of data for the state was 8.9 (min 0 – max 23) for 2019 and 12.9 (min 0 – max 28) for 2020]. During the restrictions, this value increases to approximately 50% [2019: 12.4 (min 1 – max 24); 2020: 17.5 (min 2 – max 28)]. This occurs due to the greater presence of clouds during the rainy season (October to March), with increased cloudiness in early spring and maximum peak occurring in summer (MOURA; MARTINS; ASSIREU, 2016).

Locally, data availability can be even more affected. For example, for MASP, the availability of valid days was less than 30% before and approximately 40% during the restrictions (data available for the period before the restrictions: 2019 – 7.3 and 2020 - 6.4, during the restrictions: 2019 – 8.3 and 2020 – 14.7). This fact is due to the high cloudiness present over the region in all year periods compared to other areas of the state (Figure C.10). In the specific case of MASP, few data were used in the comparison presented in Figure 7.10; this fact shows that unrealistic results can be obtained when satellite data is used to track short-term changes, once days with low or high concentrations of pollutants can be considered on the statistics and thus generate under- or overestimated averages. (PRUNET et al., 2020) also observed low availability of valid days for Paris (approximately 40%) due to the presence of clouds. The authors also indicate that the lack of valid data for some weeks analyzed for Milan may have contributed to artificially reducing the 2020 averages, probably intensifying the reductions observed during the lockdown period. Therefore, considering the uncertainties related to the lack of data associated with constant presence of clouds worldwide (Figure C.11), the findings must also be associated with a more careful analysis. For instance, the review of Gkatzelis et al. (2021) distinguishes studies based on observations alone and studies that involve models that can account for the weather impacts on the data. Besides, one way to improve satellite data availability is associated with using mathematical and/or statistical methods, such as interpolation and regression methods (CERSOSIMO; SERIO; MASIELLO, 2020; WU et al., 2021; ZHENG et al., 2019).

7.4 Conclusions

This study examined the capability of satellite products, tropospheric NO₂ column density from the TROPOMI sensor and MAIAC-AOD from MODIS sensors, as predictors of NO₂ and

particulate matter (PM₁₀ and PM_{2.5}) concentrations at ground level for the state of São Paulo during the period of vehicle circulation restrictions as a function of the COVID-19 pandemic. This approach was performed by calculating the correlation between ground-based air quality data and satellite data at spatial windows of 3, 5, and 10 km, and at a temporal window of $\pm 1h$. The pandemic outbreak period was chosen due to the recurrent use of these products to describe the trend in pollutant concentrations, especially in large urban areas. The main findings of this study were:

- 1) For the State of São Paulo, the distribution of changes in pollutants measured by satellite (relative difference between observed and expected data) presented a very heterogeneous behavior. Overall, about 57% of the state of São Paulo showed some reduction in Tropospheric NO₂ density values. The regions with the greatest reductions were those with industrial economic vocation, such as MASP. On the other hand, increases in tropospheric NO₂ were observed in regions with an economic vocation for Farming and Livestock, of up to 87%.
- 2) The assessment of temporal data availability demonstrated that, due to factors that make data acquisition difficult by passive sensors - especially clouds, there is a large presence of failures in the data series. Considering the satellite data available for thirty days, the analyses show that retrievals are available less than 20% of the time, depending on the region of the state. This fact can provide misleading assessments of the real variability of pollutants in the lower atmosphere when satellite information is used as ground truth. This misleading can occur when few data from days with high or low concentrations of pollutants are used, generating over- or underestimated values, respectively.
- 3) Comparisons between AOD data and particulate matter (PM₁₀ and PM_{2.5}) showed that AOD has a low potential to explain the spatiotemporal behavior of PM in the study area. Correlations between data were low and frequently not significant, demonstrating the weak direct relationship between data. Furthermore, although the performance of the MAIAC AOD retrievals varied in time and space, PM_{2.5} data were better represented by the AOD in some specific stations, reaching correlations of up to 0.61 in a few stations. The low correlations between AOD and PM data observed for most stations are corroborated by previous studies, and they may be related to the type of aerosol present in the atmosphere of the state of São Paulo, which is very different from other regions where the correlations between these data proved to be promising.

4) Tropospheric NO₂ data demonstrated the potential to represent changes in NO₂ concentrations at ground level. The correlations between Tropospheric NO₂ column density and NO₂ concentrations at ground level were greater than 0.60. The highest correlations were observed especially in industrialized regions, which are significant sources of NO₂ and NO. On the other hand, low correlations were observed for regions with an economic vocation for Farming and Livestock. These regions had low tropospheric NO₂ densities due to the low number of sources present in the area, making the correlations generally low and not significant.

Finally, based on the findings of this research, it is possible to conclude that the Tropospheric NO₂ column density product from the TROPOMI sensor is promising for evaluating NO₂ concentration in the lower atmosphere of the state of São Paulo. On the other hand, the MODIS-AOD data retrieved through the MAIAC algorithm showed low potential for directly predicting MP concentrations at ground level. Thus, our findings demonstrate that, although MAIAC-AOD has shown relatively good results as a PM predictor in some areas around the world, its use may be limited to locations with less uncertainty regarding atmospheric parameters and other assumptions and approximations used in AOD retrieval.

8 EVALUATION AND COMPARISON OF MODIS AEROSOL OPTICAL DEPTH RETRIEVAL ALGORITHMS OVER BRAZIL

Abstract: Brazil experiences significant aerosol loads throughout the year, particularly during the biomass burning season in the Amazon (August to November). Therefore, it is of utmost importance to accurately estimate and understand the geographical distribution of aerosols due to their significant impact on the Earth's energy budget and human health. Aerosol Optical Depth (AOD) serves as a primary indicator for mapping aerosol distribution and load, with data primarily derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor. Thus, given the importance of aerosols, this research aimed to validate and compare AOD products obtained using three reference algorithms: Dark Target (DT) at 3 and 10 km resolution, Deep Blue (DB), and Multi-Angle Implementation of Atmospheric Correction (MAIAC). For this purpose, data from 17 sunphotometers of the AErosol RObotic NETwork (AERONET) were utilized. Overall, the results demonstrated a strong correlation ($R > 0.9$) between all AOD products retrieved from MODIS and ground-based data. However, it is essential to note that the AOD products obtained through these algorithms tend to overestimate or underestimate AOD values, depending on the specific AOD value and algorithm evaluated. Additionally, the performance of the algorithms is influenced by factors such as land cover type, view geometry, and the spatiotemporal distribution of aerosols. In particular, challenges were encountered when retrieving robust AOD data for Savanna and Urban cover classes. In conclusion, the results indicate that MAIAC and DB algorithms demonstrate greater stability in retrieving AOD values. Nevertheless, caution should be exercised when applying these products to map aerosols on specific surfaces, such as urban areas.

Keywords: AOD; MODIS; Biomass burning emissions; view geometry dependence; Land cover type dependency.

8.1 Introduction

Aerosols, which are tiny particles suspended in the air (solid or liquid), have a significant impact on the planet's climate and human health (KOK et al., 2023; LIU et al., 2019b; MARTINS et al., 2018; PÖSCHL, 2005; SEINFELD; PANDIS, 2016). Therefore, the accurate mapping and monitoring of aerosols is crucial for understanding their behavior and effects (KOK et al., 2023; LIU; MAUZERALL; HOROWITZ, 2009; MHAWISH et al., 2019; PU; YOO, 2021). Aerosol Optical Depth (AOD) measurement is a approach that has proven particularly useful in this regard (FILONCHYK et al., 2019). AOD measures the extent to which sunlight is prevented

from transmitting through the atmosphere due to the absorption or scattering of aerosol particles (FILONCHYK et al., 2019). It can be measured using satellite or ground-based instruments. By analyzing the spatiotemporal distribution of AOD, it is possible to infer aerosols' distribution, concentration, and transport, helping the understanding of the complex interactions between aerosols, climate, and human well-being (KOK et al., 2023; LIU; MAUZERALL; HOROWITZ, 2009; LIU et al., 2019b; PALVE; NEMADE; GHUDE, 2016; PÖSCHL, 2005; REMER et al., 2013b).

Satellite data has become an essential tool for monitoring and mapping AOD on a global scale (REMER et al., 2013b). In this context, Moderate Resolution Imaging Spectroradiometer (MODIS) is one satellite-based instrument that has been widely used for AOD retrievals (LEVY et al., 2010, 2013; LYAPUSTIN et al., 2011, 2018; REMER et al., 2013a). With a spatial resolution of 10 to 1 kilometer, MODIS can observe the entire globe in just one or two days, making it an ideal instrument for studying the impact of aerosols on regional and global climate patterns (EDDY, 2012; PALVE; NEMADE; GHUDE, 2016; RAMANATHAN et al., 2001). The data collected by MODIS has been used in a range of applications, including air quality forecasting, wildfire smoke monitoring, and health studies (DI et al., 2016; KLOOG et al., 2014; LEE et al., 2011, 2012; LI et al., 2021; MARTINS et al., 2018; TARGINO et al., 2019). Overall, MODIS-AOD data has contributed significantly to understanding the complex interactions between aerosols and the environment.

Although AOD retrieved from satellite sensors such as MODIS has revolutionized the ability to study aerosols on a global scale, it is essential to note that the quality of the data can vary depending on various factors, such as cloud cover, instrument calibration, surface reflectivity, and aerosol type (CHEN et al., 2021; HE et al., 2017b; LIU et al., 2019a; MARTINS et al., 2017b; MHAWISH et al., 2019; NICHOL; BILAL, 2016; TIAN; GAO, 2019; WEI et al., 2019). As such, it is crucial to evaluate the quality of the AOD data obtained from satellite-based instruments before using it for local assessments. By comparing the satellite AOD data with ground-based measurements, it is possible to identify and correct any biases or errors in the satellite data, ensuring that the data is of high quality and suitable for local assessments (KLIEWER et al., 2018; LEMMOUCHI et al., 2023; ZHONG et al., 2022). This is especially important when using the AOD data for health studies or air quality forecasting, where accuracy is critical for making informed decisions (HE; WANG; YIM, 2021; LI et al., 2018a, 2018c, 2018b; PU; YOO, 2021; XIAO et al., 2017). In summary, while satellite data such as MODIS

can provide a comprehensive view of aerosol distribution and behavior on a global scale, it is crucial to evaluate the data's quality to ensure that it is suitable for local assessments.

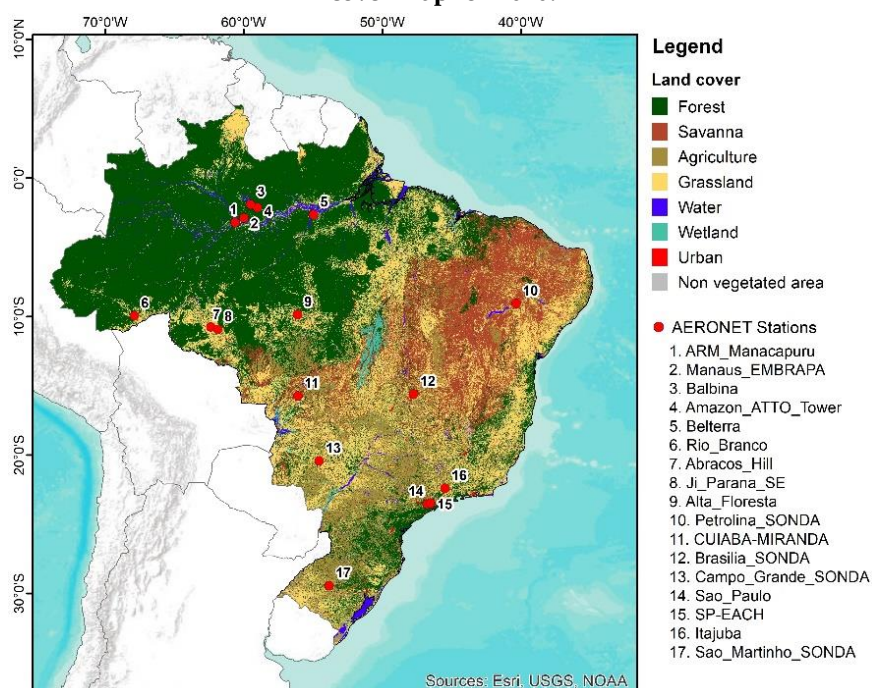
In this context, this study represents the first in-depth validation and comparison of the AOD products retrieved by using three different algorithms [Dark Target (DT) at 3 and 10 km resolution, Deep Blue (DB), and Multi-Angle Implementation of Atmospheric Correction (MAIAC)] over Brazil. This research proposes to examine various factors that may impact product quality, such as: spatiotemporal patterns, spatiotemporal completeness, land cover class and view geometry dependencies, and predominant aerosol type.

8.2 Methodology

8.2.1 Description of the Study Area

Brazil is a country of continental proportions, with a diverse range of ecosystems and a variety of land cover types (Figure 8.1). These land cover characteristics play a crucial role in the distribution and concentrations of aerosols, which can have significant impacts on air quality, climate, and human health. Despite this, few studies have investigated the relationship between land cover and aerosol loads in Brazil. The study area will focus on various regions across Brazil, including the Amazon rainforest, Cerrado (known as Brazilian savanna), and the Atlantic Forest, to assess how land cover characteristics affect the aerosol loads.

Figure 8.1 – Location of the AERONET sites in Brazil. The background image shows Mapbiomas land cover map for 2020.



8.2.2 Data Sets

8.2.2.1 AOD – Satellite data

AOD data were obtained through images from the MODIS sensors on board the Aqua (EOS/PM-1) and Terra (EOS/AM-1) satellites, available free of charge on the National Aeronautics and Space Administration (NASA) website, <https://ladsweb.modaps.eosdis.nasa.gov>). With distinct orbits, Terra satellite has a daily pass over Ecuador at around 10:30 am (local time) and Aqua at around 1:30 pm in the opposite direction, providing almost daily AOD coverage across the globe. (LIN et al., 2015).

Over the last few years, the AOD retrieval algorithms estimated through MODIS sensor data have been updated. Currently, three operational algorithms are in use: Dark Target (DT), Deep Blue (DB), and Multi-Angle Implementation of Atmospheric Correction (MAIAC). This research uses MODIS AOD products from collection 6.1 (C6.1) for all aerosol algorithms. Information about the satellite and AOD product used in this study is listed in Table 8.1.

Table 8.1 – Overview of Aerosol Optical Depth (AOD) data sets utilized in the study.

Satellite	Product	Algorithm	Spatial resolution	Start date
Aqua/Terra	MCD19A2	MAIAC	1 km	01/02/2000
Aqua	MYD04_3K	DT	3 km	01/07/2002
Terra	MOD04_3K	DT	3 km	01/02/2000
Aqua	MYD04_L2	DT and DB	10 km	01/07/2002
Terra	MOD04_L2	DT and DB	10 km	01/02/2000

Both products have a temporal resolution of 1 day. The DT algorithm has products in two spatial resolutions (3 and 10 km). Although the 3 km product is based on the same algorithm and uses the same Look Up Tables as the standard Dark Target aerosol product retrievals, some differences include the pixel arrays used for each retrieval box and the minimum percentage of "good" pixels required for a retrieval. In addition, the 10 km algorithm also attempts a "poor quality" retrieval when the number of available pixels after screening is small, while the 3 km algorithm will not attempt a retrieval under these circumstances. The 10 km product proved to be more accurate in some regions (HE et al., 2017b; NICHOL; BILAL, 2016; REMER et al., 2013a); higher uncertainties of the 3 km products were observed in urban areas, especially those dominated by coarse fraction aerosols (NICHOL; BILAL, 2016). In this way, both products were evaluated.

The presence of snow, clouds, and highly reflective surface types (e.g., desert) will increase the uncertainty of AOD data retrievals. Thus, all three products provide information about Quality

Assurance (QA) (HUBANKS, 2017). In this sense, only AOD data with the best retrieval quality were used [MAIAC: highest quality, DT: QA = 3 and DB: best estimate layer (“Deep_Blue_Aerosol_Optical_Depth_550_Land_Best_Estimate”)].

8.2.2.2 AOD – Ground-level data

For validation of MODIS-AOD products, AOD data measured on surface by AErosol RObotic NETwork (AERONET) were used. The AERONET network provides globally distributed AOD observations. The data set is made available by the NASA website (https://aeronet.gsfc.nasa.gov/new_web/index.html). For the evaluation in Brazil, 17 monitoring sites were used (Table 8.2).

Table 8.2 – AERONET sites used in this study.

AERONET sites	Latitude	Longitude	Elevation (m)	Period	Data availability (days)
Abracos Hill	-10.76	-62.36	259	2000-2005	848
Alta Floresta	-9.87	-56.10	277	2000-2020	3722
Amazon ATTO Tower	-2.14	-59.00	210	2016-2020	824
ARM Manacapuru	-3.21	-60.60	50	2013-2015	247
Balbina	-1.92	-59.49	80	2000-2002	432
Belterra	-2.65	-54.95	70	2000-2005	777
Brasília-SONDA	-15.60	-47.71	992	2015-2018	245
Campo Grande-SONDA	-20.44	-54.54	677	2003-2019	2148
Cuiabá-MIRANDA	-15.73	-56.07	210	2001-2020	3447
Itajuba	-22.41	-45.45	856	2013-2020	981
Ji-Parana-SE	-10.93	-61.85	218	2006-2020	2373
Manaus-EMBRAPA	-2.89	-59.97	115	2011-2019	1008
Petrolina-SONDA	-9.07	-40.33	381	2004-2016	1783
Rio Branco	-9.97	-67.87	212	2000-2020	3240
São Martinho-SONDA	-29.44	-53.82	489	2008-2014	851
São Paulo	-23.56	-46.73	786	2000-2020	2779
SP-EACH	-23.48	-46.50	754	2016-2020	580

^a The ID number in parentheses is a reference to site location in Figure 8.1.

The most current version of AERONET data (version 3) has three quality levels: Level 1.0 (unscreened), raw data; Level 1.5 (cloud-screened and quality controlled), in which the measurements undergo processing to eliminate cloudiness, and; Level 2.0 (quality-assured), in which the data undergoes pre- and post-calibration when the photometer returns to NASA. In this research, only data with Level 2.0 quality were used for validation of satellite data, as well as in previous research (LIU et al., 2019; MARTINS et al., 2017b; MHAWISH et al., 2019).

As the AOD measurements retrieved by satellite are at a wavelength of 550 nm, the AERONET data were interpolated to 550 nm using the equation (EIBEDINGIL et al., 2021; FALAH et al., 2021):

$$\tau_{550\text{nm}} = \frac{\tau_{500\text{nm}}}{\exp\left(-\alpha * \ln\left(\frac{\lambda_{500\text{nm}}}{\lambda_{550\text{nm}}}\right)\right)} \quad (8.1)$$

Where, $\tau_{550\text{nm}}$, $\tau_{500\text{nm}}$ denote AOD values at wavelengths $\lambda_{550\text{nm}}$, $\lambda_{500\text{nm}}$, respectively; and α corresponds to the Angstrom exponent at 440-675 nm ($\alpha_{440-675}$).

8.2.2.3 Land cover data

It is known that AOD retrieval by satellite is highly dependent on the surface characteristics to which it is being mapped. This occurs due to the variation of the land cover features, which can significantly affect atmosphere properties since the algorithms' decoupling of surface and atmosphere reflectance can be complex (Xu et al., 2018; Feng and Zou, 2019). Therefore, it is crucial to comprehend the uncertainties of AOD retrievals across various land cover classes.

This research used land cover data provided by the Mapbiomas platform. Mapbiomas land cover data are generated annually from images from Landsat satellite series (spatial resolution of 30 m). To characterize the AERONET sites, a radius of 15km from the AERONET sites was considered. The majority classes, with at least 50% of the area, were defined as the land cover class for each location in the land cover map developed for 2020. AERONET sites where the majority classes had an area of less than 50% were defined as “mixed”. Table 8.3 presents the land cover class defined for each AERONET site.

Table 8.3 – Basic Statistics of AERONET Sites Grouped According to Major Land Cover Type.

LULC	AERONET site	Major Land Cover (%)	Biome	AOD ₅₅₀
				(mean ± std)
Forest	Amazon ATTO Tower	95.4	Amazon Rainforest	0.163 ± 0.117
	ARM Manacapuru	59.1	Amazon Rainforest	0.239 ± 0.242
	Balbina	61.4	Amazon Rainforest	0.17 ± 0.104
	Manaus-EMBRAPA	76.7	Amazon Rainforest	0.195 ± 0.17
Savanna	Petrolina-SONDA	56.9	Caatinga	0.09 ± 0.057
Grassland	Abracos Hill	85	Amazon Rainforest	0.36 ± 0.397
	Alta Floresta	62.6	Amazon Rainforest	0.278 ± 0.395
	Itajuba	54.5	Atlantic Forest	0.112 ± 0.106
	Ji-Parana-SE	75.6	Amazon Rainforest	0.242 ± 0.36
	Rio Branco	61.8	Amazon Rainforest	0.238 ± 0.311
Cropland	São Martinho-SONDA	56.2	Pampas	0.1 ± 0.183
Urban	São Paulo	81.8	Atlantic Forest	0.213 ± 0.158
	SP-EACH	70.8	Atlantic Forest	0.19 ± 0.142
Mixed	Belterra	-	Amazon Rainforest	0.252 ± 0.211
	Brasilia-SONDA	-	Cerrado	0.127 ± 0.105
	Campo Grande-SONDA	-	Cerrado	0.146 ± 0.227
	Cuiabá-MIRANDA	-	Cerrado	0.235 ± 0.343

8.2.3 Data processing

Measurements made by the AERONET solar photometer network provide a high sampling rate (~15 min) at the local point, while satellite imagery provides AOD measurements only during satellite overpass time (once a day) and with great spatial coverage. Thus, the direct comparison between the pixel value located on the surface measurement site, considering just the exact moment of the satellite overpass, restricts the probability of correspondence between the data; due to cloud cover or time delay between satellite and surface measurements. For this reason, a spatiotemporal window is generally adopted for the evaluation of MODIS-AOD products with AERONET-AOD (LIU et al., 2019a; MARTINS et al., 2017b; MHAWISH et al., 2019). In this way, this procedure is used to increase the number of data available for validation. In this context, the AOD data were linked using the following spatiotemporal windows: Firstly, the AERONET-AOD data were averaged over a temporal window of ± 30 minutes around the overpass time of the Aqua and Terra satellites. Secondly, a spatial window of 15 km was adopted, centered on the AERONET sampling sites.

8.2.4 Spatiotemporal AOD retrieval

To assess the spatiotemporal retrieval frequency of the algorithms, the temporal and spatial completeness were calculated. For this purpose, we calculated the annual and seasonal retrieval frequency averages for each pixel and also determined the spatial completeness on a daily basis using equation 8.2 (LIU et al., 2019a)

$$\text{Spatial completeness} = \frac{\text{Daily count of available AOD pixels}}{\text{Total number of pixels in the study area}} \times 100 \quad (8.2)$$

To account for the varying spatial resolutions of the AOD products, a 10x10 km grid was created for the evaluation of both temporal and spatial completeness. Only pixels with a centroid falling within the 10x10 km grid were considered valid for the analysis. The percentage of days with valid retrievals was then calculated for each grid cell. To avoid double-counting frequencies, only one retrieval was considered for AOD retrievals performed by both Aqua and Terra on the same day and in the same grid cell.

8.2.5 Aerosol Classification

To classify the aerosol type for each land cover class and AERONET site, it was used the classification system suggested by Logothetis et al. (2020). Fine Mode Fraction (FMF_{500nm}) was obtained considering the Aerosol Optical Depth version 3 and Spectral Deconvolution Algorithm (SDA) at level 2.0 for each AERONET site. Single Scattering Albedo (SSA_{440nm}) were obtained through the data set Inversions version 2 and considering the same data levels of FMF (500nm).

The Aerosol classification propoused by Logothetis, Salamalikis, And Kazantzidis (2020) considers the following classes: I. Fine highly absorbing ($SSA \leq 0.85$, $FMF > 0.6$ and $AE > 1.2$); II. Fine moderately absorbing ($0.85 < SSA \leq 0.9$, $FMF > 0.6$ and $AE > 1.2$); III. Fine slightly absorbing ($0.9 < SSA \leq 0.95$, $FMF > 0.6$ and $AE > 1.2$); IV. Fine non-absorbing ($SSA > 0.95$, $FMF > 0.6$ and $AE > 1.2$); V. Mixed absorbing ($SSA \leq 0.95$, $0.4 \leq FMF \leq 0.6$ and $0.6 \leq AE \leq 1.2$); VI. Mixed non-absorbing ($SSA > 0.95$, $0.4 \leq FMF \leq 0.6$ and $0.6 \leq AE \leq 1.2$); VII. Coarse absorbing ($SSA \leq 0.95$, $FMF < 0.4$ and $AE < 0.6$); VIII. Coarse non-absorbing ($SSA > 0.95$, $FMF < 0.4$ and $AE < 0.6$); IX. Other absorbing (divergences between the intervals in FMF and AE and $SSA \leq 0.95$); X. Other non-absorbing (divergences between the intervals in FMF and AE and $SSA > 0.95$).

8.2.6 Statistical Approach

The retrieval accuracy of different MODIS algorithms was evaluated through graphical comparison and statistical analysis, seeking to determine the quality and reliability of satellite measurements. Statistical analyzes include correlation coefficient (R), coefficient of determination (R^2), bias, Root Mean Square Error (RMSE), and Mean Absolute Error (MAE), often used in this type of analysis. assessment (LIU et al., 2019a; MARTINS et al., 2017b; MHAWISH et al., 2019; TIAN; GAO, 2019). The expected error (EE) was also evaluated, with defines the radial boundaries in which 67% of the matched AERONET and MODIS points fall. We used the EE definition of the dark target algorithm $\pm (0.05 + 0.15 \text{ AOD}_{\text{AERONET}})$.

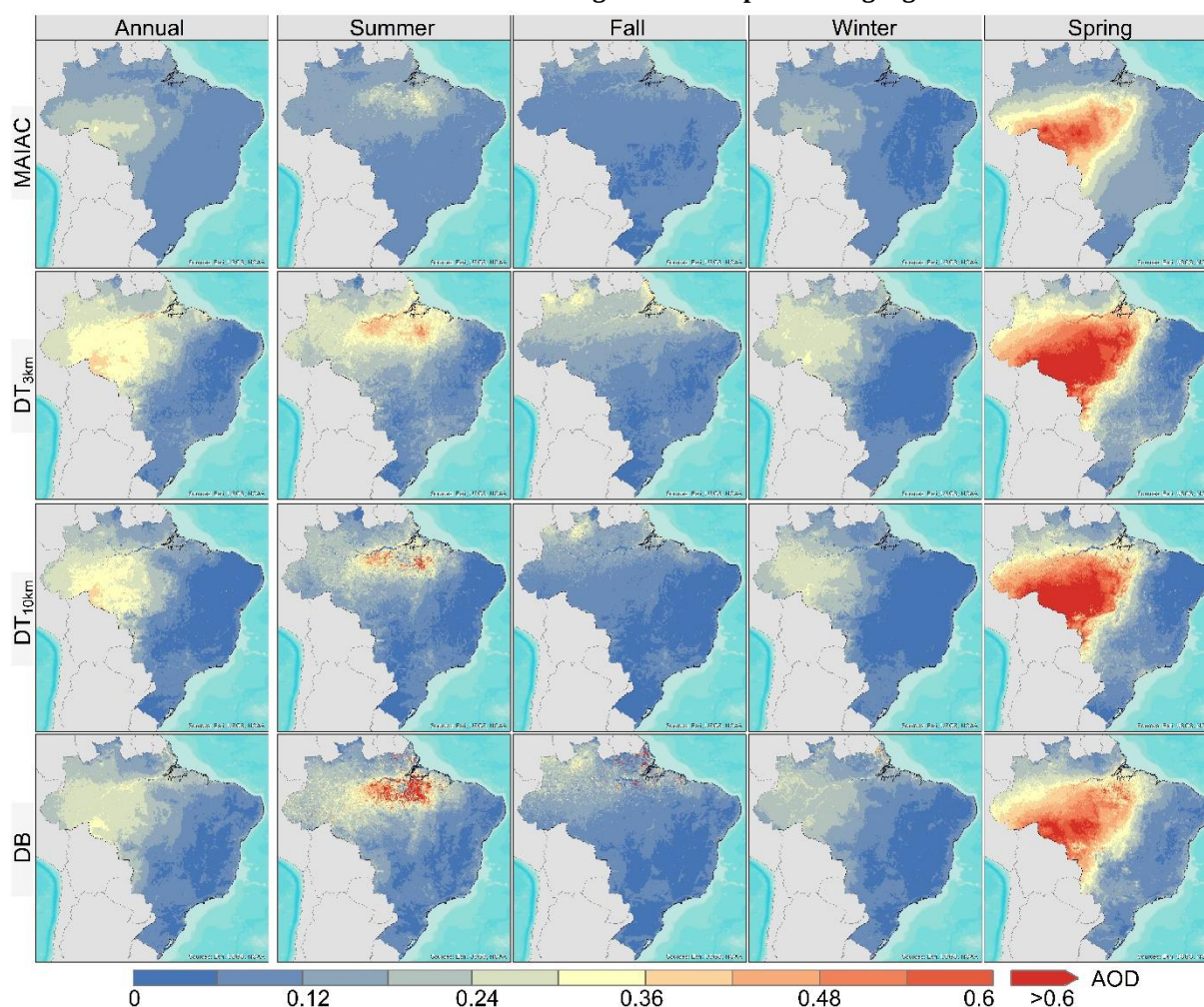
8.3 Results and discussion

8.3.1 Spatial intercomparison of MAIAC, DT, and DB

As the sources, types, and loading of aerosols that persist throughout the vast Brazilian territory are quite heterogeneous, the spatial intercomparison between MAIAC, DT (3 and 10 km), and DB retrieval algorithms was performed. By employing multi-year averages of annual and seasonal AOD, the spatiotemporal variations in AOD retrieval were explored, as well as the frequency and the spatial coverage of retrievals. The seasons consider typical southern weather for Brazil that are: "Summer" (Dec, Jan, and Feb); "Autumn" (Mar, Apr, and May); "Winter" (Jun, Jul, and Aug); and "Spring" (Sep, Oct, and Nov).

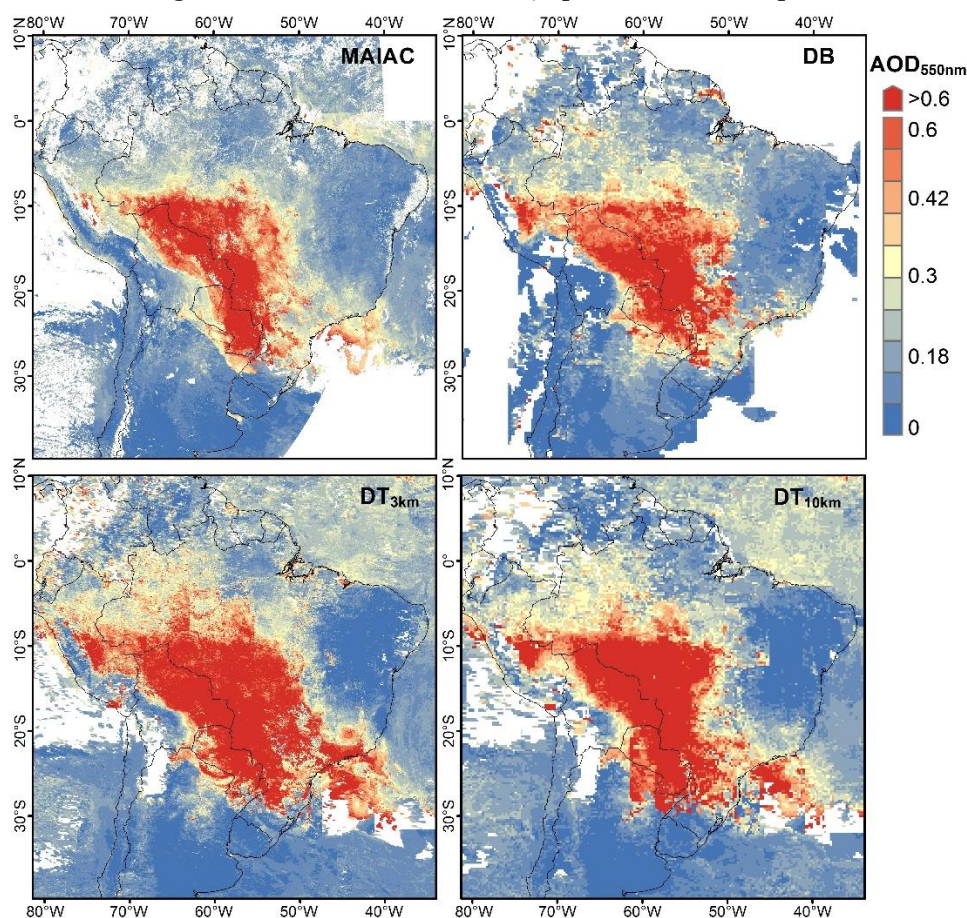
Spatial and temporal variations in the average (Figure 8.2) and frequency (Figure 8.4) of AOD are observed across Brazil. Considering all algorithms, it was observed a consistent spatial pattern in AOD, characterized by higher values over the Amazon region (MAIAC: 0.17 ± 0.05 , $\text{DT}_{3\text{km}}$: 0.28 ± 0.06 , and $\text{DT}_{10\text{km}}$: 0.23 ± 0.08 , DB: 0.22 ± 0.06) compared with non-Amazon region (MAIAC: 0.10 ± 0.02 , $\text{DT}_{3\text{km}}$: 0.10 ± 0.05 , and $\text{DT}_{10\text{km}}$: 0.06 ± 0.05 , DB: 0.08 ± 0.06). The high aerosol loads observed in the Amazon Biome are predominantly caused by emissions from biomass burning, which are closely linked to deforestation and agribusiness activities (MARTIN et al., 2010; SILVEIRA et al., 2020).

Figure 8.2 – Annual and seasonal averaged AOD distributions for MAIAC, DT (3 and 10km), and DB, retrieved over Brazil from 2000 to 2020. The averages were computed using a grid with cells of 10x10 km.



Divergences in the AOD values and spatial distribution may be attributed to different assumptions regarding aerosol properties and surface reflectance, which vary between MAIAC, DT, and DB (MHAWISH et al., 2019). In general, it is possible to observe that AOD values are significantly lower in MAIAC retrievals in Amazon, but slightly higher in the non-amazon area. The lower values observed in the MAIAC algorithm may also be attributed to its more refined resolution compared to other algorithms. This is evident when analyzing specific events, such as the aerosol transport event shown in Figure 8.3, which occurred between September 16th and 20th, 2019. The figure demonstrates how the higher resolution of MAIAC improves the ability to determine the source location and fine aerosol characteristics accurately. Conversely, algorithms with coarse spatial resolution tend to present more homogeneous values over large areas due to the larger pixel sizes, which can mask spatial variability in aerosol properties.

Figure 8.3 – Comparison between MAIAC, DT (3 and 10 km), and DB retrievals for long-range transport of biomass burning aerosols over South America (September 16th to September 20th, 2019).

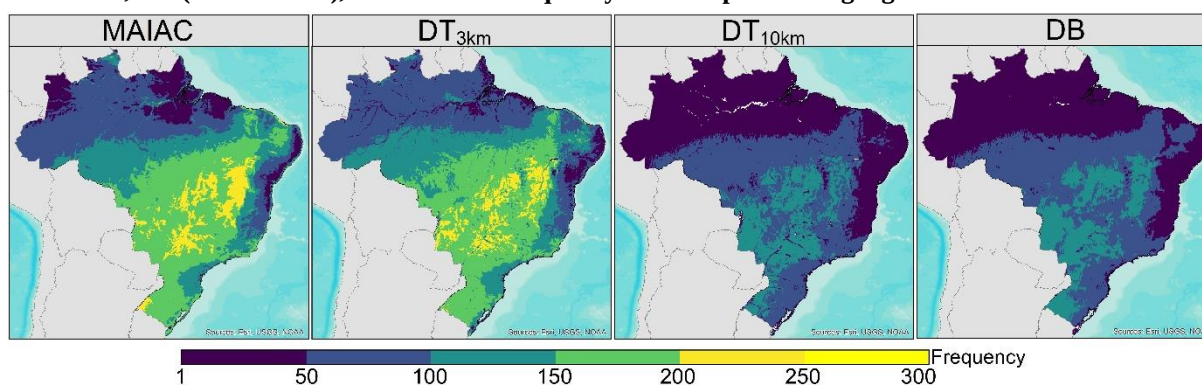


Temporally, the spring season in the Amazon is characterized by the highest AOD values, with the DT_{3km} (0.45 ± 0.16) and DT_{10km} (0.41 ± 0.08) exhibiting the highest mean values. In comparison, DB showed a mean AOD value of 0.36 ± 0.14 and MAIAC of 0.29 ± 0.14 . The highest seasonal averages of AOD were also observed during spring in the non-Amazon region (MAIAC: 0.16 ± 0.08 , DT_{3km} : 0.17 ± 0.13 , and DT_{10km} : 0.13 ± 0.12 , DB: 0.15 ± 0.10). Higher AOD values are typically observed during the spring season, as this period presents a greater concentration of biomass burning emissions, with the majority occurring in September and October, despite fire activity remaining high throughout the dry season, from June to November (POPE et al., 2020). After Spring, the season with the highest AOD values is Summer (MAIAC: 0.13 ± 0.04 , DT_{3km} : 0.19 ± 0.10 , and DT_{10km} : 0.14 ± 0.10 , DB: 0.17 ± 0.15). This is followed by Winter (MAIAC: 0.10 ± 0.05 , DT_{3km} : 0.13 ± 0.10 , and DT_{10km} : 0.10 ± 0.09 , DB: 0.12 ± 0.08) and Fall (MAIAC: 0.09 ± 0.03 , DT_{3km} : 0.14 ± 0.07 , and DT_{10km} : 0.09 ± 0.06 , DB: 0.11 ± 0.12).

The retrieval frequency varies significantly among the different algorithms, both in terms of the frequencies themselves and their spatial distribution (Figure 8.4). Regarding the spatial

distribution, there is a lower frequency of AOD retrievals observed in the central-northern portion of the Amazon region and along the Brazilian coast. These regions have an average annual frequency of less than 50%. This is particularly attributed to the high occurrence of clouds in these regions, which can be attributed to the abundance of water vapor, promoting the formation of clouds at a higher frequency (LIU et al., 2022). Algorithms with higher spatial resolution, MAIAC and DT_{3km}, showed a higher frequency of retrievals, as in other regions worldwide (BELLE; LIU, 2016; LIU et al., 2022; MHAWISH et al., 2019). On the other hand, for the coarser spatial resolution products (10 km), namely DT_{10km} and DB, average annual frequencies lower than 50% were observed throughout Brazil, with regions characterized by high cloudiness (Amazon region and coast) exhibiting an annual average frequency of less than 50 retrievals. These discrepancies in the amount of data retrieved daily by the algorithms, in addition to spatial resolution, are related to the ability of each algorithm to retrieve different aerosol types, such as smoke and dust (MHAWISH et al., 2019).

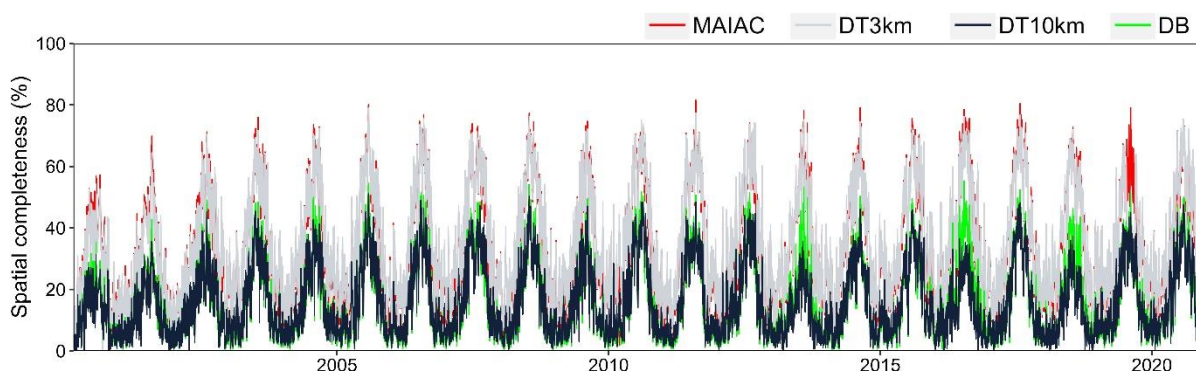
Figure 8.4 – Annual AOD retrieval frequency over Brazil from different MODIS retrieval algorithms: MAIAC, DT (3 and 10 km), and DB. The frequency was computed using a grid with cells of 10x10 km.



In Brazil, rainy periods are prevalent from late spring to early autumn. As a result, the AOD retrievals are reduced during this time due to a higher presence of clouds. Seasonal AOD retrieval frequencies for the algorithms are presented in Figure D.1 (Appendix D) and daily Spatial completeness in Figure 8.5. As expected, the summer season has the lowest data frequency, with several regions having a frequency of less than 20%, particularly for DT_{10km} and DB. The daily spatial completeness indicates that the coverage of AOD is significantly limited during the rainy months, with less than 20% of the study area covered on several days. The algorithms with coarse spatial resolution, such as DT_{10km} and DB, have almost no coverage during this period. On the other hand, during winter, a predominantly dry season, there is a higher frequency of data retrieval. The central region of Brazil, in particular, has the greatest availability of data during this period, with some locations having a frequency greater than 80%

(MAIAC and DT_{3km}). This can be attributed to the region's majority non-forest cover (as shown in Figure 8.1), leading to lower evapotranspiration rates (LOARIE et al., 2011; RODRIGUES et al., 2022), and resulting in less moisture in the atmosphere (LIU et al., 2022). The reduced moisture content can limit cloud formation, especially since this region is more inland from the continent, avoiding the sea breeze, and experiences fewer cold fronts compared to the southern and southeastern regions of the country (ANJOS; LOPES, 2019; SATYAMURTY; NOBRE; SILVA DIAS, 1998).

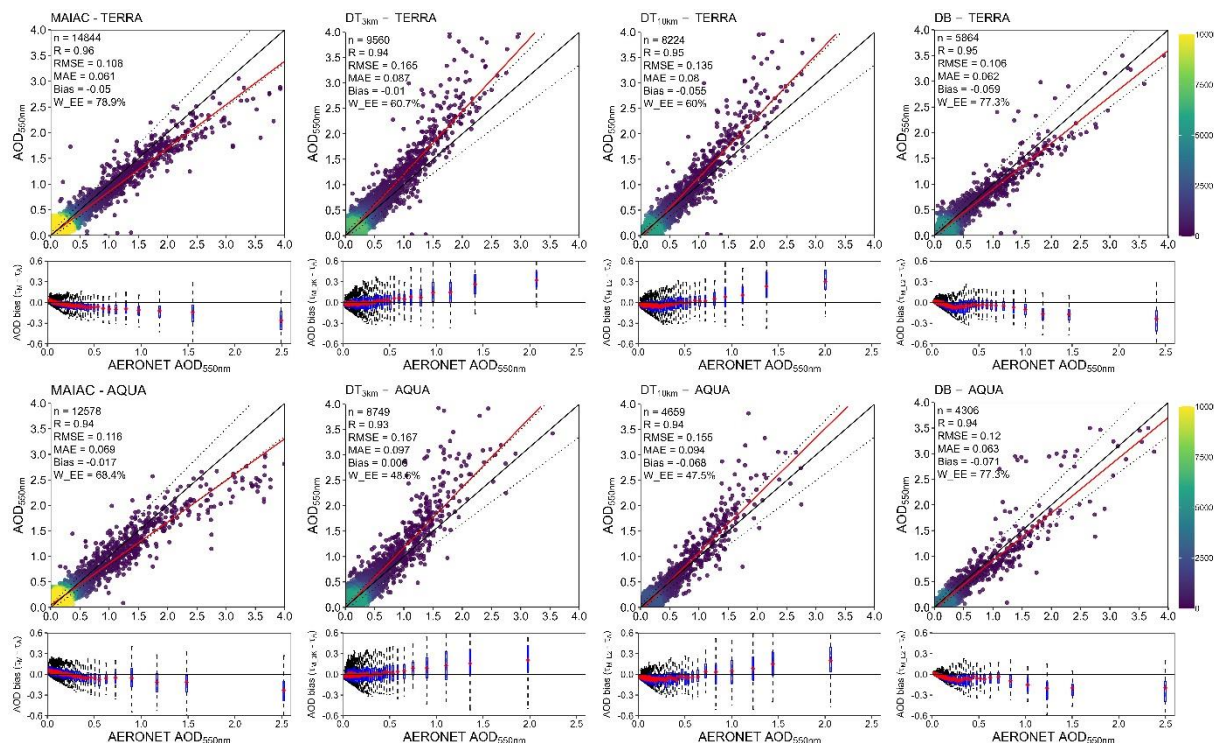
Figure 8.5 – Daily spatial completeness for MAIAC, DT (3 and 10km), and DB from 2000 to 2020.



8.3.2 Overall accuracy evaluation

The overall quality assessment of the MODIS-AOD products was evaluated based on average values, standard deviation (SD), retrievals within the EE envelope, RMSE, MAE, R, and R^2 . This evaluation was conducted separately for the data obtained from the MODIS sensors on both the Aqua and Terra satellites. The aim was to assess the consistency of the data retrieval algorithms. Figure 8.6 displays the scatter plot for each algorithm and satellite, accompanied by a summary of the evaluated statistics. In general, the results demonstrate significant positive correlations ($R > 0.9$) between the four satellite products and the AERONET ground truth data. The strong correlation also remains when the AOD products are compared (Figure D.2), whether evaluated globally across algorithms or separately for each satellite. In this case, the correlation values consistently exceed 0.95, indicating a robust relationship between the AOD measurements.

Figure 8.6 – Overall accuracy of MODIS (Aqua and Terra) AOD algorithms MAIAC, DT (3 and 10 km), and DB (all AERONET sites across Brazil). The matchup pairs are separated into 100 bins along with AERONET AOD values. The red, black, and dashed lines on the scatterplot stand in for the regression line, the 1:1 reference line, and the expected error [EE = $\pm (0.05 + 0.15 \text{ AOD})$], respectively. The AOD bias box plot uses the 25 % and 75 % percentiles for each of 100 bins. The red points in the box plot are the mean bias for each of 100 bins.



Upon observing the scatter plots, it is evident that there is a tendency for underestimation of the AOD values in the case of MAIAC and DB algorithms. In contrast, overestimation is observed for the DT (3 km and 10 km) algorithms. However, a different behavior can be observed upon comparing the bias boxplots (subfigure in Figure 8.6), which helps explain this variation. It is evident that the DT algorithm tends to underestimate low AOD levels (generally less than 0.5) and overestimate higher AOD levels. On the other hand, MAIAC and DB algorithms tend to overestimate low levels and underestimate higher AOD levels. Similar patterns of overestimation of low AOD values by MAIAC and underestimation of high AOD levels have been observed in various locations worldwide, including China, Central and South Asia, and California (USA) (CHEN et al., 2021; FALAH et al., 2021; LIU et al., 2019a; MHAWISH et al., 2019). In contrast, divergences are observed in other regions, where the MAIAC algorithm tends to either consistently underestimate or overestimate AOD values (FALAH et al., 2021; ZHANG et al., 2019c). This implies that although MAIAC exhibits overall consistency, its behavior can change depending on the region and properties of the aerosol loads (e.g., size and absorption). Regarding DT and DB algorithms, there is a greater variation observed depending on the region of analysis (CHEN et al., 2021; HE et al., 2017b; LIU et al., 2019a; MHAWISH

et al., 2019). However, the general behavior remains similar to what was observed in this study, with DT showing overestimated values and DB underestimating the AOD values.

When comparing the performance of different satellites, overall consistency is observed across most statistical analyses. However, Terra tends to exhibit slightly better performance compared to Aqua. In general, Terra presents slightly lower RMSE, MAE, and absolute Bias values and slightly greater R and R^2 . The most notable difference between the satellites is observed when examining the percentage of retrievals within the EE envelope. In this aspect, Terra typically exhibits noticeably higher values, indicating better agreement with the expected error range. However, it is important to note that there is an exception in the case of DB, where Aqua and Terra showed the same performance. According to Levy et al. (2010) and Remer et al. (2005), a satisfactory accuracy for an AOD product occurs when more than 66% of retrievals are within the EE limits. By using this approach, it is possible to conclude that the DT did not meet the requirement for confident retrievals, while the MAIAC and DB did. In the case of DT, less reliable retrievals are mainly observed for the Aqua satellite (retrievals within EE - DT_{3km}: 48.6% and DT_{10km}: 47.5%). In addition, it is important to mention that the EE envelope used for this evaluation [EE = $\pm (0.05 + 0.15 \text{ AOD})$] was determined for the DT algorithm. When using the EE envelope defined by Martins et al. (2017b) for MAIAC in South America [EE = $\pm (0.05 + 0.05 \text{ AOD})$], only the Terra satellite attends more than 66% of retrievals within the EE envelope (MAIAC-Terra: 66.4% and MAIAC-Aqua: 58.3%).

As previously mentioned, the variation in product quality may be attributed to the different AOD levels. Therefore, to examine the effects of AOD levels on the retrieval quality, the dataset was divided and analyzed based on four distinct bin levels: low level (< 0.2), moderate level (0.2–0.4), moderate-high level (0.4–0.6), and high level (> 0.6), as indicated by Liu et al. (2019a). Based on Table 8.4, it is observed that the majority of AOD retrievals correspond to levels lower than 0.4. However, it is at the higher AOD values, greater than 0.6, that the more expressive correlations are found, with R values around 0.9 (except for DB-Aqua, R: 0.78). Regarding the error measures, it is notable that MAIAC consistently exhibits the lowest values of RMSE, MAE, and absolute Bias for the low-level bin, as expected. At intermediate AOD values ($0.2 < \text{AOD} < 0.4$), DT_{10km} demonstrates low values of RMSE, MAE, and absolute Bias. For higher AOD values, DB exhibits the best results. Consistent with the overall analysis, Terra generally outperforms Aqua in terms of performance.

Table 8.4 – Accuracy evaluation of MAIAC, DT, and DB at different size levels. The bold number represents the highest performance among the four products for each indicator and level.

		MAIAC		DT (3km)		DT (10km)		DB	
		Terra	Aqua	Terra	Aqua	Terra	Aqua	Terra	Aqua
n	<0.2	10714	8579	6589	5992	6091	3603	4538	3310
	0.2-0.4	2534	2830	1507	1540	1007	508	520	314
	0.4-0.6	690	496	612	549	445	223	357	193
	>0.6	906	673	852	668	681	325	449	232
R	<0.2	0.56	0.41	0.48	0.41	0.50	0.51	0.47	0.61
	0.2-0.4	0.49	0.53	0.42	0.41	0.53	0.48	0.54	0.48
	0.4-0.6	0.58	0.51	0.40	0.35	0.43	0.46	0.49	0.48
	>0.6	0.92	0.89	0.91	0.87	0.92	0.88	0.92	0.78
RMSE	<0.2	0.059	0.060	0.076	0.084	0.085	0.097	0.073	0.066
	0.2-0.4	0.104	0.117	0.105	0.126	0.087	0.116	0.095	0.106
	0.4-0.6	0.114	0.118	0.147	0.161	0.106	0.109	0.124	0.114
	>0.6	0.332	0.370	0.474	0.494	0.371	0.461	0.268	0.338
MAE	<0.2	0.042	0.046	0.058	0.068	0.065	0.079	0.047	0.044
	0.2-0.4	0.082	0.097	0.084	0.104	0.067	0.089	0.071	0.079
	0.4-0.6	0.091	0.090	0.117	0.127	0.084	0.083	0.087	0.084
	>0.6	0.202	0.236	0.293	0.317	0.234	0.275	0.179	0.235
Bias	<0.2	-0.021	0.003	-0.039	-0.043	-0.053	-0.070	-0.029	-0.032
	0.2-0.4	0.003	0.046	0.042	0.060	-0.005	-0.017	-0.014	-0.058
	0.4-0.6	-0.052	-0.009	0.083	0.084	0.033	0.002	0.004	-0.047
	>0.6	-0.146	-0.119	0.264	0.275	0.191	0.198	-0.072	-0.141

8.3.3 Land cover class dependency

The retrieval of aerosols is intricately linked to the characteristics of the terrestrial surface, as it requires distinguishing between surface reflectance and the influence of aerosol scattering/absorption, which becomes particularly challenging under low aerosol loading conditions (LEVY et al., 2007). Therefore, in order to assess the impact of land surface on AOD retrievals, an investigation was conducted to examine the relationship between retrieval quality and land cover. Table 8.5 and Figure D.3 to Figure D.8 provide the statistical results of the accuracy assessment for each land cover class and retrieval algorithm. Overall, the algorithms displayed higher correlation and lower errors over Forest, Cropland Grassland, and Mixed cover classes. On the other hand, lower statistical quality was observed for Savanna and Urban classes, demonstrating that AOD retrievals are sensitive to land cover types.

Table 8.5 – Comparison of the MAIAC, DT, and DB retrieval accuracy for different land cover classes. The bold number is the highest performance among the products by each indicator.

		MAIAC		DT (3km)		DT (10km)		DB	
		Terra	Aqua	Terra	Aqua	Terra	Aqua	Terra	Aqua
R	Forest	0.89	0.80	0.94	0.92	0.91	0.90	0.85	0.91
	Savanna	0.51	0.46	0.49	0.55	0.47	0.50	0.61	0.71
	Grassland	0.97	0.95	0.97	0.96	0.96	0.94	0.98	0.97
	Cropland	0.88	0.77	0.94	0.95	0.93	0.92	0.93	0.96
	Urban	0.79	0.77	0.88	0.90	0.84	0.85	0.80	0.89
	Mixed	0.94	0.94	0.92	0.94	0.91	0.91	0.94	0.90
R ²	Forest	0.79	0.64	0.88	0.84	0.82	0.82	0.71	0.83
	Savanna	0.26	0.21	0.24	0.30	0.22	0.25	0.37	0.51
	Grassland	0.94	0.91	0.94	0.91	0.93	0.89	0.95	0.95
	Cropland	0.77	0.60	0.88	0.91	0.86	0.84	0.87	0.91
	Urban	0.63	0.60	0.78	0.80	0.71	0.72	0.65	0.78
	Mixed	0.89	0.88	0.85	0.88	0.84	0.83	0.88	0.81
RMSE	Forest	0.070	0.081	0.105	0.111	0.150	0.161	0.080	0.090
	Savanna	0.091	0.082	0.108	0.099	0.097	0.093	0.051	0.038
	Grassland	0.114	0.135	0.155	0.208	0.202	0.216	0.092	0.107
	Cropland	0.055	0.059	0.057	0.071	0.136	0.079	0.044	0.045
	Urban	0.096	0.095	0.090	0.087	0.089	0.092	0.138	0.123
	Mixed	0.118	0.111	0.130	0.132	0.144	0.138	0.128	0.120
MAE	Forest	0.050	0.060	0.070	0.085	0.111	0.127	0.055	0.073
	Savanna	0.062	0.060	0.095	0.087	0.081	0.079	0.037	0.029
	Grassland	0.062	0.078	0.076	0.100	0.091	0.112	0.052	0.055
	Cropland	0.035	0.037	0.041	0.055	0.050	0.053	0.026	0.024
	Urban	0.074	0.071	0.070	0.068	0.066	0.063	0.108	0.101
	Mixed	0.059	0.064	0.095	0.105	0.089	0.096	0.070	0.063
Bias	Forest	-0.018	0.036	0.059	0.078	0.104	0.124	0.040	0.053
	Savanna	0.043	0.042	-0.094	-0.086	-0.068	-0.063	0.010	-0.002
	Grassland	-0.030	0.010	0.014	0.004	0.045	0.057	-0.006	-0.029
	Cropland	0.013	0.012	-0.021	-0.046	-0.006	-0.033	-0.012	-0.018
	Urban	-0.055	-0.035	-0.040	-0.051	-0.010	-0.010	-0.097	-0.099
	Mixed	-0.026	-0.006	-0.077	-0.086	-0.046	-0.052	-0.054	-0.046
Within EE	Forest	80.0	69.2	66.6	47.6	42.7	24.0	75.1	58.6
	Savanna	67.3	63.1	23.2	27.7	40.2	36.8	83.7	90.8
	Grassland	83.1	66.3	73.3	57.8	70.7	52.2	88.0	87.4
	Cropland	85.6	82.2	77.3	58.2	75.7	62.4	94.8	96.3
	Urban	60.5	67.0	61.2	68.6	67.2	74.1	36.7	41.7
	Mixed	80.6	72.1	39.9	32.4	48.4	40.5	73.0	79.8

With an increase in vegetation coverage, it is possible to observe an enhancement in retrieval quality. This can be attributed to a combination of factors: the inherent surface characteristics, which typically result in lower albedos compared to urban areas, and the higher aerosol loads found in the Amazon rain Forest and Cerrado Biomes (ROTHMUND et al., 2018), where most AERONET sites with better statistical results are located. Higher albedo values influence TOA reflectance making it more complex to distinguish between surface and atmospheric contributions, which poses a significant challenge in aerosol retrieval on land, particularly in wavelengths used to retrieve AOD (VIS and NIR spectra) (REMER et al., 2013a). In addition, the high aerosol loads during intense biomass burning in Brazil are the primary determinant of validation statistics in the region (HYER; REID; ZHANG, 2011). Therefore, sites outside the

axis of major aerosol emissions and transport due to biomass burning tend to exhibit lower AOD values and, consequently, lower statistical quality. The AERONET site in the savanna area exemplifies this, exhibiting poor validation results despite having similar albedo characteristics to the grassland class. The AERONET site classified as savanna (Petrolina-SONDA) presents significantly low AOD values ($AOD < 0.1$). In these cases, over semi-arid regions, MODIS tends to approach its noise threshold, resulting in a general lack of skill and frequently overestimating AOD values (HYER; REID; ZHANG, 2011; MARTINS et al., 2017b). When considering Petrolina-SONDA as an example, the RMSE values generally exceed 0.08 (except for the DB algorithm), demonstrating comparable or even higher values than the other land cover classes. It is noteworthy that Petrolina-SONDA exhibits this pattern despite having the lowest AOD values among the AERONET sites.

In Forest areas (Table 8.5 and Figure D.3), the retrievals correlated well with ground measurements, with $R > 0.8$ for all the products. Although the DT algorithm showed better correlations with AERONET sites classified as Forest ($R > 0.9$ for DT 3 and 10km), lower errors and bias were observed for MAIAC, which also presented a higher percentage of retrievals within the EE envelope (T: 80% and A: 69.2%). This demonstrates that although the DT algorithm can better capture the variation of AOD over forest areas, the frequent overestimation of AOD values (higher bias values in Table 8.5) tends to lead to more significant errors (higher MAE and RMSE and a lower number of retrievals within EE envelope for this algorithm). This result is in line with Lyapustin et al. (2018), which indicates that the MAIAC algorithm improves the accuracy of dark target retrievals better than the DT algorithm.

All algorithms presented good accuracy results for the Grassland cover type (Table 8.5 and Figure D.5), with DB showing slight higher performance (better correlation and retrievals within EE envelope and lower errors, in general). One important aspect to mention is the consistency of the DB retrievals, which show good accuracy results for the AOD data retrieved using reflectance from both Aqua and Terra satellites. On the other hand, for MAIAC and DT algorithms, it can be observed that the data obtained from the MODIS sensor on the Terra satellite perform better (with approximately +15% retrievals within the EE envelope) compared to the Aqua satellite.

The DB algorithm also yielded the best results for the Savanna and Cropland cover classes (Table 8.5 and Figure D.4 and Figure D.6). These classes are represented by only one

AERONET site and have the lowest average AOD values (Table 8.3). The AERONET site in the Savanna area is in a semi-arid region where agricultural use is limited due to harsh climatic conditions (CUNHA et al., 2010). The soils in the region have sandy and sandy loam textures with very hard consistency and the presence of surface and subsurface rock fragments (CUNHA et al., 2010). These factors, combined with sparse vegetation (CUNHA et al., 2010), result in a surface with higher reflectivity. For the Agriculture class, higher reflectivity is also expected during specific periods of the year, as agriculture is a class with significant intra-annual variation, including periods of bare areas due to the transition between agricultural cycles (RUDKE et al., 2019). The mentioned aspects result in Cropland and Savanna having brighter surfaces, for which the DB algorithm generally performs better than MAIAC and DT (TAO et al., 2019; WEI et al., 2019). MAIAC also presented good results, demonstrating accuracy over brighter surfaces, as previously suggested (e.g., JETHVA; TORRES; YOSHIDA, 2019; LYAPUSTIN et al., 2018; TAO et al., 2019; WEI et al., 2019).

Over urban areas, MAIAC exhibited the lowest correlations compared to DT and DB (MAIAC < 0.8 ; DT_{3km} ~ 0.9 ; DT_{10km}: 0.85; and DB: 0.8 and 0.89). DB, however, has the lowest percentage of retrievals within the EE (T: 36.7 and A: 41.7). In the study conducted by Damascena et al. (2021) in São Paulo city, it was found that DT_{3km} and MAIAC yielded comparable results, with MAIAC showing a slightly stronger correlation for Terra and DT_{3km} exhibiting a slightly stronger correlation for Aqua. Given the substantial spatial variation of AOD over urban areas resulting from diverse aerosol emission sources, adopting a 15km radius for collocating satellite and surface data may influence the measurements obtained by higher-resolution products, specifically MAIAC. On the other hand, MAIAC presented the best results over Mixed land cover.

Table 8.6 shows the seasonal retrieval accuracy for evaluated products. Most of the products and land cover classes showed better correlations during winter and spring. This can be attributed to the characteristic dry periods that occur in most locations where the AERONET sites are situated. These dry periods favor the biomass burning season, which tends to coincide with these seasons, leading to elevated aerosol loads (Figure 8.2). As a result, the higher aerosol loads during this period contribute to the enhanced performance observed in winter or spring.

Table 8.6 – Seasonal comparison of the retrieval accuracy of the MAIAC, DT, and DB for different land cover classes. The bold number is the highest performance among the four seasons by each indicator. The red number indicates the non-significant correlation ($\alpha > 0.05$).

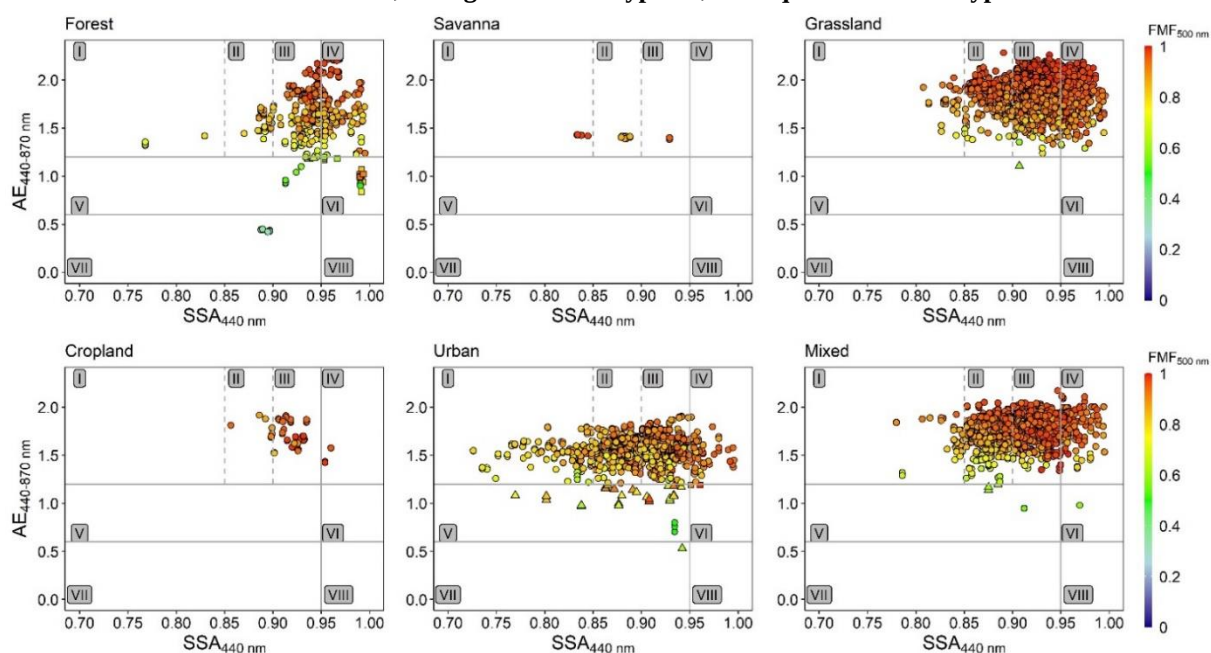
			Forest	Savanna	Grassland	Cropland	Urban	Mixed
R	MAIAC	Summer	0.90	0.54	0.83	0.41	0.47	0.47
		Fall	0.30	0.52	0.31	0.59	0.28	0.58
		Winter	0.74	0.18	0.97	0.95	0.93	0.79
		Spring	0.82	0.46	0.96	0.77	0.95	0.85
	DT _{3km}	Summer	0.94	0.65	0.87	0.65	0.60	0.73
		Fall	0.26	0.48	0.44	0.72	0.51	0.37
		Winter	0.80	0.30	0.96	0.94	0.77	0.89
		Spring	0.92	0.48	0.95	0.88	0.89	0.93
	DT _{10km}	Summer	0.94	0.56	0.92	0.70	0.70	0.64
		Fall	0.43	0.29	0.56	0.60	0.45	0.40
		Winter	0.92	0.43	0.97	0.97	0.77	0.89
		Spring	0.92	0.46	0.96	0.91	0.93	0.94
	DB	Summer	0.74	0.61	0.75	0.72	0.50	0.53
		Fall	0.09	0.54	0.53	0.67	0.51	0.42
		Winter	0.90	0.53	0.98	0.97	0.74	0.86
		Spring	0.82	0.44	0.97	0.90	0.85	0.91
Within EE	MAIAC	Summer	69.2	64.3	49.9	74.3	51.9	63.3
		Fall	62.0	54.1	66.2	84.7	75.6	73.4
		Winter	78.4	67.4	84.3	89.4	88.2	65.4
		Spring	77.3	68.3	71.7	85.8	70.3	53.9
	DT _{3km}	Summer	28.9	41.5	42.3	61.2	57.4	61.0
		Fall	24.0	54.6	58.8	72.4	71.2	56.5
		Winter	41.5	37.2	68.8	82.4	70.9	33.2
		Spring	28.6	31.4	58.4	59.4	72.1	46.5
	DT _{10km}	Summer	44.8	26.9	60.4	63.6	62.2	56.5
		Fall	50.0	38.7	80.2	74.2	65.6	49.2
		Winter	69.6	22.2	67.9	80.6	60.6	26.3
		Spring	57.0	23.7	66.1	59.9	67.1	41.0
	DB	Summer	63.6	91.4	55.9	96.7	53.6	82.9
		Fall	25.0	91.1	84.9	97.8	59.3	87.1
		Winter	72.1	90.9	94.1	89.7	41.8	76.9
		Spring	74.1	81.7	77.9	97.0	34.4	53.0

In the case of the Forest class, the highest correlations were observed during the summer, which can be attributed to the high aerosol loads in the region where the AERONET sites with this characteristic are located (Figure 8.2). During the summer, the region where the AERONET sites classified as Forest are situated is notably influenced by two factors: local biomass burning, particularly in December, and the long-range transport of aerosols from Africa (predominantly contribute to the composition of black carbon during the months of January and February) (HOLANDA et al., 2023). Similar to Forest, Savanna tends to exhibit the highest correlations during the summer, except for the MAIAC product, which shows the best correlation for Savanna during the fall. In the case of Savanna, a possible explanation could be linked to higher precipitation during the summer, as increased relative humidity (RH) tends to induce hygroscopic growth (aerosol swelling) of particles, resulting in both increased volume and particle size (see Figure D.9). Studies have demonstrated that increasing RH typically induces hygroscopic growth of particles, leading to an enhancement in the scattering coefficient

and, consequently, an increase in AOD (e.g., LIU et al., 2013; PRASAD; BASHA; RATNAM, 2023; STIRNBERG; CERMAK; ANDERSEN, 2018).

To assess the impact of aerosol properties, the classification of aerosols was examined for each land cover class, based in the classification proposed by Logothetis, Salamalikis, and Kazantzidis (2020) (Figure 8.7; The detailed classification for each of the AERONET sites can be found in Figure D.10). Through the Figure 8.7 it is verified significant variability in the characteristics of the aerosol, both in terms of inter- and intra-classes. AERONET sites located in forested areas predominantly exhibit slightly absorbing or non-absorbing fine aerosols (classes III and IV). AERONET sites located in grassland and mixed-class areas predominantly show fine aerosols ranging from non-absorbing to moderately absorbing (classes II to IV). Aerosols in urban areas also exhibit a predominance of fine particles, but with a wide range of radiation absorption, varying from non-absorbing to highly absorbing (classes I to IV). Although there are only a few data points for verification, it is observed that the AERONET site in the savanna area also exhibits fine particles, but with a tendency towards high to moderate absorption (mainly classes I and II). For agriculture, it is observed that almost 80% of the evaluated data are represented by class II, which indicates fine aerosols with moderate absorption.

Figure 8.7 – Aerosol classification considering each land cover class. The dots show the Aerosol Type between I and VIII, triangles Aerosol Type IX, and squares Aerosol Type X.



A predominance of fine-mode aerosols is observed through evaluating aerosol type classification for each land cover class, although with highly variable absorption. This may

demonstrate that in addition to the impacts related to the variation in reflectance of different surfaces (land cover classes), there may also be an impact on the type of aerosol present in each of these classes. In general, it can be observed that aerosols present in Forest, Grassland, and Cropland classes predominantly contain components with low or non-absorbing properties. Studies conducted mainly in the Amazon region (where most of the AERONET sites with this coverage are located) have shown that aerosols from biomass burning are primarily found in the accumulation mode, corresponding to the fine mode (BRITO et al., 2014; MORGAN et al., 2020; ORDOU; AGRANOVSKI, 2019; REID et al., 2005). Furthermore, the studies observed a high contribution of non-refractory organic components, followed by inorganic components (nitrate, sulfate, ammonium, and chloride), and black carbon (MORGAN et al., 2020). A higher presence of black carbon was observed in the case of biomass burnings in Savanna (MORGAN et al., 2020). This may explain the potentially higher absorptive properties of the aerosols observed for this land cover class. For the Forest, at the AERONET site Amazon ATTO Tower, aerosols classified as Class VII (Coarse aerosols - absorbing) were observed, which may be related to the long-range transport of dust originating from Africa, as previously verified for this site (HOLANDA et al., 2023; NOGUEIRA et al., 2021). The urban class exhibits a wide variation in aerosol types, which is associated with the diverse sources of aerosol emissions in urban centers (MARTINS et al., 2017b; RUDKE et al., 2023). Thus, the multiple natural and anthropogenic sources and the different sets of aerosol optical properties can pose additional challenges for satellite aerosol retrievals at distinct surfaces.

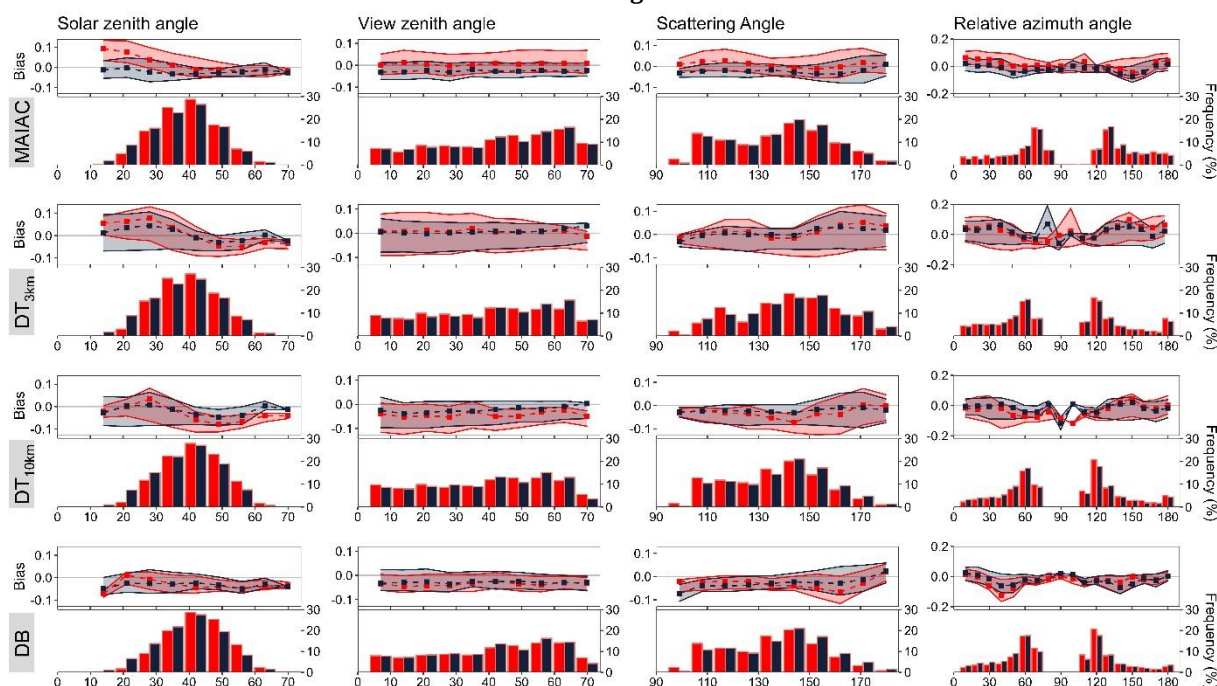
8.3.4 Angular dependence of AOD retrieval

Satellite-based aerosol retrievals entail numerous sources of uncertainty, with varying degrees of magnitude. These uncertainties encompass factors such as satellite and solar geometries, which can introduce potential errors into the retrieval process (MHAWISH et al., 2019). Therefore, to investigate the potential influence of angular factors on the accuracy of AOD retrievals, we examined the impact of solar zenith angle (SZA), view zenith angle (VSA), scattering angle (SA), and relative azimuth angle (RAA) on AOD retrieved by MAIAC, DT (3 and 10 km), and DB products (Figure 8.8).

In terms of the SZA, all algorithms showed some dependency to retrieval angle. The algorithms generally showed more matches for angles between 25 and 50° (representing approximately 80 to 85% of the matchups, depending on the algorithm and satellite). DB showed the least dependency on SZA, although the AOD retrieved by the Aqua satellite exhibited a slight

tendency towards negative bias as SZA increased. For MAIAC, a slightly decreasing trend was observed with increasing SZA for the Aqua satellite (the mean biases for Aqua range from -0.02 to 0.09). Specifically, there is a slight tendency for overestimation when SZA is less than 35°. MAIAC retrievals using Terra data showed a reduced dependence on SZA. In this scenario, the bias variation was smaller, with mean values ranging from -0.03 to -0.003, indicating a slight negative bias. DT 3 and 10 km show similar behavior but with different magnitudes, with the mean bias increasing for small SZAs; after the maximum mean bias (around 30°), bias tends to decrease as SZA increases. Compared to DT_{10km}, DT_{3km} tends to present higher absolute bias for small SZAs, the opposite happens for higher SZAs, with DT_{10km} showing higher absolute bias.

Figure 8.8 – Dependency of AOD bias to solar zenith angle, view zenith angle, scattering angle, and relative azimuth angle for MAIAC, DT, and DB products. The black and red points represent the mean bias for Terra and Aqua satellites, respectively. The top and bottom lines in shaded areas are the 75% and 25% percentiles for Terra (grey) and Aqua (red). The vertical bars represent the percentage of matchups in each angle bin.



For VZA, slight or no angular dependency was observed. Consistent with previous studies (e.g., LIU et al., 2019a; MHAWISH et al., 2019), the algorithms exhibited higher match frequency at the scan edge (most occurring between 40 and 65°, ~55% of matchup), while fewer matches were observed at the nadir. MAIAC and DB presented no dependency on the VZA; bias lines did not fluctuate much for both Aqua and Terra satellites. For DT, slight fluctuations in bias

values are observed with variations in VZA, especially for data acquired from the Terra satellite. In this case, a slight positive trend is observed for VZA above 55° , particularly for DT_{3km}.

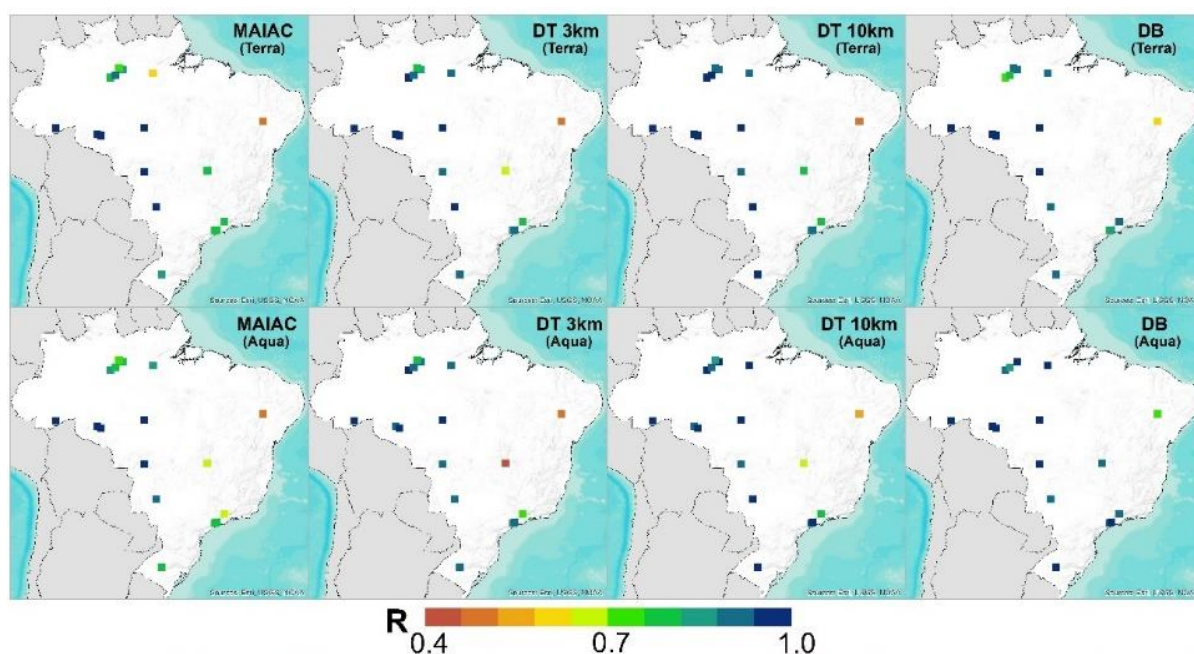
As for collection 6 (C6) (LIU et al., 2019a; MHAWISH et al., 2019), C6.1 also demonstrated dependence on variations in the Scattering Angle. In this context, MAIAC appears to be the algorithm with the least angular dependence, showing only a slightly positive trend for SA $> 160^\circ$. MAIAC-Terra predominantly exhibits negative biases, indicating underestimation (bias range: -0.037 to 0.008 for Terra). On the other hand, MAIAC-Aqua shows overestimation for angles between 100 and 125° (bias range: 0.009 to 0.025), while achieving a good fit for the range between 130 and 140° (bias range: -0.005 to 0.001). The DB algorithm also exhibited a positive bias trend for SA $> 160^\circ$ for Aqua and Terra. However, for SA $< 160^\circ$, DB-Aqua and DT-Terra showed different behaviors, with DT-Aqua displaying a slight negative bias trend and DT-Terra showing a slight positive bias trend. Similar behavior is observed for DT 3 and 10km, with a dip in the interval between $130^\circ < SA < 150^\circ$, particularly noticeable for DT_{10km}-Aqua. Additionally, a positive bias trend is observed for SA $> 150^\circ$. However, DT_{10km}-Terra displays less sensitivity to changes in SA.

The accuracy of AOD retrieval algorithms using measurements from single-view sun-synchronous orbit sensors is influenced by the anisotropic properties of surface reflectance (MHAWISH et al., 2019). This is attributed to variations in surface brightness in response to the view direction (MHAWISH et al., 2019); according to, in general, the surface tends to be brighter for backscattering geometry (RAA $< 90^\circ$) than for forward scattering (RAA $> 90^\circ$). Very low percentage of matches is found for interval between 70 and 110° , which is related to the MODIS orbit (MHAWISH et al., 2019; QIN et al., 2021b). DB and MAIAC exhibited less dependence on RAA compared to DT. However, even for these algorithms, small variations can be observed depending on RAA. For DB, this variation is particularly evident during backscattering ($30^\circ < RAA < 50^\circ$), and for MAIAC, it is observed during forward scattering ($120^\circ < RAA < 150^\circ$). As mentioned by Mhawish et al. (2019) and Qin et al. (2021b), accounting for surface anisotropy (BRDF) in MAIAC seemed to reduce dependency on RAA. For the DT algorithm, there was a decrease in the positive mean bias as RAA increased for backscattering, while for forward-scattering, the bias initially increased and then decreased.

8.3.5 Spatiotemporal retrieval accuracy

To explore the influence of spatiotemporal factors on AOD retrievals, we analyzed the spatial and temporal distribution accuracy statistics. Figure 8.9 presents the spatial distribution of the correlation coefficient for each product and site AERONET analyzed (spatial distribution of RMSE, Bias, and percentage of retrievals within EE envelope can be assessed in the Appendix D – Figure D.11). Regarding the spatial correlation distribution, consistent results were observed across the algorithms, indicating higher correlations ($R > 0.9$) in regions traditionally known for biomass burning emissions and where higher aerosol loads are typically observed (Figure 8.2 Figure 8.3).

Figure 8.9 – Correlation coefficient (R) for MAIAC, DT, and DB algorithms in each AERONET site.



Through Figure 8.10, which presents the performance statistics of each algorithm for each AERONET site, heterogeneity is observed regarding the algorithms' performance in correlation with surface data. However, it can be observed that DB achieved the highest correlations in 11 AERONET sites. The heterogeneous behavior is also maintained when evaluating RMSE, although DB and MAIAC exhibit the lowest RMSE values (Figure D.11). In terms of bias, MAIAC shows the best overall performance, being the algorithm with the highest performance in 8 out of 17 AERONET sites. The superior performance of MAIAC and DB is also evident when observing the percentage of retrievals within the EE envelope (Figure 8.10 and Figure D.11). It is clear that the lower performance of DT is attributed to overestimation of AOD

values, as indicated by the high bias values, particularly in the northern regions (Figure D.11), once again demonstrating DT's tendency to overestimate high AOD values.

Figure 8.10 – Statistical results for MAIAC, DT, and DB algorithms in each AERONET site.

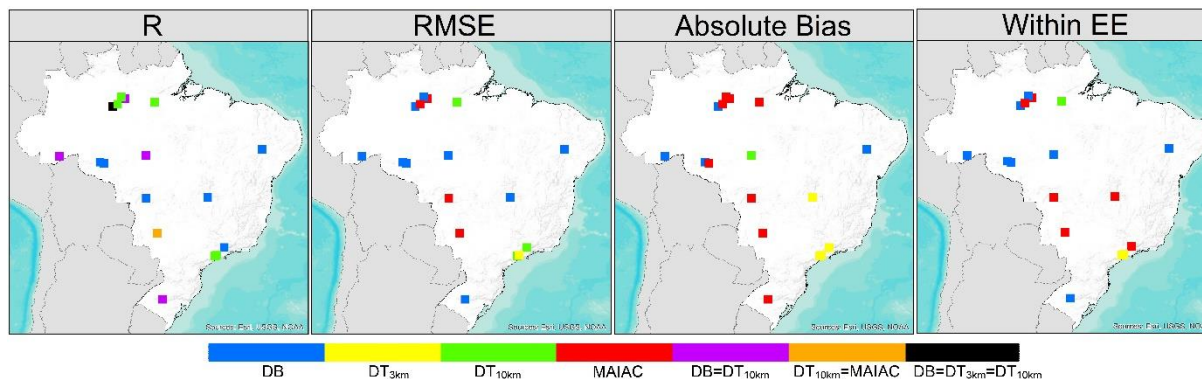
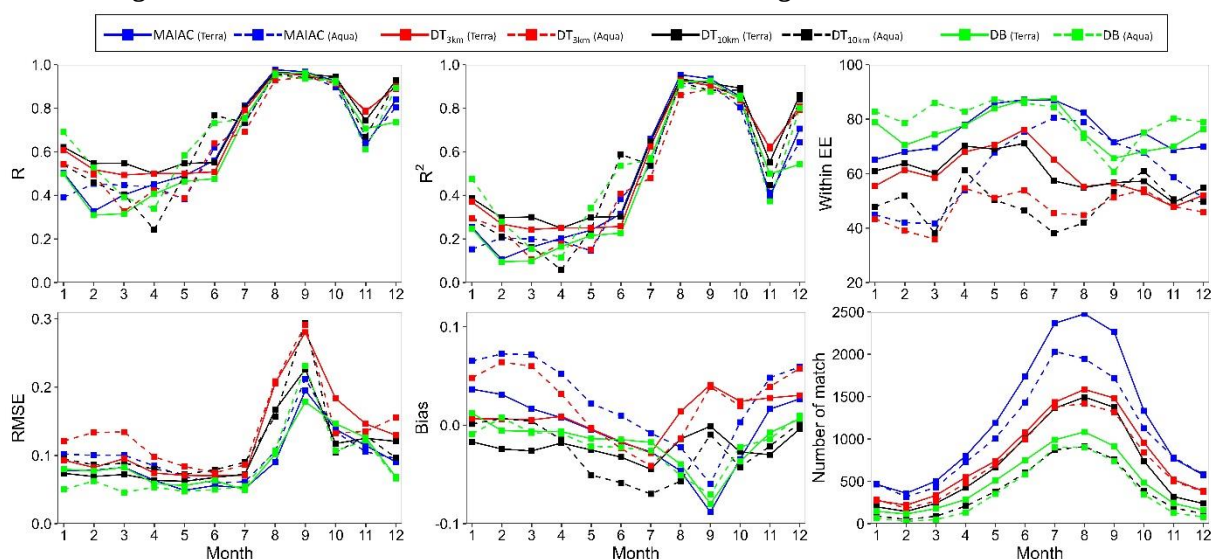


Figure 8.11 presents the monthly validation results for the evaluated products. Overall, it is observed that the highest values of R and R^2 are obtained during periods traditionally associated with biomass burning (MARTIN et al., 2010; POPE et al., 2020; SILVEIRA et al., 2020), once again highlighting that the strongest correlations between satellite and surface measurements occur when high aerosol loads are present in the atmosphere. However, high aerosol loads during biomass burning emissions also lead to more significant errors and biases, with errors reaching up to two orders of magnitude.

Figure 8.11 – Statistical results for MAIAC, DT, and DB algorithms in different months.



8.4 Conclusions

This study provides the first comprehensive validation and comparison of four AOD-MODIS products [MAIAC, DT (3 and 10km), and DB] over Brazil. We assess their overall accuracy, dependency on land cover and view geometry, as well as their spatiotemporal retrieval accuracy and completeness. These validation results serve as valuable guidance for users in utilizing the products effectively and with increased caution. The main findings of this study were:

- 1) The spatial distribution of AOD across all the products revealed consistent patterns with variations in magnitude. The Amazon region exhibited higher AOD values, particularly in areas historically associated with elevated aerosol loads during the biomass burning season.
- 2) Due to MODIS being a passive sensor, AOD retrievals are greatly affected by the presence of clouds, which are more prevalent in the northern region and along the northeastern coast of Brazil. The combination of cloud cover and other intrinsic algorithm characteristics results in a significantly low percentage of data retrieval for the study area during specific periods of the year, particularly the rainy season, with values below 20%.
- 3) In terms of overall accuracy, MAIAC and DB algorithms demonstrated higher accuracy compared to the DT. Generally, DT tended to underestimate the low values of AOD and overestimate high values. In contrast, MAIAC and DB exhibited the opposite behavior, tending to overestimate low values and underestimate high values.
- 4) The algorithms showed dependency on the land cover type. Overall, surfaces classified as Urban and Savannah exhibited the poorest accuracies for all products. Over forest areas, DT tended to show slightly better results compared to MAIAC. MAIAC and DB presented the best results for the other land cover classes.
- 5) The four products exhibited varying degrees of dependency on SZA, SA, and RAA. The influence of VZA on retrieval accuracy was found to be minimal. Among them, DB demonstrated the least angular dependency, followed by MAIAC.
- 6) Based on the spatiotemporal distribution of accuracy statistics, it is evident that the strongest correlations between satellite-derived and ground-measured AOD data are observed during the biomass burning season. This finding supports the information provided by Hyer, Reid, and

Zhang (2011), which suggests that good validation statistics in Brazil are mainly influenced by the presence of high aerosol loads during intense biomass burning episodes.

9 CONCLUSIONS

The present work primarily aimed to assess the impact of mobility restrictions implemented during the COVID-19 pandemic on air pollutant concentrations in Brazil. Additionally, the study aimed to investigate the capability of remote sensing data to capture the observed variations in pollutant levels. To achieve this objective, the research was divided in 4 general phases: (1) the first phase was focused on comprehending how the diverse state-level public policies implemented to reduce the transmission of COVID-19 (isolation, quarantine, and mobility restrictions) affected the regional air quality in Brazil. (2) The second phase investigated the localized effects of mobility restrictions on air quality within the most affected state in Brazil, São Paulo. Additionally, it aimed to illustrate how atmospheric conditions can influence temporal comparisons in air quality assessments. (3) The third phase focused on assessing the capability of remote sensing data in capturing variations in air pollution in the lower atmosphere. Specifically, it evaluated the potential of TROPOMI and MAIAC-AOD data as predictors of ground-level NO₂ and PM concentrations, respectively. Lastly, due to the low relationship between MAIAC-AOD and PM observed in phase 3, (4) in the fourth phase, we conducted a validation and comparison of all AOD products derived from MODIS data [MAIAC, DT (3 and 10 km), and DB]. This phase aimed to investigate the factors that can influence the retrieval accuracy of these algorithms. The main conclusions are presented as follows:

For Brazil, the results revealed a significant decrease in CO, NO₂, PM₁₀, and PM_{2.5} concentrations, mainly for a short period when the isolation was more restrictive (first 30 days after implementing mobility restrictions). Contrary trends were observed for O₃, exhibiting significant concentration increases for all states under evaluation. The O₃ increases during mobility restrictions were observed worldwide, suggesting potential changes in the chemical formation regime of O₃ due to decreases in NO_x. For longer periods (>90 days), there is a decrease in the magnitude of variations in air pollution levels. This can be attributed to the “relaxation” of isolation measures implemented by state governments, leading to increased population mobility and, consequently, higher emissions of air pollutants. Finally, the findings of this study confirmed that the mobility restrictions implemented due to COVID-19 led to a significant decrease in the concentrations of various atmospheric pollutants, even during more extended periods, although the effects were less pronounced compared to shorter time frames.

Furthermore, the results demonstrated that densely populated and industrialized regions observed the most significant impacts.

Based on the findings of Phase 1, which highlighted the significant impacts of mobility restrictions during shorter periods and in densely populated and industrialized regions, a chapter of this thesis was dedicated to investigate the localized effects of mobility restrictions on air pollution levels. The state of São Paulo was chosen as the study area for this phase due to its high population density, level of industrialization, and well-established air quality monitoring network, providing suitable conditions for conducting such studies. By comparing the data from 2019 and 2020 with the historical averages of pollutants, a consistent reduction trend in most air pollutants was observed. This decline can be primarily attributed to the implementation of public policies aimed at controlling and reducing emissions. As for the period during pandemics, the findings for the State of São Paulo revealed a diverse pattern of air quality during mobility restrictions, indicating a heterogeneously localized behavior. Reinforcing the findings observed in Phase 1, it was demonstrated that the most significant decreases in pollutant levels were observed for primary pollutants, such as CO and NO_x, while increases in O₃ concentrations were observed. Spatially, some peripheral industrial areas showed an increase in pollutant concentrations even during the mobility restrictions, which may be associated with specific industrial activities. Non-significant variations or even increases in PM concentrations were observed at various monitoring stations, indicating a potential impact of biomass burning emissions. In contrast to studies conducted in other regions worldwide, the impact of mobility restrictions in São Paulo was less pronounced. This could be attributed to the absence of stricter social isolation measures (e.g., lockdown) and the influence of atmospheric conditions prevailing during mobility restrictions. Indeed, the findings of this research demonstrate that an above-average number of fire incidents in the months following the pandemics, together with the abnormal distribution of rainfall, have contributed to the lack of clear evidence indicating improved air quality resulting from mobility restrictions. Therefore, this study recommends considering atmospheric conditions and regionalization when evaluating the effectiveness of emission control measures.

The results obtained in the first two phases of the research highlight the tangible impact of policies aimed at reducing air pollutant concentrations. This real-world experiment served as an open-air laboratory, providing valuable insights. Namely, it was evident that reducing emissions from both mobile and stationary sources holds substantial potential for significantly

lowering primary pollutant levels. The results also highlight that biomass burning in the Amazon, Cerrado, and, more recently, in the Pantanal regions can impact pollutant concentrations even in major urban centers. Addressing this issue necessitates specific measures aimed at reducing fire incidents within these biomes. Furthermore, an important observation was the increase in O_3 levels despite reductions in primary pollution sources. O_3 , alongside $PM_{2.5}$, poses significant health risks. Hence, comprehending the intricacies related to emissions, the spatial distribution of O_3 precursors, and the underlying mechanisms associated with its atmospheric formation is essential for implementing effective control strategies to safeguard human health.

Given the observed variations in atmospheric pollutant concentrations during Phases 1 and 2, the capability of remote sensors to capture these changes was evaluated, as remote sensing has been extensively utilized for this specific purpose. As mentioned, São Paulo has the most robust air quality monitoring network, making it the chosen region for conducting Phase 3. The findings of this study demonstrate that TROPOMI exhibits strong predictive capabilities for NO_2 concentrations at ground-level. The correlations between the tropospheric density of NO_2 and ground-level concentrations exceeded 0.6 for all analyzed periods. Moreover, tropospheric NO_2 data proved to be valuable in capturing the temporal behaviors of the pollutant during the mobility restrictions period. Locally, it was particularly notable that stations in urbanized and/or industrialized regions, where NO_2 sources are more concentrated, showed strong correlations. Based on the spatially continuous data from TROPOMI, the conclusions drawn in Phase 2 were further confirmed, highlighting that the most substantial changes in air pollution during mobility restrictions occurred in regions with economic vocations characterized as industrial or in the process of industrialization. In contrast, the countryside, where agricultural activities dominate, experienced smaller reductions or even increases in NO_2 . On the other hand, MAIAC-AOD data showed a low potential for directly predicting PM concentrations at ground level. The study highlighted the limitations of using satellite data as ground truth due to data acquisition difficulties and the presence of different aerosol types in the atmosphere.

AOD data play a crucial role in estimating $PM_{2.5}$ levels, particularly in the context of epidemiological studies. Therefore, recognizing the significant impact of $PM_{2.5}$ on human health and considering the limitations of PM predictions based on MAIAC-AOD observed in Phase 3, extensive efforts were undertaken to identify the most suitable AOD product for Brazil. The results of this phase highlight the strong influence of land cover type, view geometry,

geographical location, and time period on the performance of AOD retrieval algorithms (MAIAC, DT, and DB). While these algorithms exhibit a considerable dependency on geographical location and period, they demonstrate similar behavior overall, with higher accuracy observed in regions and periods characterized by significant aerosol loads resulting from biomass burning emissions. However, the algorithms display more varied performance with respect to the land cover type and view geometry. Specifically, DT performs better in forested and urban areas, while MAIAC and DB show greater effectiveness in other land cover classes. Moreover, MAIAC and DB exhibit lower dependency on the analyzed angular variations (SZA, SA, and RAA), indicating greater consistency in the AOD algorithms. Notably, the urban and savanna land cover classes demonstrate lower accuracy values, suggesting challenges in accurately retrieving AOD in brighter and more heterogeneous surfaces.

Finally, the findings in Phases 3 and 4 demonstrate the potential of TROPOMI data and underscore the importance of further research on the relationship between AOD and PM. It is noteworthy that the evaluated AOD retrieval algorithms generally encountered more significant challenges in capturing spatial variations in urban areas. Given that PM measurements are primarily conducted in urban areas, it is expected that the lower quality of the retrieved AOD for these locations may not directly reflect the variations in ground-level concentrations of PM. Hence, there is a need for studies focused on improving the quality of remote sensing data for PM_{2.5} estimations.

From the results obtained and the limitations of this study, some future works are suggested:

- It is recommended that future studies assess the impacts of mobility restrictions in other Brazilian states to highlight local characteristics that may either facilitate or impede improvements in air quality due to reduced emissions of air pollutants. During the development of Phase 1, we utilized all the available data for Brazil. However, although measurements for 2020 were available in some states, the datasets were not accessible to the public during the data analysis.
- To comprehensively quantify how wildfire emissions in the northern region of the country impact atmospheric pollutant concentrations in the southern and southeastern regions.

- To dissociate and quantify the impacts on air quality induced by atmospheric conditions from those occurring due to mobility restrictions.
- To evaluate the predictive capacity of the remaining AOD products for PM estimations. It is particularly suggested to assess DT, which exhibited superior performance in urban areas and offers a product with a relatively higher spatial resolution (3km) compared to the 10km resolution of the other untested algorithms.
- To develop a method (empirical or physical) capable of overcoming the difficulty of algorithms in retrieving accurate AOD data in urban areas in Brazil.
- Finally, in addition to evaluating the AOD products, it is suggested to assess the predictive potential of MODIS Level-1 satellite images, which have demonstrated their capability for estimating PM in other locations (IMANI, 2021).

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**APPENDIX A – IMPACTS OF STRATEGIC MOBILITY RESTRICTIONS
POLICIES DURING 2020 COVID-19 OUTBREAK ON BRAZIL’S REGIONAL AIR
QUALITY**

Table A.1 - IEMA air quality monitoring stations for the states of Espírito Santo (ES), Paraná (PR), Rio de Janeiro (RJ), Rio Grande do Sul (RS) e São Paulo (SP).

State	Lat	Lon	Station name	Station Code	Organization	Air pollutant
ES	-20.31	-40.29	Enseada do Suá	ES03	IEMA - ES	CO, PM ₁₀
	-20.19	-40.26	Laranjeiras	ES05	IEMA - ES	CO
	-20.35	-40.32	Vila Velha	ES07	IEMA - ES	CO, PM ₁₀ , O ₃
PR	-25.57	-49.38	CSN	PR03	IAT - PR	NO ₂ , O ₃
	-25.55	-49.39	REPAR	PR06	IAT - PR	CO, NO ₂ , O ₃
RJ	-22.54	-44.20	BM - Bocaininha	RJ57	INEA - RJ	PM ₁₀
	-22.51	-44.20	BM - Vista Alegre	RJ59	INEA - RJ	PM ₁₀
	-22.71	-43.27	DC - Campos Elíseos	RJ20	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.67	-43.29	DC - Jardim Primavera	RJ22	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.71	-43.31	DC - Pilar	RJ21	INEA - RJ	NO ₂ , PM ₁₀ , O ₃
	-22.74	-43.31	DC - São Bento	RJ23	INEA - RJ	NO ₂ , PM ₁₀ , O ₃
	-22.78	-43.29	DC - Vila São Luiz	RJ24	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.70	-42.88	Itb - Porto das Caixas	RJ15	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.67	-42.79	Itb - Sambaetiba	RJ16	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.90	-43.87	Itg - Coroa Grande	RJ18	INEA - RJ	PM ₁₀
	-22.87	-43.77	Itg - Monte Serrat	RJ28	INEA - RJ	NO ₂ , PM ₁₀ , O ₃
	-22.29	-44.34	Itt - Campo Alegre	RJ52	INEA - RJ	PM ₁₀
	-22.67	-43.59	Jp - Engenheiro Pedreira	RJ26	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.31	-41.75	Mc - Cabiúnas	RJ71	INEA - RJ	CO, NO ₂ , O ₃
	-22.35	-41.96	Mc - Fazenda Aires	RJ68	INEA - RJ	CO, NO ₂ , O ₃
	-22.31	-41.88	Mc - Fazenda Severina	RJ70	INEA - RJ	CO
	-22.38	-41.81	Mc - Pesagro	RJ69	INEA - RJ	CO
	-22.93	-43.91	Mt - Itacuruçá	RJ19	INEA - RJ	PM ₁₀
	-22.83	-43.61	NI - Jardim Guandu	RJ80	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.42	-44.29	PR - Porto Real	RJ50	INEA - RJ	CO, NO ₂
	-22.89	-43.47	Pref - Bangu	RJ90	Prefeitura RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.89	-43.56	Pref - Campo Grande	RJ91	Prefeitura RJ	CO, PM ₁₀ , O ₃
	-22.91	-43.18	Pref - Centro	RJ85	Prefeitura RJ	PM ₁₀ , O ₃
	-22.83	-43.33	Pref - Irajá	RJ89	Prefeitura RJ	NO ₂ , PM ₁₀ , PM _{2.5} , O ₃
	-23.00	-43.63	Pref - Pedra de Guaraíba	RJ92	Prefeitura RJ	PM ₁₀ , O ₃
	-22.90	-43.22	Pref - São Cristóvão	RJ87	Prefeitura RJ	CO, PM ₁₀ , O ₃
	-22.92	-43.23	Pref - Tijuca	RJ88	Prefeitura RJ	CO, PM ₁₀ , O ₃
	-22.40	-44.32	Qt - Bom Retiro	RJ51	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.77	-43.11	RJ - Ilha de Paqueta	RJ11	INEA - RJ	O ₃
	-22.80	-43.18	RJ - Ilha do Governador	RJ10	INEA - RJ	O ₃
	-22.28	-44.29	Rs - Cidade Alegria	RJ53	INEA - RJ	CO, PM ₁₀ , O ₃
	-22.89	-43.72	SC - Adalgisa Nery	RJ25	INEA - RJ	NO ₂ , O ₃
	-22.84	-43.71	Sp - Piranema	RJ17	INEA - RJ	CO
	-22.52	-44.13	VR - Belmonte	RJ47	INEA - RJ	CO, NO ₂ , PM ₁₀ , O ₃
	-22.50	-44.12	VR - Retiro	RJ48	INEA - RJ	CO, O ₃
	-22.52	-44.11	VR - Santa Cecília	RJ49	INEA - RJ	CO, NO ₂ , O ₃
RS	-29.92	-50.99	Gravataí - Condomínio Jardim Timbaúva	RS05	FEPAM - RS	PM ₁₀ , O ₃
	-30.11	-51.33	Guaíba - Parque 35	RS07	FEPAM - RS	CO, NO ₂ , PM ₁₀ , O ₃
SP	-21.19	-50.44	Araçatuba	SP02	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-21.78	-48.19	Araraquara	SP03	CETESB - SP	PM ₁₀ , O ₃
	-22.33	-49.09	Bauru	SP06	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-22.90	-47.06	Campinas - Centro	SP07	CETESB - SP	CO, PM ₁₀
	-22.87	-47.06	Campinas - Taquaral	SP08	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-22.95	-47.12	Campinas - Vila União	SP09	CETESB - SP	O ₃
	-23.67	-46.78	Capão Redondo	SP11	CETESB - SP	PM ₁₀ , O ₃
	-23.53	-46.84	Carapicuíba	SP12	CETESB - SP	CO, PM ₁₀ , O ₃
	-21.14	-48.98	Catanduva	SP13	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-23.55	-46.67	Cerqueira César	SP14	CETESB - SP	NO ₂ , PM ₁₀
	-23.57	-46.74	Cid.Universitária USP - IPEN	SP15	CETESB - SP	PM _{2.5} , O ₃
	-23.62	-46.66	Congonhas	SP16	CETESB - SP	CO, NO ₂ , PM ₁₀ , PM _{2.5}
	-23.88	-46.42	Cubatão - Centro	SP18	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-23.83	-46.37	Cubatão - Vale do Mogi	SP19	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-23.85	-46.39	Cubatão - Vila Parisi	SP20	CETESB - SP	NO ₂ , PM ₁₀
	-23.69	-46.61	Diadema	SP21	CETESB - SP	PM ₁₀ , O ₃
	-23.78	-46.70	Grajaú - Parelheiros	SP43	CETESB - SP	PM ₁₀ , PM _{2.5} , O ₃
	-22.82	-45.19	Guaratinguetá	SP04	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-23.46	-46.52	Guarulhos - Paço Municipal	SP24	CETESB - SP	PM ₁₀ , PM _{2.5} , O ₃
	-23.44	-46.41	Guarulhos - Pimentas	SP25	CETESB - SP	NO ₂ , PM ₁₀ , PM _{2.5} , O ₃
	-23.59	-46.66	Ibirapuera	SP26	CETESB - SP	CO, NO ₂ , PM _{2.5} , O ₃
	-23.68	-46.68	Interlagos	SP27	CETESB - SP	PM ₁₀ , O ₃
	-23.50	-46.42	Itaim Paulista	SP28	CETESB - SP	PM ₁₀ , PM _{2.5} , O ₃
	-23.58	-46.47	Itaquera	SP29	CETESB - SP	O ₃
	-22.30	-48.57	Jaú	SP33	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
	-23.19	-46.90	Jundiaí	SP34	CETESB - SP	PM ₁₀ , O ₃
	-22.56	-47.41	Limeira	SP36	CETESB - SP	NO ₂ , PM ₁₀ , O ₃

-23.52	-46.74	Marg. Tietê - Pte dos Remédios	SP37	CETESB - SP	CO, NO ₂ , PM ₁₀ , PM _{2.5}
-22.20	-49.96	Marília	SP38	CETESB - SP	NO ₂
-23.67	-46.47	Mauá	SP39	CETESB - SP	PM ₁₀ , O ₃
-23.55	-46.60	Mooca	SP40	CETESB - SP	CO, O ₃
-23.48	-46.69	Nossa Senhora do Ó	SP41	CETESB - SP	PM ₁₀ , O ₃
-23.53	-46.79	Osasco	SP42	CETESB - SP	CO, NO ₂ , PM ₁₀
-23.54	-46.63	Parque Dom Pedro II	SP44	CETESB - SP	CO, NO ₂ , PM ₁₀ , PM _{2.5} , O ₃
-22.72	-47.15	Paulínia	SP45	CETESB - SP	PM ₁₀ , O ₃
-23.46	-46.77	Pico do Jaraguá	SP49	CETESB - SP	NO ₂ , PM _{2.5} , O ₃
-23.56	-46.70	Pinheiros	SP50	CETESB - SP	CO, NO ₂ , O ₃
-22.70	-47.65	Piracicaba	SP51	CETESB - SP	NO ₂ , PM ₁₀ , PM _{2.5} , O ₃
-22.12	-51.41	Presidente Prudente	SP53	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
-22.46	-47.54	Santa Gertrudes	SP57	CETESB - SP	PM ₁₀
-23.51	-46.63	Santana	SP59	CETESB - SP	O ₃
-23.65	-46.71	Santo Amaro	SP60	CETESB - SP	CO, PM ₁₀ , O ₃
-23.64	-46.49	Santo André - Capuava	SP61	CETESB - SP	PM ₁₀ , O ₃
-23.98	-46.30	Santos - Ponta da Praia	SP64	CETESB - SP	NO ₂ , PM ₁₀ , PM _{2.5} , O ₃
-23.70	-46.55	São Bernardo do Campo - Centro	SP65	CETESB - SP	CO, NO ₂ , PM _{2.5} , O ₃
-23.67	-46.58	São Bernardo do Campo - Paulicéia	SP66	CETESB - SP	PM ₁₀
-23.62	-46.56	São Caetano do Sul	SP67	CETESB - SP	CO, NO ₂ , PM ₁₀ , O ₃
-20.78	-49.40	São José do Rio Preto	SP69	CETESB - SP	NO ₂ , PM ₁₀ , PM _{2.5}
-23.22	-45.89	São José dos Campos	SP70	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
-23.22	-45.89	São José dos Campos - Jardim Satélite	SP71	CETESB - SP	CO, NO ₂ , PM ₁₀ , PM _{2.5} , O ₃
-23.50	-47.48	Sorocaba	SP73	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
-23.61	-46.76	Taboão da Serra	SP75	CETESB - SP	CO, PM ₁₀
-23.36	-47.87	Tatuí	SP77	CETESB - SP	NO ₂ , PM ₁₀ , O ₃
-23.03	-45.58	Taubaté	SP78	CETESB - SP	NO ₂ , PM ₁₀ , PM _{2.5} , O ₃

Table A.2 - Averages concentrations and standard deviation for the analyzed air pollutants ($\mu\text{g m}^{-3}$) before (2015-2019) and during 2020 considering different temporal intervals; Changes (%) in air pollutant concentrations and mobility averaged for the five selected Brazilian States analyzed.

Air pollutant		15 days			30 days			90 days			180 days			290 days		
		2015-2019	2020	Mobility	2015-2019	2020	Mobility	2015-2019	2020	Mobility	2015-2019	2020	Mobility	2015-2019	2020	Mobility
ES	CO	321.6 ± 40.8 (-18.8%)	261.3 ± 51.1		343.8 ± 47.4 (-24.4%)	259.9 ± 43.2		368.4 ± 50.5 (-17.2%)	305.1 ± 70.5		369.6 ± 48.0 (-14.0%)	317.9 ± 65.8		363.0 ± 45.3 (-10.5%)	324.9 ± 62.8	
	PM ₁₀	21.3 ± 2.2 (-21.6%)	16.7 ± 3.7	-52.3%	21.0 ± 2.8 (-23.3%)	16.1 ± 3.5	-56.9%	24.3 ± 4.7 (-28.4%)	17.4 ± 4.3	-54.7%	24.9 ± 4.4 (-28.9%)	17.7 ± 4.4	-33.5%	25.4 ± 4.5 (-26.4%)	18.7 ± 5.2	-35.1%
	O ₃	34.4 ± 2.6 (-9.6%)	37.7 ± 7.4		33.4 ± 2.9 (5.1%)	35.1 ± 7.9		33.5 ± 5.7 (11.3%)	37.3 ± 8.6		33.4 ± 7.2 (27.5%)	42.6 ± 10.8		34.5 ± 8.5 (24.3%)	42.9 ± 13.3	
PR	CO	202.9 ± 43.7 (-41.1%)	119.6 ± 57.5		208.6 ± 63.6 (-45.1%)	114.6 ± 55.4		236.7 ± 105.8 (-29.5%)	166.9 ± 98.1		265.7 ± 97.9 (-30.6%)	184.3 ± 99.5		237.9 ± 90.0 (-27.1%)	173.5 ± 95.1	
	NO ₂	21.8 ± 2.1 (-5.0%)	20.7 ± 5.5	-44.9%	23.5 ± 2.3 ± (-16.6%)	19.6 ± 5.0	-48.9%	24.7 ± 4.3 (8.1%)	26.7 ± 10.6	-42.5	27.9 ± 6.5 (7.2%)	29.9 ± 11.6	-37.6%	25.8 ± 6.5 (4.7%)	27.0 ± 11.5	-29.1%
	O ₃	17.4 ± 1.8 (-46.0%)	25.4 ± 6.1		18.7 ± 3.4 (43.9%)	26.9 ± 5.5		16.2 ± 3.9 (74.7%)	28.3 ± 10.3		17.7 ± 4.7 (57.1%)	27.8 ± 9.7		21.2 ± 6.4 (40.1%)	29.7 ± 9.8	
RJ	CO	439.6 ± 17.8 (-16.5%)	367.2 ± 78.7		451.8 ± 35.9 (-22.6%)	349.7 ± 61.4		474.5 ± 52.6 (-12.2%)	416.6 ± 111.2		498.8 ± 59.6 (-14.2%)	427.8 ± 92.8		500.1 ± 56.9 (-12.6%)	436.9 ± 88.2	
	NO ₂	15.7 ± 1.0 (-28.0%)	11.3 ± 2.4		16.4 ± 1.2 (-34.1%)	10.8 ± 1.9		17.6 ± 1.8 (-23.9%)	13.4 ± 3.4		19.0 ± 2.4 (-25.3%)	14.2 ± 3.3		18.1 ± 2.5 (-22.1%)	14.1 ± 3.1	
	PM ₁₀	29.4 ± 3.5 (-19.7%)	23.6 ± 5.7	-51.3%	31.4 ± 4.0 (-23.6%)	24.0 ± 5.3	-54.2%	33.1 ± 5.2 (-11.2%)	29.4 ± 9.4	-53.3%	36.6 ± 7.5 (-10.4%)	32.8 ± 11.3	-42.9%	34.5 ± 7.6 (-9.6%)	31.2 ± 11.5	-32.9%
	PM _{2.5}	14.3 ± 2.2 (-14.7%)	16.4 ± 5.7		14.6 ± 2.1 (8.2%)	15.8 ± 4.5		16.3 ± 4.6 (9.8%)	17.9 ± 11.3		20.0 ± 7.2 (-1.0%)	19.8 ± 11.7		17.9 ± 7.2 (0.6%)	18.0 ± 11.3	
	O ₃	31.5 ± 2.3 (-17.5%)	37.0 ± 6.4		32.6 ± 2.3 (10.7%)	36.1 ± 5.7		29.6 ± 3.6 (19.3%)	35.3 ± 5.7		31.0 ± 4.7 (14.8%)	35.6 ± 7.5		34.7 ± 6.8 (10.7%)	38.4 ± 8.8	
RS	CO	994.5 ± 352.6 (-57.4%)	423.9 ± 52.7		890.3 ± 277.2 (-53.1%)	417.3 ± 56.3		884.7 ± 317.5 (-35.1%)	574.5 ± 316.0		926.9 ± 271.5 (-29.5%)	653.8 ± 329.4		803.4 ± 384.4 (-25.4%)	599.6 ± 297.5	
	NO ₂	7.2 ± 1.6 (-36.1%)	9.8 ± 3.5		8.3 ± 1.8 (-2.4%)	8.1 ± 3.8		14.4 ± 3.7 (-9.0%)	13.1 ± 8.6		13.3 ± 4.5 (21.8%)	16.2 ± 10.1		11.7 ± 4.4 (15.4%)	13.5 ± 9.2	
	PM ₁₀	19.0 ± 2.7 (-1.1%)	19.2 ± 6.2	-48.8%	20.8 ± 4.2 (-7.7%)	19.2 ± 5.4	-52.0%	20.8 ± 5.0 (9.1%)	22.7 ± 10.8	-45.6%	23.3 ± 7.1 (-1.3%)	23.0 ± 11.9	-41.1%	22.7 ± 6.5 (-7.0%)	21.1 ± 10.6	-31.7%
	O ₃	24.3 ± 1.9 (-37.4%)	33.4 ± 10.6		25.9 ± 3.4 (26.3%)	32.7 ± 8.1		23.8 ± 4.0 (18.5%)	28.2 ± 7.6		25.3 ± 5.2 (22.5%)	31.0 ± 9.2		29.1 ± 6.8 (13.1%)	32.9 ± 8.9	
SP	CO	651.1 ± 46.5 (-37%)	410.0 ± 155.5		659.7 ± 45.2 (-39.5%)	399.1 ± 130.6		± 744.1 ± 150.8 (-20.2%)	594.1 ± 325.5		815.0 ± 180.4 (-16.5%)	680.7 ± 315.6		754.2 ± 170.1 (-10.9%)	671.9 ± 293.1	
	NO ₂	22.7 ± 1.8 (-24.2%)	17.2 ± 5.5		23.6 ± 2.1 (-28.8%)	16.8 ± 4.7		26.9 ± 4.2 (-14.5%)	23.0 ± 9.2		30.3 ± 5.6 (-14.9%)	25.8 ± 9.2		28.0 ± 6.1 (-11.4%)	24.8 ± 9.1	
	PM ₁₀	23.9 ± 2.6 (-10.0%)	21.5 ± 4.7	-47%	26.7 ± 4.2 (-13.9%)	23.0 ± 5.4	-51.7%	28.9 ± 4.6 (-1.0%)	29.2 ± 10.9	-50.6%	34.0 ± 7.7 (-3.2%)	32.9 ± 13.1	-42.2%	31.2 ± 8.4 (-0.6%)	31.0 ± 15.1	-32.6%
	PM _{2.5}	12.2 ± 1.8 (-12.3%)	10.7 ± 3.5		13.7 ± 2.7 (-20.4%)	10.9 ± 3.2		15.6 ± 3.2 (-4.5%)	14.9 ± 6.9		18.2 ± 4.5 (-10.4%)	16.3 ± 7.3		16.8 ± 4.7 (-7.1%)	15.6 ± 8.1	
	O ₃	36.5 ± 2.6 (-15.9%)	42.3 ± 5.1		38.3 ± 3.9 (15.4%)	44.2 ± 6.0		32.1 ± 7.1 (20.2%)	38.6 ± 8.7		33.4 ± 6.9 (16.5%)	38.9 ± 10.0		35.6 ± 9.2 (22.2%)	43.5 ± 11.9	

Note: The intervals shown correspond to the number of days counted from March 16 2020, and the corresponding period for 2015-2019: 15 days (March 16 to March 30), 30 days (March 16 to April 14), 90 days (March 16 to June 13), 180 days (March 16 to September 11) and 290 days (March 16 to December 31).

Figure A.1 - Synoptic area (A, black dashed line), box plot of Convective Available Potential Energy (B, CAPE) and Precipitable Water (C, PWater) for the study area.

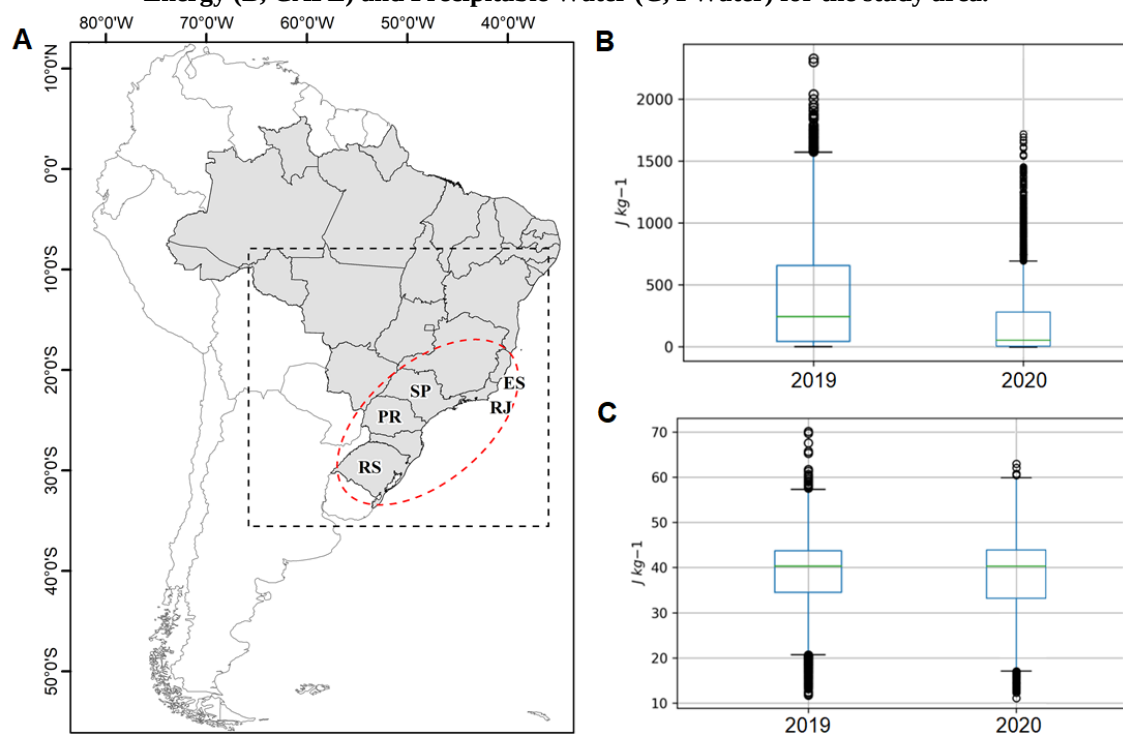


Figure A.2 - Fine particulate matter (PM_{2.5}) concentration annual behavior for the period before (yellow) and during (green) the COVID-19 pandemic for São Paulo (SP) and Rio de Janeiro (RJ) states. The dashed line indicates the date March 16 2020, representing the beginning of Brazil's mobility restrictions.

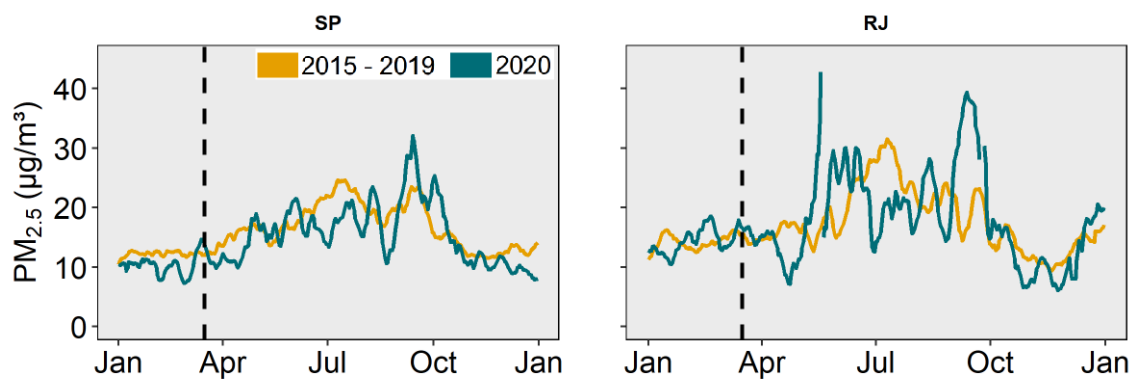


Figure A.3 - Accumulated precipitation between January 1 and March 16 from 2015 to 2020 for the study area. The data presented are predicted values by the numerical weather forecast model of the Brazilian National Institute of Meteorology (Instituto Nacional de Meteorologia – INMET, available in: <http://sisdagro.inmet.gov.br/sisdagro/app/monitoramento/bhs/mapaperiodoacum>).

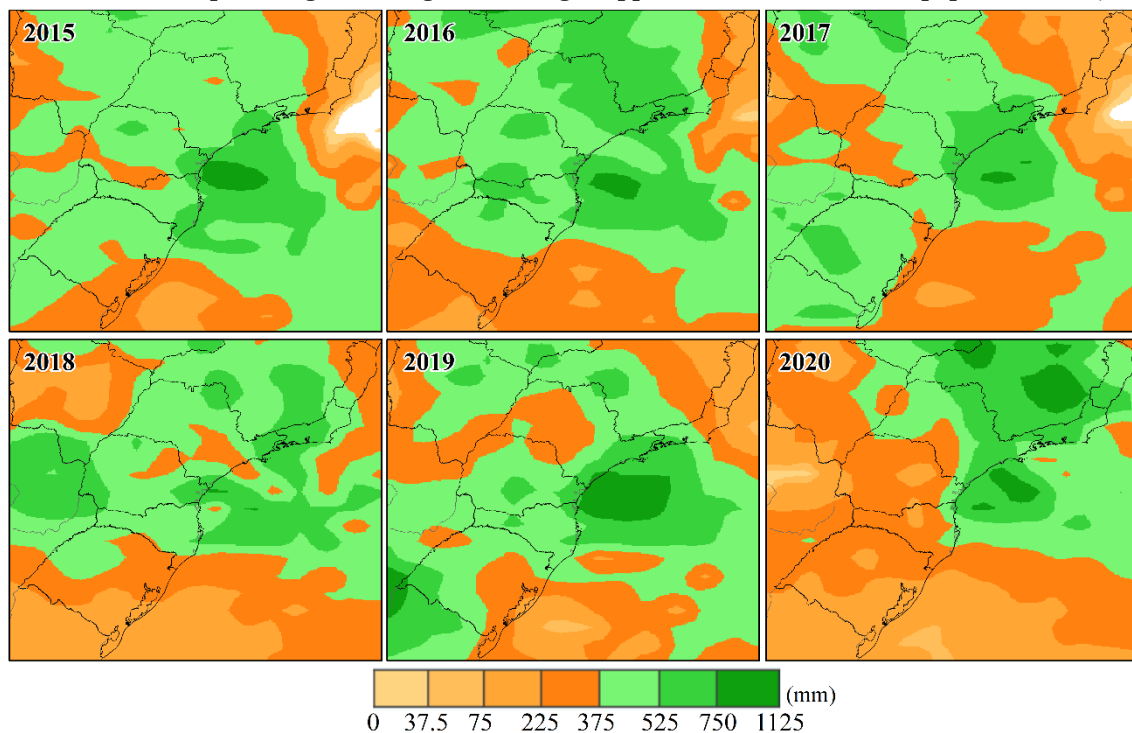
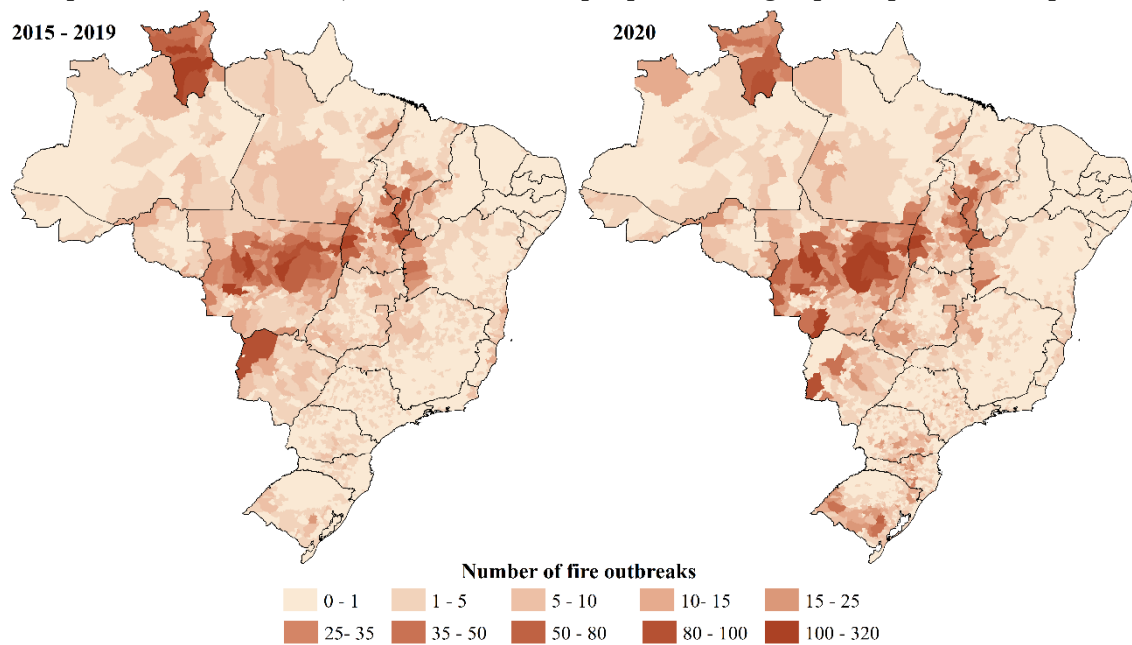


Figure A.4 - Average number of fire outbreaks observed between March 26th and June 11th for the period 2015-2019 and 2020. The data used represent the number of fire outbreaks retrieved daily by the Aqua and Terra satellites (data available at: <http://queimadas.dgi.inpe.br/queimadas/bdqueimadas/>)



**APPENDIX B – HOW MOBILITY RESTRICTIONS POLICY AND ATMOSPHERIC
CONDITIONS IMPACTED AIR QUALITY IN THE STATE OF SÃO PAULO
DURING THE COVID-19 OUTBREAK**

Table B.1 – CETESB air quality monitoring station for São Paulo state used in this research.

HUWRM	Station name	Lon	Lat	Initial date	PM ₁₀	PM _{2.5}	NO _x	NO ₂	NO	SO ₂	CO	O ₃
2	Guaratinguetá	-45.191	-22.802	2017	X		X	X	X			X
	Jacaré	-45.968	-23.294	2011	X							X
	S.José Campos	-45.871	-23.188	2000	X		X ⁷	X ⁷	X ⁷	X		X
	S.José Campos-Jd.Satelite	-45.891	-23.224	2015	X	X	X	X	X		X	X
	Taubaté	-45.576	-23.032	2015	X	X ¹¹	X	X	X			X
	Total				5	2	3	4	4	1	1	5
5	Campinas-Centro	-47.057	-22.903	2000	X						X	
	Campinas-Taquaral	-47.059	-22.875	2015			X	X				X
	Campinas-V.União	-47.119	-22.947	2015		X						X
	Jundiaí	-46.897	-23.192	2008	X							X
	Limeira	-47.414	-22.564	2016	X							
6	Paulínia	-47.155	-22.772	2000	X					X		X
	Piracicaba	-47.65	-22.701	2008	X	X ⁷	X	X	X			X
	Santa Gertrudes	-47.536	-22.46	2014	X	X ¹³	X ¹³	X ¹³	X ¹³			
	Total				6	2	3	3	2	1	1	5
	Capão Redondo	-46.78	-23.668	2012	X							X
7	Carapicuíba	-46.836	-23.531	2012	X						X	X
	Cerqueira César	-46.673	-23.554	1998	X		X	X		X	X	
	Congonhas	-46.663	-23.616	1998	X	X ⁶	X	X		X	X	
	Diadema	-46.612	-23.686	1998	X							X ¹
	Grajaú-Parelheiros	-46.697	-23.776	2007	X	X ⁸					X	
6	Guarulhos-Paço Municipal	-46.519	-23.456	2012	X	X ¹²						X
	Guarulhos-Pimentas	-46.41	-23.44	2015			X	X	X	X	X	X
	Ibirapuera	-46.661	-23.592	1998		X ⁹	X	X			X	X
	Interlagos	-46.675	-23.681	2012	X					X		X
	Itaim Paulista	-46.421	-23.502	2012		X ¹⁰	X ¹³	X ¹³	X ¹³	X ¹³		X
6	Marg.Tietê-Pte Remédios	-46.743	-23.519	2012	X	X	X	X	X	X	X	
	Mooca	-46.6	-23.55	2005		X ¹³					X	
	N.Senhora do Ó	-46.692	-23.48	1998	X							
	Osasco	-46.792	-23.527	1998	X	X ¹²	X	X	X	X	X	
	Parque D.Pedro II	-46.628	-23.545	1998		X ¹¹	X	X	X		X	X
7	Pico do Jaraguá	-46.766	-23.456	2016		X		X				
	Pinheiros	-46.702	-23.561	1999			X	X	X		X ³	X
	S.André-Capuava	-46.492	-23.64	1998	X					X ¹⁰		X ²
	S.Bernardo-Centro	-46.546	-23.699	2014		X	X ¹⁰	X	X		X	X
	S.Bernardo-Paulicéia	-46.585	-23.671	1998	X							
10	Santana	-46.629	-23.506	1999		X ¹²						X
	São Caetano do Sul	-46.556	-23.618	1998	X	X ¹³	X	X	X	X		X
	Taboão da Serra	-46.758	-23.609	1998	X							
	Total				15	13	11	13	8	9	12	14
	Cubatão-Centro	-46.418	-23.879	1998	X		X	X		X		
7	Cubatão-V.Parisi	-46.389	-23.849	1998	X		X ⁵	X ⁴	X ⁵	X		
	Santos-Ponta da Praia	-46.301	-23.981	2011	X	X ⁷		X ⁷		X ⁷		X ⁷
	Total				3	1	2	3	1	3	0	1
	Sorocaba	-47.479	-23.502	2000	X			X ³	X ³			X
	Tatuí	-47.871	-23.361	2011	X			X	X			X
	Total				2	0	0	2	2	0	0	2
13	Araraquara	-48.186	-21.783	2008	X			X				
	Bauru	-49.093	-22.327	2008	X		X	X				X
	Jaú	-48.567	-22.299	2008	X		X	X	X			X
	Total				3	0	2	3	1	0	0	2
	Catanduva	-48.983	-21.142	2009	X		X	X				X
15	São José do Rio Preto	-49.398	-20.785	2008	X	X ⁸	X	X	X			
	Total				2	1	2	2	1	0	0	1
	Araçatuba	-50.439	-21.187	2008	X							X
	Total				1	0	0	0	0	0	0	1
	Marília	-49.96	-22.2	2008	X		X	X	X			X
	Total				1	0	1	1	1	0	0	1
22	Presidente Prudente	-51.409	-22.12	2008	X		X	X	X			X
	Total				1	0	1	1	1	0	0	1
TOTAL					39	19	25	32	21	14	14	33

Note: X indicates that pollutant monitoring started in the year stated in the "Initial date" column; X¹ is used to demonstrate that pollutant monitoring began in 1999, X² in 2000, X³ in 2002, X⁴ in 2004, X⁵ in 2009, X⁶ in 2010, X⁷ in 2012, X⁸ in 2013, X⁹ in 2014, X¹⁰ in 2015, X¹¹ in 2016, X¹² in 2017, and X¹³ in 2018.

Table B.2 – Mann-Whitney U test results compare average daily concentrations between the years 2019 and 2020, considering the period before (January 1 to March 15) and during (March 16 to June 30) the COVID-19 outbreak.

Air Pollutants	HUWRM	Station name	Before COVID-19			During COVID-19		
			2019	2020	p-value	2019	2020	p-value
			[Mean±SD]	[Mean±SD]		[Mean±SD]	[Mean±SD]	
PM ₁₀	2	Guaratinguetá	16 ± 6.3	12.8 ± 4.2	0.004	21.7 ± 8.5	20.8 ± 9.3	0.496
		Jacareí	19.3 ± 9.3	16.7 ± 6.1	0.066	21.8 ± 8.9	20.4 ± 8.5	0.747
		S.José Campos	18.1 ± 4.6	17.6 ± 5.2	0.453	22.1 ± 7.5	23.1 ± 7.5	0.292
		S.José Campos-Jd.Satelite	15.5 ± 5.8	14.7 ± 5	0.897	21.6 ± 9.3	21.5 ± 9.3	0.806
		Taubaté	15.5 ± 5.1	13.3 ± 4.8	0.007	20.8 ± 8.6	21.1 ± 9.5	0.996
		Campinas-Centro	20.1 ± 3.6	19 ± 4.6	0.099	25 ± 6.8	23.1 ± 6.4	0.046
	5	Jundiaí	20.1 ± 6.9	16.4 ± 5.7	0.003	26.3 ± 11.5	24 ± 10.3	0.147
		Limeira	15.6 ± 6	20.5 ± 7.8	0.015	35.8 ± 14	34 ± 13.4	0.185
		Paulínia	23.1 ± 6.5	19 ± 5	0.000	31.1 ± 9.6	29.3 ± 10.8	0.057
		Piracicaba	21 ± 5.9	24.2 ± 8.9	0.005	31.9 ± 13.3	41.2 ± 18.4	0.000
		Santa Gertrudes	30.8 ± 13.4	31.5 ± 13.1	0.504	56.7 ± 25.5	45.5 ± 17.9	0.000
		Capão Redondo	-	-	-	26.7 ± 13.7	25.2 ± 15	0.323
	6	Carapicuíba	20.2 ± 7.6	18.8 ± 6.7	0.505	30 ± 15.6	27 ± 15.1	0.064
		Cerqueira César	19.4 ± 7.4	17.1 ± 7.2	0.033	23.5 ± 10.3	22.6 ± 10.4	0.320
		Congonhas	23.2 ± 7.1	23.1 ± 7.8	0.553	31.3 ± 11.8	29.8 ± 13.4	0.160
		Diadema	21.2 ± 5.9	18.6 ± 5.9	0.008	25.6 ± 9.8	24 ± 9.6	0.161
		Grajaú-Parelheiros	25.8 ± 9.5	22.7 ± 11.7	0.201	36.5 ± 21.3	36.7 ± 24.1	0.689
		Guarulhos-Paço Municipal	19 ± 7.7	17.3 ± 8.4	0.221	28.5 ± 13	27 ± 13.6	0.303
		Interlagos	18.7 ± 7	19 ± 9	0.290	29.6 ± 13.3	26 ± 14.2	0.112
		Marg.Tietê-Pte Remédios	27.8 ± 9.1	21.1 ± 8.6	0.000	36.7 ± 16.7	32.8 ± 16.4	0.035
		N.Senhora do Ó	20.5 ± 7.3	18.7 ± 7.3	0.271	28 ± 11.2	28.2 ± 13.1	0.776
		Osasco	33.7 ± 10.5	32.6 ± 12	0.303	45.1 ± 18.5	43.6 ± 18.7	0.519
		S.André-Capuava	20.1 ± 7.2	17.3 ± 7.4	0.010	25.3 ± 10.1	21.5 ± 9	0.004
		S.Bernardo-Paulicéia	20.3 ± 6.6	19.9 ± 7.6	0.559	28.8 ± 13.1	25.4 ± 11.2	0.028
		São Caetano do Sul	24.3 ± 9.3	20.2 ± 9.1	0.059	32.4 ± 12.8	26.3 ± 13	0.106
		Taboão da Serra	22.3 ± 9.9	21.6 ± 9.6	0.974	34.2 ± 17.7	29.2 ± 15.6	0.014
	7	Cubatão-Centro	23.7 ± 7.2	18.9 ± 6.9	0.001	26.1 ± 10.7	25.8 ± 9.1	0.520
		Cubatão-V.Parisi	72 ± 38.6	61.3 ± 46	0.086	76 ± 42.2	86.5 ± 43.2	0.041
		Santos-Ponta da Praia	20.6 ± 6.8	21.7 ± 7.9	0.382	23.5 ± 10.5	37.3 ± 19	0.000
	10	Sorocaba	-	-	-	23.3 ± 10.2	24.5 ± 11.7	0.881
		Tatuí	14.2 ± 4.7	14.7 ± 7.3	0.951	20.9 ± 12.5	23.2 ± 14.8	0.073
	13	Araraquara	14.2 ± 5.4	12.8 ± 4.9	0.275	25.5 ± 11.6	28.4 ± 11.3	0.004
		Bauru	13.8 ± 4.4	13 ± 5.4	0.583	24.8 ± 11.6	24.7 ± 11.4	0.689
		Juá	14.7 ± 4.8	12.7 ± 4.5	0.024	25.9 ± 10.5	22.9 ± 10	0.325
	15	Catanduva	15.8 ± 3.8	16.6 ± 5.5	0.383	35 ± 14.5	34.2 ± 13.1	0.542
		São José do Rio Preto	16.5 ± 5.5	15.7 ± 6	0.450	31.4 ± 13.7	31.8 ± 11.8	0.705
	19	Araçatuba	14.4 ± 4.6	15 ± 6.7	0.601	25.8 ± 10.5	28.3 ± 11.2	0.050
	21	Marília	12.8 ± 4.4	13 ± 5	0.718	20.8 ± 9.1	20.5 ± 8.1	0.804
	22	Presidente Prudente	14.8 ± 5.3	13 ± 5	0.044	20.2 ± 8	19.9 ± 7.8	0.514
PM _{2.5}	2	S.José Campos-Jd.Satelite	9 ± 3.4	7.1 ± 3	0.001	12.4 ± 5.4	12.1 ± 6.2	0.793
		Taubaté	8.9 ± 2.6	7.3 ± 2.7	0.000	11.6 ± 4.2	12.1 ± 6	0.910
	5	Campinas-V.União	11.1 ± 3.9	10.2 ± 3.9	0.150	21.1 ± 10.3	15.9 ± 6.4	0.418
		Piracicaba	8.8 ± 3.4	8.1 ± 3.6	0.074	13 ± 5.2	13.1 ± 5.5	0.920
	6	Santa Gertrudes	12.9 ± 5.3	11.7 ± 4.4	0.093	18.8 ± 6.9	13.3 ± 5.7	0.000
		Congonhas	14.7 ± 4.8	13.2 ± 4.8	0.212	18.6 ± 7.1	16.4 ± 8.7	0.006
		Grajaú-Parelheiros	14.9 ± 5.8	10.5 ± 6.5	0.044	18.9 ± 11.6	16.5 ± 11.3	0.048
		Guarulhos-Paço Municipal	11.4 ± 4.6	10.2 ± 5.2	0.289	16.4 ± 7	15.5 ± 8.3	0.440
		Ibirapuera	9.7 ± 4.5	8.4 ± 4.6	0.050	13.8 ± 7.1	11 ± 6.2	0.003
		Itaim Paulista	17 ± 8.8	9.8 ± 5.8	0.000	19.9 ± 12.9	17.7 ± 12.1	0.076
		Marg.Tietê-Pte Remédios	15.1 ± 5.7	13.3 ± 5.9	0.020	20.5 ± 11	18.7 ± 10	0.066
		Mooca	11.7 ± 4.6	10.8 ± 4.9	0.057	17.8 ± 8.1	14.2 ± 7.4	0.002
		Osasco	16.1 ± 5.9	12.8 ± 5.7	0.001	23.7 ± 10.4	18.7 ± 9.8	0.000
		Parque D.Pedro II	-	-	-	17.5 ± 8	16.4 ± 8	0.235
		Pico do Jaraguá	15.7 ± 7.4	15.5 ± 4.6	0.719	15.4 ± 4.6	11.9 ± 4.4	0.000
		S.Bernardo-Centro	14.1 ± 5.4	13.1 ± 5.1	0.371	16.3 ± 7.1	15.3 ± 7.3	0.201
		Santana	13.3 ± 4.4	11 ± 6.1	0.012	17.7 ± 6.6	15.2 ± 7.4	0.006
		São Caetano do Sul	-	-	-	19.4 ± 9	15 ± 8.6	0.066
	7	Santos-Ponta da Praia	13 ± 4.3	11 ± 3.6	0.002	16.5 ± 7.6	13.6 ± 5.9	0.000
	15	São José do Rio Preto	8 ± 2.9	8 ± 3	0.958	14.7 ± 5.7	14.9 ± 5.7	0.805
NO _x	2	Guaratinguetá	6.1 ± 2	5.1 ± 1.6	0.004	9.9 ± 5.2	7.6 ± 4.5	0.000
		S.José Campos	11.9 ± 4	9.9 ± 3.7	0.001	23.1 ± 14.6	17.4 ± 12.3	0.000
		S.José Campos-Jd.Satelite	-	-	-	18 ± 15.1	11.6 ± 10.2	0.003
	5	Campinas-Taquaral	6.1 ± 2.8	4.6 ± 2.2	0.005	13.5 ± 9.2	8.7 ± 6.9	0.000
		Piracicaba	-	-	-	12.2 ± 4.5	10.3 ± 4.5	0.006
		Santa Gertrudes	21.2 ± 7.9	40.6 ± 14.6	0.000	36.5 ± 15.6	58.3 ± 17	0.000
	6	Cerqueira César	33.5 ± 15.2	20.8 ± 11.4	0.000	37.3 ± 21.9	23.5 ± 18	0.000
		Congonhas	56.1 ± 18.9	54.9 ± 18.2	0.908	72.8 ± 30.3	66 ± 47.4	0.001
		Guarulhos-Pimentas	17.1 ± 9.9	12 ± 4.6	0.000	22.6 ± 14.7	18.8 ± 14.6	0.001
		Ibirapuera	16.1 ± 10.5	12.7 ± 4.8	0.005	23.7 ± 16.7	18.6 ± 15.6	0.000
		Itaim Paulista	14.1 ± 9	10.7 ± 5.1	0.002	18.9 ± 13.6	17.1 ± 13.9	0.001
		Marg.Tietê-Pte Remédios	75.2 ± 36.9	53.6 ± 33.2	0.000	96.7 ± 63.9	73.7 ± 52.1	0.000
		Osasco	53.4 ± 21.3	41.7 ± 12.7	0.000	71.4 ± 31.5	51.4 ± 27.6	0.000
		Parque D.Pedro II	30.6 ± 17.5	19.1 ± 7.6	0.000	35 ± 22.8	28.9 ± 24.9	0.000
		Pinheiros	39.2 ± 24.5	23.9 ± 13.6	0.000	57.1 ± 41.8	29.3 ± 25.6	0.000
		S.Bernardo-Centro	-	-	-	22.1 ± 15.4	18 ± 13.3	0.003
		São Caetano do Sul	-	-	-	33.1 ± 24.1	24.6 ± 20.4	0.000
	7	Cubatão-Centro	30.7 ± 10	29.9 ± 9.3	0.451	47.4 ± 16.3	42.7 ± 15.6	0.008

	13	Cubatão-V.Parisi	73 ± 30.4	77.2 ± 24.7	0.924	103.4 ± 30.8	139.9 ± 38.4	0.000
		Bauru	6.6 ± 2.4	5.4 ± 2.2	0.007	13.8 ± 8.9	11.9 ± 7.6	0.030
		Jaú	7.1 ± 1.7	5.6 ± 1.5	0.000	9.8 ± 3.7	8.1 ± 4.2	0.000
	15	Catanduva	5.4 ± 1.9	5.1 ± 2	0.816	14.2 ± 7.3	11 ± 5.9	0.000
		São José do Rio Preto	9.1 ± 3.5	8.7 ± 3.4	0.603	22.8 ± 13.5	20.3 ± 12.8	0.259
	22	Marília	6 ± 1.7	4.5 ± 1.5	0.000	9.2 ± 3.6	6.1 ± 2.9	0.000
		Presidente Prudente	4.5 ± 2.1	4.9 ± 2.3	0.439	9.1 ± 4.9	6.7 ± 4	0.000
NO ₂	2	Guaratinguetá	9.5 ± 2.8	8.1 ± 2.2	0.005	13.3 ± 4.9	10.7 ± 5	0.000
		S.José Campos	16 ± 3.9	13.2 ± 3.7	0.000	21.3 ± 5.7	17.4 ± 7.4	0.000
		S.José Campos-Jd.Satelite	15.1 ± 5.3	12.2 ± 4.5	0.011	19.3 ± 9.1	12.7 ± 8.6	0.000
		Taubaté	13.5 ± 4.2	10.8 ± 3	0.000	18.1 ± 7.1	14.4 ± 7.8	0.000
	5	Campinas-Taquaral	10.3 ± 4.2	7.6 ± 3.5	0.001	18.3 ± 9	12.5 ± 8.6	0.000
		Piracicaba	10.2 ± 3.2	11.9 ± 3.8	0.097	15.4 ± 4.7	14.4 ± 5.1	0.201
		Santa Gertrudes	25.5 ± 8.9	26.4 ± 8.5	0.098	33.9 ± 8	34.5 ± 8.7	0.710
	6	Cerqueira César	35.5 ± 12.9	24 ± 10.6	0.000	37.2 ± 16.2	26.1 ± 15.6	0.000
		Congonhas	57 ± 15.2	50.7 ± 12.5	0.122	62.5 ± 15	51.6 ± 18	0.000
		Guarulhos-Pimentas	20.9 ± 8	15.9 ± 5.3	0.001	23.5 ± 8.7	20.6 ± 9.6	0.004
		Ibirapuera	23.4 ± 10.3	20.4 ± 6.7	0.015	29.3 ± 11.7	23.3 ± 11.4	0.000
		Itaim Paulista	19.5 ± 8.4	15 ± 5.8	0.000	22.4 ± 9.2	20.1 ± 10.8	0.000
		Marg.Tietê-Pte Remédios	56.6 ± 19	44 ± 16.6	0.000	61.8 ± 22.8	51.9 ± 22.7	0.001
		Osasco	41.1 ± 13.2	34.3 ± 9.8	0.002	47.3 ± 12.2	38.4 ± 12.6	0.000
		Parque D.Pedro II	35.8 ± 13.1	27 ± 8.4	0.001	37.7 ± 12.8	30.5 ± 14.8	0.000
		Pico do Jaraguá	10.2 ± 6.5	11.6 ± 4.7	0.699	13.8 ± 6	11.8 ± 5.2	0.000
		Pinheiros	36.9 ± 13.5	27 ± 9.1	0.000	44 ± 17.6	25 ± 10.9	0.000
		S.Bernardo-Centro	24.8 ± 11.4	18 ± 7.8	0.001	29.5 ± 14.8	24.2 ± 14.2	0.001
		São Caetano do Sul	31.3 ± 14.2	22.5 ± 8.2	0.009	33.8 ± 12.1	25.8 ± 13.2	0.000
	7	Cubatão-Centro	28.8 ± 6.3	26 ± 6	0.023	36.7 ± 9.6	34.5 ± 12	0.012
		Cubatão-V.Parisi	42.5 ± 15.1	41.7 ± 10.3	0.340	51.9 ± 10.5	59.8 ± 11.9	0.000
	10	Santos-Ponta da Praia	25.3 ± 7.5	23.1 ± 5.5	0.051	32.4 ± 11.5	33 ± 13.3	0.165
		Sorocaba	-	-	-	22.2 ± 10.1	18.7 ± 11.7	0.003
		Tatuí	7.7 ± 3.3	6.9 ± 4.3	0.246	11.2 ± 8.5	10.1 ± 8.2	0.043
	13	Araraquara	11.5 ± 3.3	10 ± 3.5	0.009	20.5 ± 9.7	19.7 ± 9.9	0.281
		Bauru	9.6 ± 3.4	7.5 ± 3.3	0.012	17.9 ± 8.7	16 ± 8.3	0.048
	15	Jaú	7.6 ± 2.3	8.2 ± 2.3	0.181	10.4 ± 4.1	12.3 ± 5.9	0.005
		Catanduva	8.5 ± 2.9	7.6 ± 3	0.456	18.8 ± 7.7	15.6 ± 6.5	0.000
		São José do Rio Preto	11.9 ± 4.2	11.8 ± 4.8	0.939	23.2 ± 8.6	21.8 ± 8.8	0.198
	21	Marília	9.4 ± 2.7	6.5 ± 2.3	0.000	14.5 ± 5.1	9.8 ± 4.5	0.000
	22	Presidente Prudente	7 ± 2.7	7.2 ± 3.3	0.781	12.6 ± 5.2	10.3 ± 5.2	0.004
NO	2	Guaratinguetá	1.3 ± 0.9	0.9 ± 0.6	0.020	3.5 ± 3.6	2.3 ± 2.5	0.000
		S.José Campos	4.2 ± 3	3.5 ± 2.7	0.088	14.5 ± 14.9	10.1 ± 11	0.002
		S.José Campos-Jd.Satelite	2.3 ± 1.7	2 ± 1.5	0.343	9.4 ± 13.6	6.1 ± 9	0.066
		Taubaté	1.1 ± 1	1.1 ± 0.6	0.434	2.5 ± 3.7	2.7 ± 3.6	0.430
	5	Piracicaba	1.6 ± 1	2.4 ± 1.7	0.131	4.9 ± 3	3.3 ± 2.6	0.000
		Santa Gertrudes	9.5 ± 4.9	32.6 ± 13.6	0.000	22.7 ± 15.1	49.1 ± 16.1	0.000
	6	Guarulhos-Pimentas	7.4 ± 7.6	4.3 ± 2.8	0.005	12.3 ± 13.5	9.6 ± 12.4	0.001
		Itaim Paulista	4.6 ± 6.2	3.3 ± 3.8	0.129	8.6 ± 11.9	7.9 ± 10.8	0.004
		Marg.Tietê-Pte Remédios	55.7 ± 35.2	37.5 ± 31.4	0.000	78.9 ± 65.5	56.9 ± 50.4	0.000
		Osasco	38.9 ± 19.5	28.9 ± 10.3	0.000	56.8 ± 32.7	38.2 ± 26.7	0.000
		Parque D.Pedro II	14.3 ± 14	5.8 ± 4.2	0.000	18.5 ± 21.3	15.6 ± 22	0.003
		Pinheiros	24 ± 22.9	11.7 ± 11.7	0.000	41.5 ± 41.5	19.7 ± 26	0.000
		S.Bernardo-Centro	-	-	-	7.9 ± 10.9	6.3 ± 8.2	0.171
		São Caetano do Sul	-	-	-	18.6 ± 23.6	13.5 ± 17.5	0.000
	7	Cubatão-V.Parisi	64.8 ± 30.5	70.9 ± 26.3	0.843	96.3 ± 32.4	138.8 ± 41.5	0.000
	10	Sorocaba	2.8 ± 2.7	3.5 ± 2.8	0.029	10.1 ± 9.8	7.3 ± 7.7	0.004
		Tatuí	1.5 ± 1	1.3 ± 0.8	0.112	2 ± 2	2.2 ± 3.5	0.033
	13	Jaú	3.7 ± 1.1	1.5 ± 0.7	0.000	5.3 ± 2.5	1.9 ± 1.6	0.000
	15	São José do Rio Preto	3.5 ± 1.9	3.1 ± 1.5	0.088	12.8 ± 12	10.8 ± 10.9	0.187
	21	Marília	1.1 ± 0.6	1.3 ± 0.7	0.626	1.8 ± 1.5	1.1 ± 0.8	0.000
	22	Presidente Prudente	0.9 ± 1	1.3 ± 1	0.002	2.9 ± 3	1.4 ± 1.8	0.000
SO ₂	2	S.José Campos	1.3 ± 0.9	1.9 ± 1.8	0.065	1.7 ± 1.1	2 ± 1.9	0.893
	5	Paulínia	5.2 ± 3	3.2 ± 2.3	0.001	-	-	-
	6	Cerqueira César	-	-	-	1.7 ± 1.1	1.6 ± 0.8	0.819
		Congonhas	1.6 ± 1.1	1.3 ± 0.6	0.268	1.9 ± 1.1	1.6 ± 0.8	0.057
		Guarulhos-Pimentas	2.5 ± 1	1.9 ± 1.3	0.000	2.7 ± 1.6	2.7 ± 2.2	0.483
		Interlagos	-	-	-	1.7 ± 0.9	1.3 ± 0.6	0.017
		Marg.Tietê-Pte Remédios	1.9 ± 0.8	2.1 ± 1.1	0.338	2.1 ± 1.1	2.2 ± 0.9	0.644
		Osasco	2 ± 0.6	1.6 ± 0.5	0.000	3.1 ± 1.1	2 ± 0.7	0.000
		S.André-Capuava	2.3 ± 2	2 ± 1.3	0.932	3.5 ± 2.6	2.7 ± 1.5	0.058
		São Caetano do Sul	-	-	-	1.9 ± 0.9	1.5 ± 0.6	0.069
	7	Cubatão-Centro	9.5 ± 4.7	5.7 ± 3.1	0.000	9.5 ± 4.6	10.8 ± 10.6	0.196
		Cubatão-V.Parisi	9.4 ± 9	8.9 ± 6.1	0.666	11.7 ± 8.3	11.7 ± 9.6	0.664
		Santos-Ponta da Praia	13.9 ± 8.5	2.7 ± 1.7	0.000	12.6 ± 6.3	3.6 ± 1.7	0.000
CO	2	S.José Campos-Jd.Satelite	0.1 ± 0.1	0.2 ± 0	0.001	0.3 ± 0.1	0.4 ± 0.2	0.000
	5	Campinas-Centro	0.6 ± 0.1	0.6 ± 0.1	0.000	0.7 ± 0.1	0.5 ± 0.1	0.000
	6	Carapicuíba	0.5 ± 0.1	0.4 ± 0.1	0.000	0.6 ± 0.2	0.4 ± 0.2	0.000
		Cerqueira César	0.5 ± 0.1	0.4 ± 0.1	0.002	0.5 ± 0.2	0.4 ± 0.2	0.000
		Congonhas	0.7 ± 0.2	0.6 ± 0.1	0.012	0.8 ± 0.3	0.7 ± 0.4	0.000
		Grajaú-Parelheiros	0.5 ± 0.2	0.4 ± 0.2	0.000	0.6 ± 0.3	0.3 ± 0.2	0.000
		Guarulhos-Pimentas	0.5 ± 0.1	0.4 ± 0.1	0.000	0.6 ± 0.2	0.5 ± 0.2	0.000
		Ibirapuera	0.3 ± 0.1	0.3 ± 0.1	0.008	0.4 ± 0.2	0.3 ± 0.2	0.001
		Marg.Tietê-Pte Remédios	0.7 ± 0.2	0.6 ± 0.2	0.000	0.8 ± 0.4	0.6 ± 0.3	0.000
		Mooca	0.5 ± 0.1	0.4 ± 0.1	0.000	0.7 ± 0.4	0.4 ± 0.2	0.000
		Osasco	0.8 ± 0.2	0.6 ± 0.1	0.000	0.9 ± 0.3	0.6 ± 0.3	0.000
		Parque D.Pedro II	0.3 ± 0.1	0.2 ± 0.1	0.000	0.4 ± 0.2	0.3 ± 0.3	0.000
		Pinheiros	0.6 ± 0.2	0.4 ± 0.2	0.001	0.7 ± 0.4	0.4 ± 0.3	0.000

O ₃		S.Bernardo-Centro	0.5 ± 0.1	0.4 ± 0.1	0.001	0.5 ± 0.2	0.4 ± 0.3	0.000
	2	Guaratinguetá	39.9 ± 13.6	38.1 ± 8.5	0.580	36 ± 7.7	34.4 ± 8	0.134
		Jacareí	36.9 ± 15.1	28.1 ± 7.8	0.000	27.7 ± 9.4	13.8 ± 6	0.000
		S.José Campos	39.3 ± 13.6	34.3 ± 8.1	0.027	33.5 ± 7.5	32.9 ± 8.2	0.402
		S.José Campos-Jd.Satelite	36.3 ± 14.4	39.1 ± 10.5	0.511	33.7 ± 10.9	37.1 ± 10.7	0.012
		Taubaté	45.7 ± 17.1	38.3 ± 10.7	0.002	36.5 ± 10.1	35 ± 9.1	0.131
	5	Campinas-Taquaral	52.7 ± 20.8	44.2 ± 13.4	0.065	41.2 ± 13.4	42.5 ± 13.6	0.382
		Campinas-V.União	48 ± 14.4	45.6 ± 10.6	0.850	40.2 ± 10.6	34.3 ± 11	0.000
		Jundiaí	44.7 ± 18.2	44.3 ± 13.3	0.851	32.2 ± 11.2	38 ± 13.1	0.000
		Paulínia	42.4 ± 23.1	36.2 ± 14	0.289	30.8 ± 14.5	33.7 ± 14.6	0.117
		Piracicaba	48.5 ± 17.8	38 ± 12.4	0.273	36 ± 9.9	33.8 ± 10.6	0.056
	6	Capão Redondo	39.2 ± 16.3	33.5 ± 8.4	0.021	30.5 ± 10.5	34.9 ± 8.9	0.001
		Carapicuíba	38 ± 17.9	35.8 ± 12.9	0.596	29.8 ± 11.7	33.6 ± 11.2	0.005
		Diadema	42.8 ± 19.3	34.9 ± 13	0.009	30.5 ± 12	34.7 ± 10.4	0.000
		Guarulhos-Paço Municipal	36.5 ± 15	36.4 ± 13.3	0.576	28.9 ± 9.9	37.1 ± 10.8	0.000
		Guarulhos-Pimentas	43.3 ± 13.5	40.4 ± 9.6	0.251	34.8 ± 8.7	40.9 ± 9.6	0.000
		Ibirapuera	46.1 ± 19.6	44.1 ± 17	0.462	35.8 ± 13.1	47.4 ± 13.4	0.000
		Interlagos	-	-	-	34.8 ± 11.4	39.5 ± 9.6	0.004
		Itaim Paulista	43.9 ± 14.2	41 ± 12.6	0.308	37.4 ± 9.7	39 ± 10.1	0.133
		Parque D.Pedro II	39.1 ± 17.4	35 ± 12.7	0.145	30.2 ± 11.8	35.8 ± 11.6	0.000
		Pinheiros	31.3 ± 14.5	27.1 ± 12.3	0.072	24.3 ± 11.2	28.9 ± 10.6	0.000
		S.André-Capuava	45.1 ± 18.6	35.4 ± 11.5	0.004	31.6 ± 10.6	38.7 ± 9.7	0.000
		S.Bernardo-Centro	-	-	-	39.1 ± 12.5	40.9 ± 10.3	0.152
		Santana	-	-	-	31.3 ± 11.6	40.4 ± 11.9	0.000
		São Caetano do Sul	-	-	-	32.2 ± 12.3	37.5 ± 11	0.001
	7	Santos-Ponta da Praia	28.3 ± 12.1	25.8 ± 8.2	0.287	19.8 ± 6.1	25 ± 7.9	0.000
	10	Sorocaba	-	-	-	31.2 ± 9.9	36.7 ± 11.7	0.000
		Tatuí	45 ± 16	35 ± 11.3	0.000	38.6 ± 8.5	40.6 ± 9.3	0.125
	13	Bauru	42.1 ± 14.5	35.8 ± 11.5	0.216	33.7 ± 8.9	37.3 ± 10.2	0.001
		Jaú	42.5 ± 14	37.8 ± 11	0.176	35.2 ± 8.9	40.6 ± 9.3	0.000
	15	Catanduva	46.1 ± 12.3	40.4 ± 10.3	0.006	36.2 ± 9	38.6 ± 8.7	0.023
	19	Araçatuba	44.3 ± 12.3	37.8 ± 10.7	0.018	37.6 ± 9.6	40.6 ± 9.6	0.008
	21	Marília	48.6 ± 16.9	48.3 ± 15.4	0.550	47.5 ± 11.3	58.2 ± 14	0.000
	22	Presidente Prudente	46.5 ± 14.7	39.2 ± 10.9	0.009	43.2 ± 11.4	43.9 ± 11.6	0.570

Table B.3 – Mann-Whitney U test results for comparison average daily concentrations considering days without rainfall events for the period before (January 1 to March 15) and during (March 16 to June 30) COVID-19 outbreak.

HUWRM	Station name	Meteorological Station	Distance (km)	PM ₁₀	PM _{2.5}	NO _x	NO ₂	NO	SO ₂	CO	O ₃
2	Guaratinguetá	Cachoeira Paulista	22.8	↑	NA	↑	↑	↑	NA	NA	↓
	Jacareí	Taubaté	53.7	=	NA	NA	NA	NA	NA	NA	↓
	S.José Campos	Taubaté	39.4	↑	NA	↑	↑	↑	=	NA	↓
	S.José Campos-Jd.Satelite	Taubaté	42.9	↑	↑	=	=	=	NA	↑	↓
	Taubaté	Taubaté	5.7	↑	↑	NA	↑	=	NA	NA	↓
5	Campinas-Centro	Braganca Paulista	54.3	↑	NA	NA	NA	NA	NA	↓	NA
	Campinas-Taquaral	Braganca Paulista	54.9	NA	NA	↑	↑	NA	NA	NA	↓
	Campinas-V.União	Piracicaba	58.4	NA	↑	NA	NA	NA	NA	NA	↓
	Jundiaí	Barueri	36.9	↑	NA	NA	NA	NA	NA	NA	↓
	Limeira	Piracicaba	26.5	↑	NA	NA	NA	NA	NA	NA	NA
	Paulínia	Piracicaba	48.7	↑	NA	NA	NA	NA	=	NA	↓
	Piracicaba	Piracicaba	2.7	↑	↑	↑	↑	↑	NA	NA	↓
	Santa Gertrudes	Piracicaba	28.4	↑	=	↑	↑	↑	NA	NA	NA
6	Capão Redondo	São Paulo - Interlagos	12.2	↑	NA	NA	NA	NA	NA	NA	↓
	Carapicuíba	Barueri	3.5	↑	NA	NA	NA	NA	NA	=	↓
	Cerqueira César	São Paulo - Mirante	8.3	=	NA	=	=	NA	↑	=	NA
	Congonhas	São Paulo - Interlagos	12.1	↑	=	=	=	NA	=	=	NA
	Diadema	São Paulo - Interlagos	8	↑	NA	NA	NA	NA	NA	NA	↓
	Grajaú-Parelheiros	São Paulo - Interlagos	6.1	↑	=	NA	NA	NA	NA	=	NA
	Guarulhos-Paço Municipal	São Paulo - Mirante	11.3	↑	↑	NA	NA	NA	NA	NA	↓
	Guarulhos-Pimentas	São Paulo - Mirante	22.4	NA	NA	↑	↑	↑	=	↑	↓
	Ibirapuera	São Paulo - Mirante	11.4	NA	=	=	=	NA	NA	↑	=
	Interlagos	São Paulo - Interlagos	4.9	↑	NA	NA	NA	NA	=	NA	↓
	Itaim Paulista	São Paulo - Mirante	20.4	NA	↑	↑	↑	=	NA	NA	↓
	Marg.Tietê-Pte Remédios	São Paulo - Mirante	12.8	↑	↑	↑	=	↑	↑	=	NA
	Mooca	São Paulo - Mirante	6.3	NA	=	NA	NA	NA	NA	=	NA
	N.Senhora do Ó	São Paulo - Mirante	7.6	↑	NA	NA	NA	NA	NA	NA	NA
	Osasco	Barueri	7.9	↑	↑	↑	=	↑	↑	↓	NA
	Parque D.Pedro II	São Paulo - Mirante	5.4	NA	↑	↑	=	↑	NA	=	↓
	Pico do Jaraguá	Barueri	12.9	NA	↓	NA	=	NA	NA	NA	NA
	Pinheiros	São Paulo - Mirante	11	NA	NA	=	↓	=	NA	=	=
	S.André-Capuava	São Paulo - Mirante	20.6	=	NA	NA	NA	NA	=	NA	=
	S.Bernardo-Centro	São Paulo - Interlagos	13.7	NA	=	↑	↑	↑	NA	↑	↓
	S.Bernardo-Paulicéia	São Paulo - Interlagos	11.1	=	NA	NA	NA	NA	NA	NA	NA
	Santana	São Paulo - Mirante	1.4	NA	↑	NA	NA	NA	NA	NA	↓
	São Caetano do Sul	São Paulo - Mirante	15	=	=	=	=	↑	↑	NA	↓
	Taboão da Serra	Barueri	14.8	=	NA	NA	NA	NA	NA	NA	NA
7	Cubatão-Centro	Bertioga	28.3	NA	NA	NA	NA	NA	NA	NA	NA
	Cubatão-V.Parisi	Bertioga	25	NA	NA	NA	NA	NA	NA	NA	NA
	Santos-Ponta da Praia	Bertioga	22	NA	NA	NA	NA	NA	NA	NA	NA
10	Sorocaba	Sorocaba	13.8	↑	NA	NA	↑	↑	NA	NA	↓
	Tatuí	Sorocaba	30	↑	NA	NA	↑	↑	NA	NA	↑
13	Araraquara	São Carlos	38.1	↑	NA	NA	↑	NA	NA	NA	NA
	Bauru	Bauru	7.4	↑	NA	↑	↑	NA	NA	NA	=
	Jaú	Bauru	48	↑	NA	↑	↑	=	NA	NA	=
15	Catanduva	Ariranha	14.8	=	NA	↓	↓	NA	NA	NA	=
	São José do Rio Preto	José Bonifácio	63.7	↑	↑	↓	↓	=	NA	NA	NA
19	Araçatuba	Valparaíso	53	↑	NA	NA	NA	NA	NA	NA	=
21	Marília	Marília	4	↑	NA	↑	↑	=	NA	NA	↑
22	Presidente Prudente	Presidente Prudente	0	↑	NA	↑	↑	=	NA	NA	↑

Legend: ↓ Statistically different and lower concentration at a monitoring station during mobility restriction in relation to the period before the mobility restrictions; ↑ Statistically different and higher concentration at a monitoring station during mobility restriction in relation to the period before the mobility restrictions; = Monitoring station without statistically significant differences in the concentration for 2019 and 2020; NA, not available data. Distance indicates the distance between the air quality station and the closest meteorological station.

Figure B.1 - PM₁₀ concentration for Hydrographic Units for Water Resources Management (HUWRMs) in the State of São Paulo. The light gray area represents the interval between the minimum and maximum. The dark gray line represents the average for the historical series; blue and red lines represent the average for the years 2019 and 2020, respectively. The vertical dashed black line indicates the 76th day of the year 2020 when the first restrictions were started.

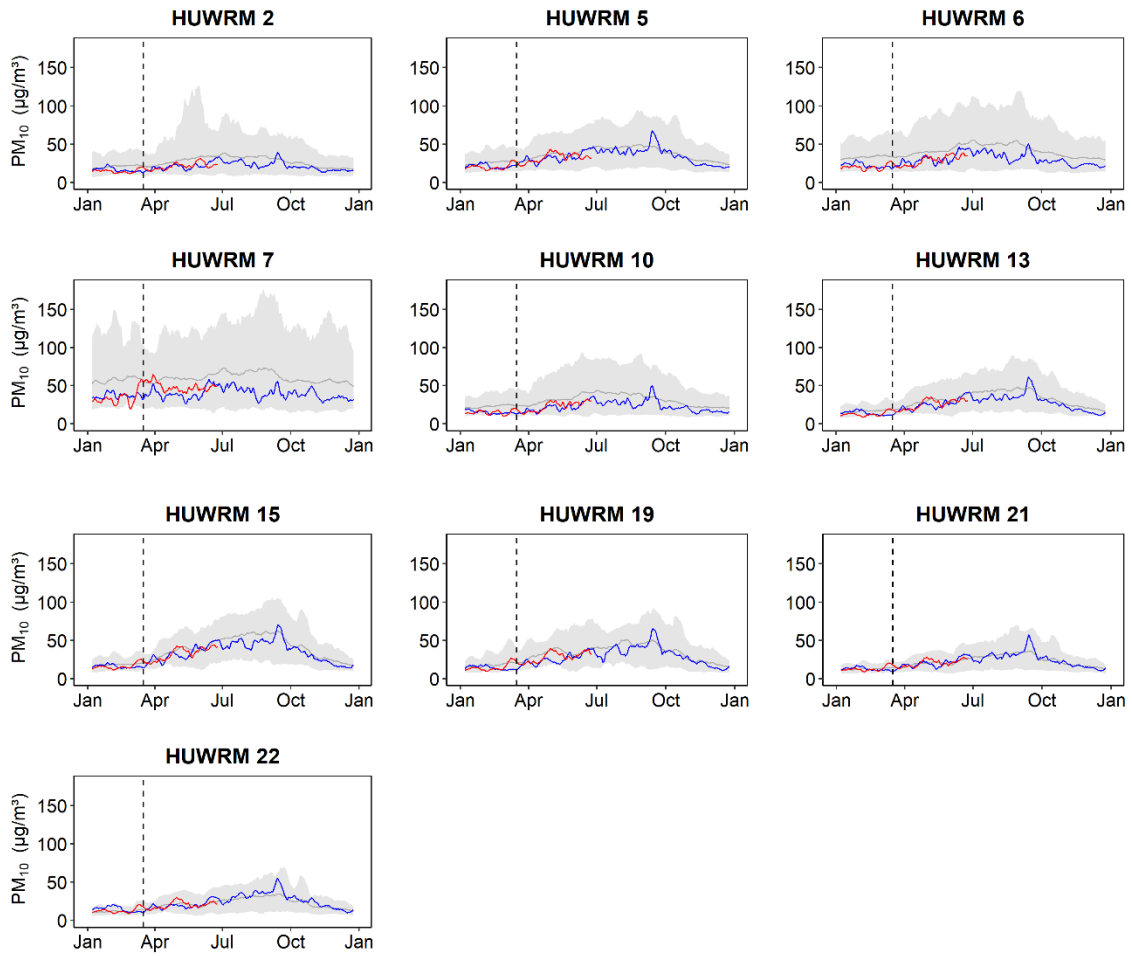


Figure B.2 - Same as in Figure B.1, but for $\text{PM}_{2.5}$ ($\mu\text{g}\cdot\text{m}^{-3}$).

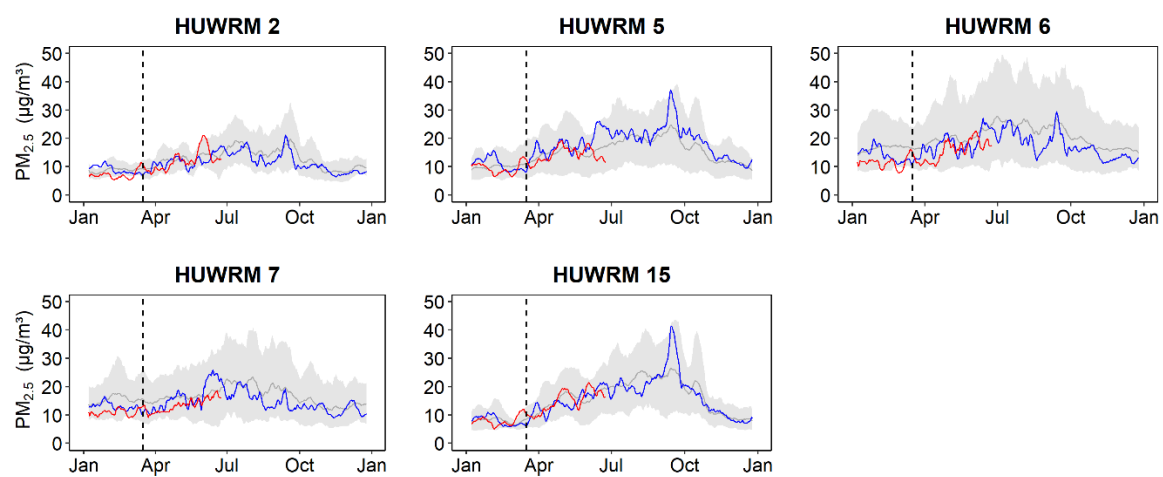


Figure B.3 - Same as in Figure B.1, but for NO_x (ppb).

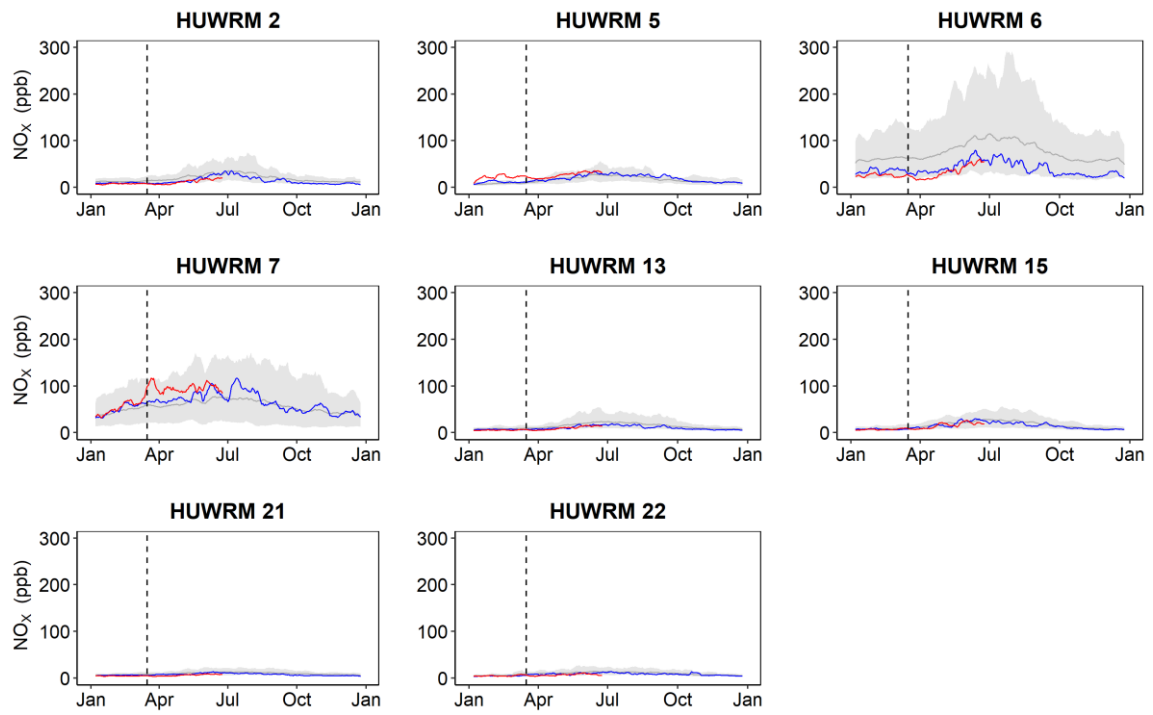


Figure B.4 - Same as in Figure B.1, but for NO₂ (µg·m⁻³).

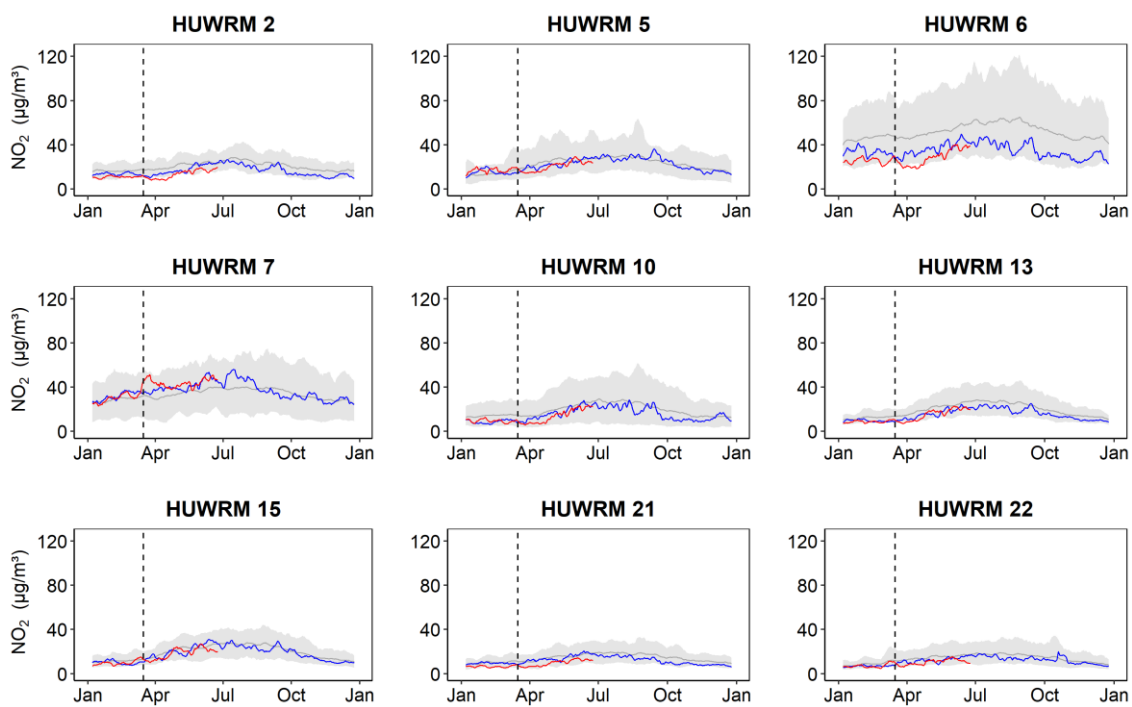


Figure B.5 - Same as in Figure B.1, but for NO ($\mu\text{g}\cdot\text{m}^{-3}$).

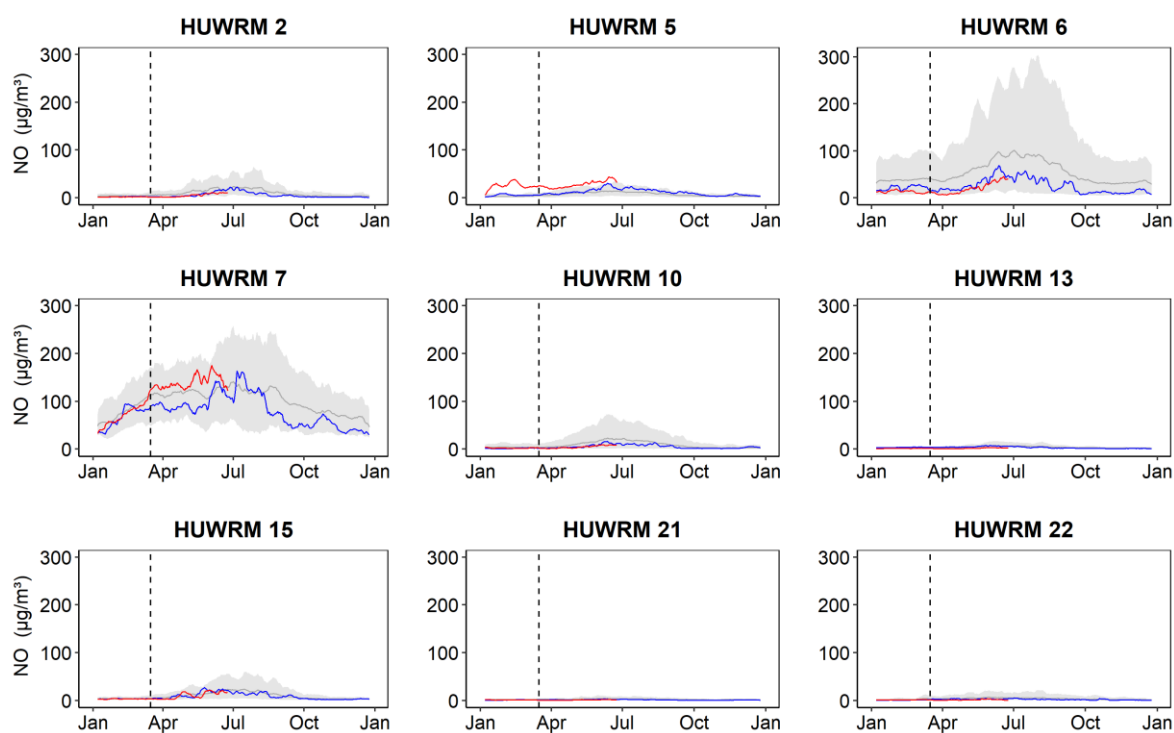


Figure B.6 - Same as in Figure B.1, but for O_3 ($\mu\text{g}\cdot\text{m}^{-3}$).

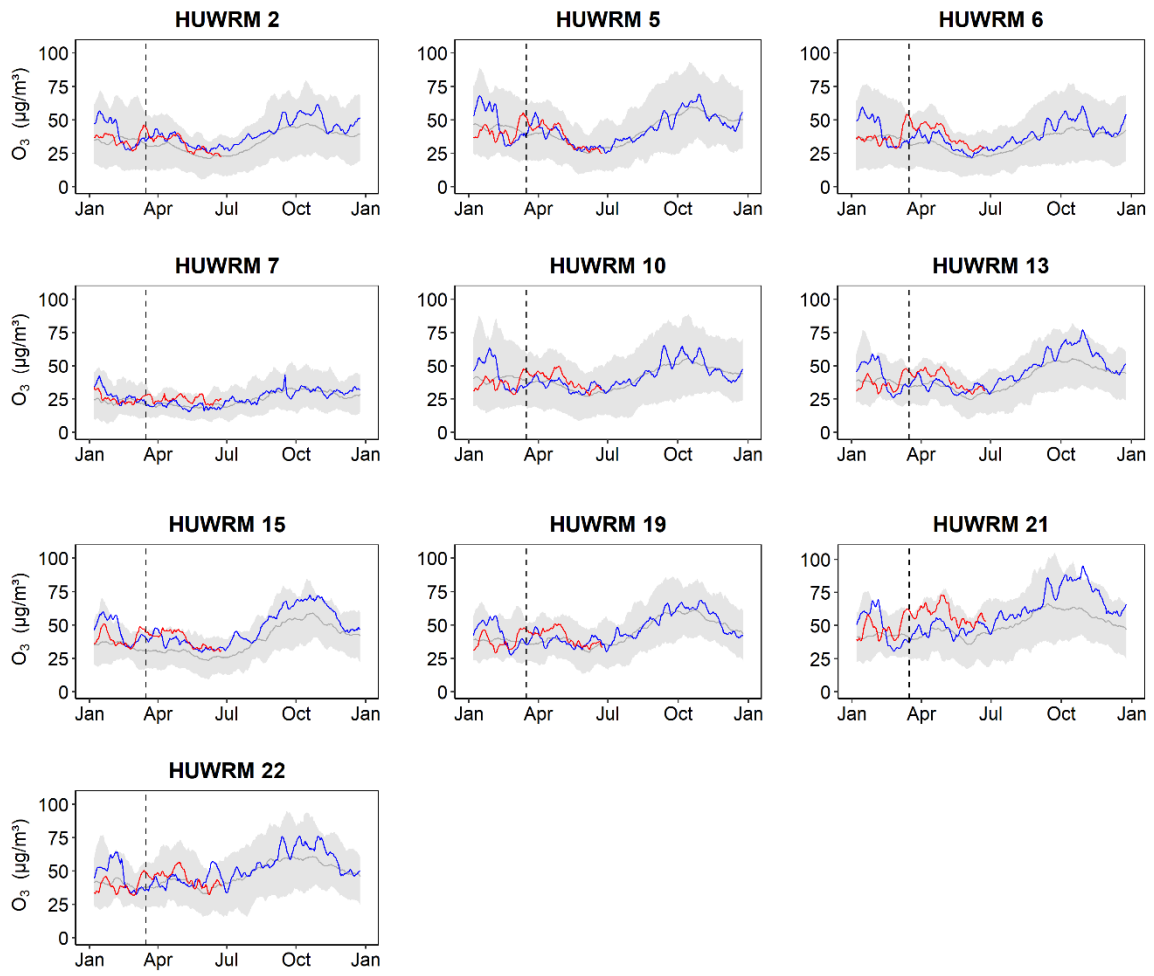


Figure B.7 - Monthly Rainfall (mm) for 2019 (blue bars), 2020 (red bars), and historical average (2003-2018) (gray bars) for the State of São Paulo.

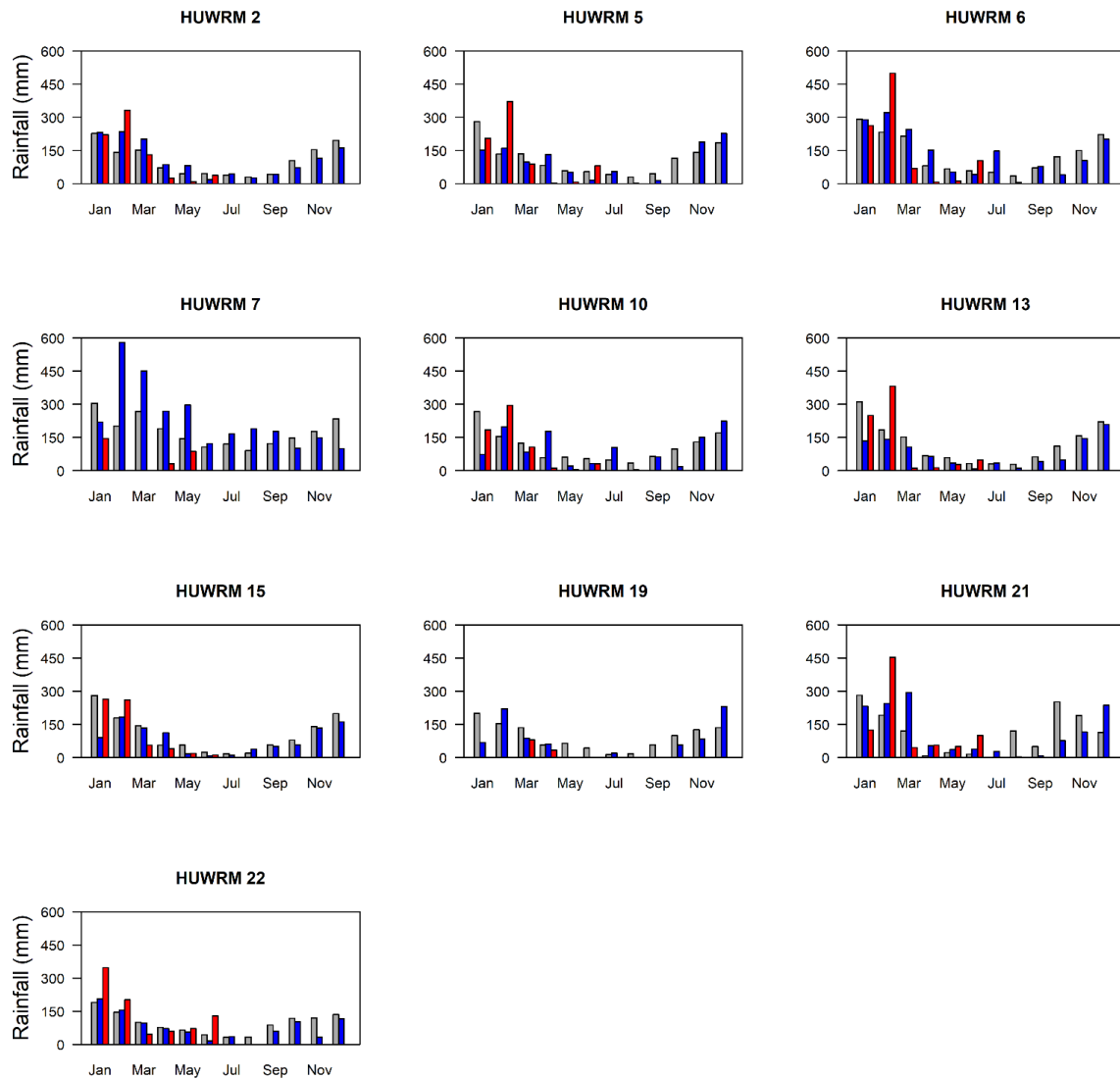


Figure B.8 - Daily variation of precipitation (blue line) and air pollutants concentrations (red line) for HUWRM 2 and HUWRM 6 in the State of São Paulo. Panel (a) shows the concentration of PM_{10} , $PM_{2.5}$, NO_2 , and NO for the air quality stations Marg.Tietê-Pte Remédios and Taubaté. Panel (b) shows the concentration of NO_x and SO_2 for the air quality stations Guarulhos-Pimentas and S.José Campos. Panel (c) shows the concentration of CO and O_3 for the air quality stations Guarulhos-Pimentas and S.José Campos-Jd.Satelite. The vertical dashed black line indicates the 76th day of the year 2020 when the first restrictions were started.

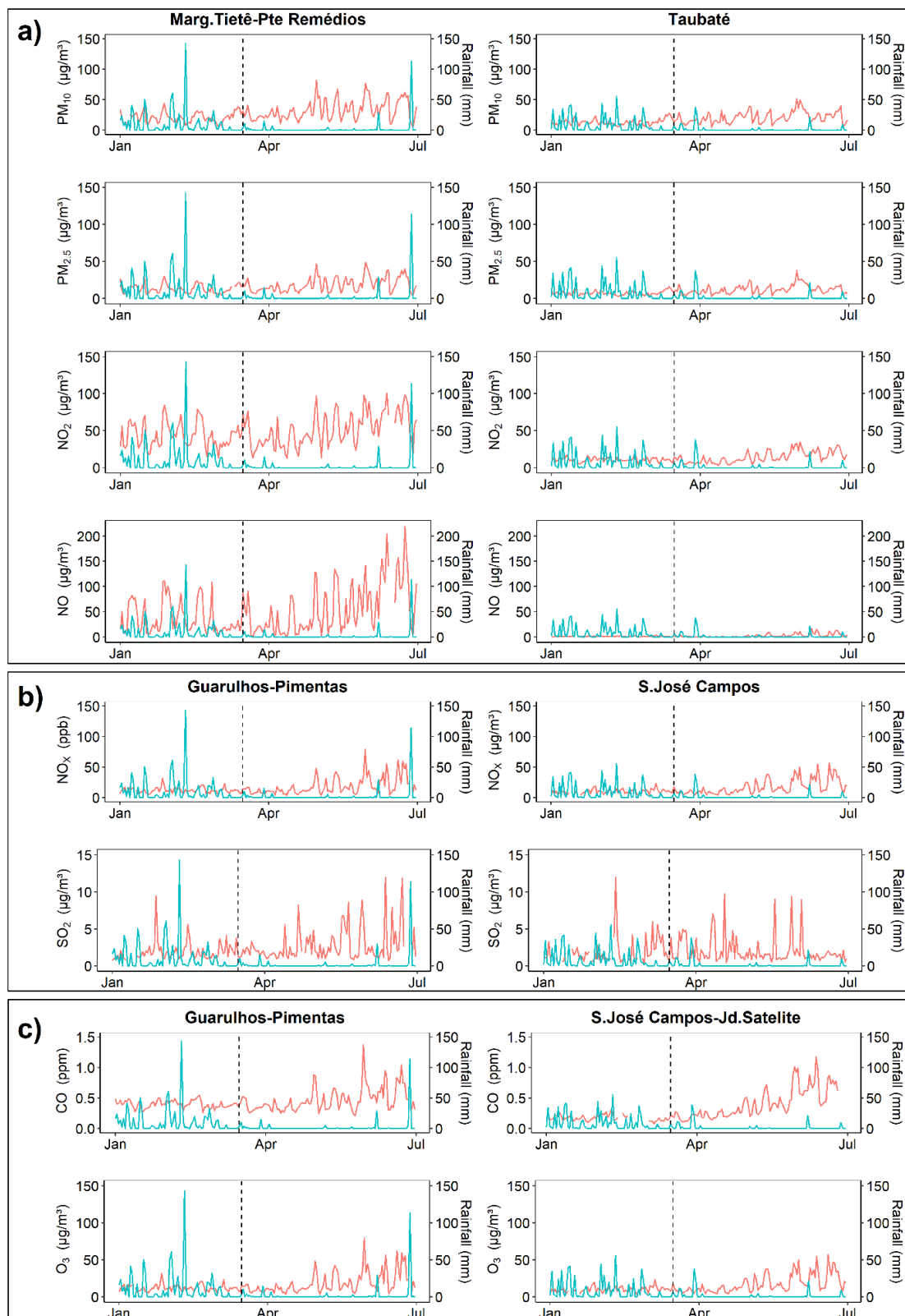
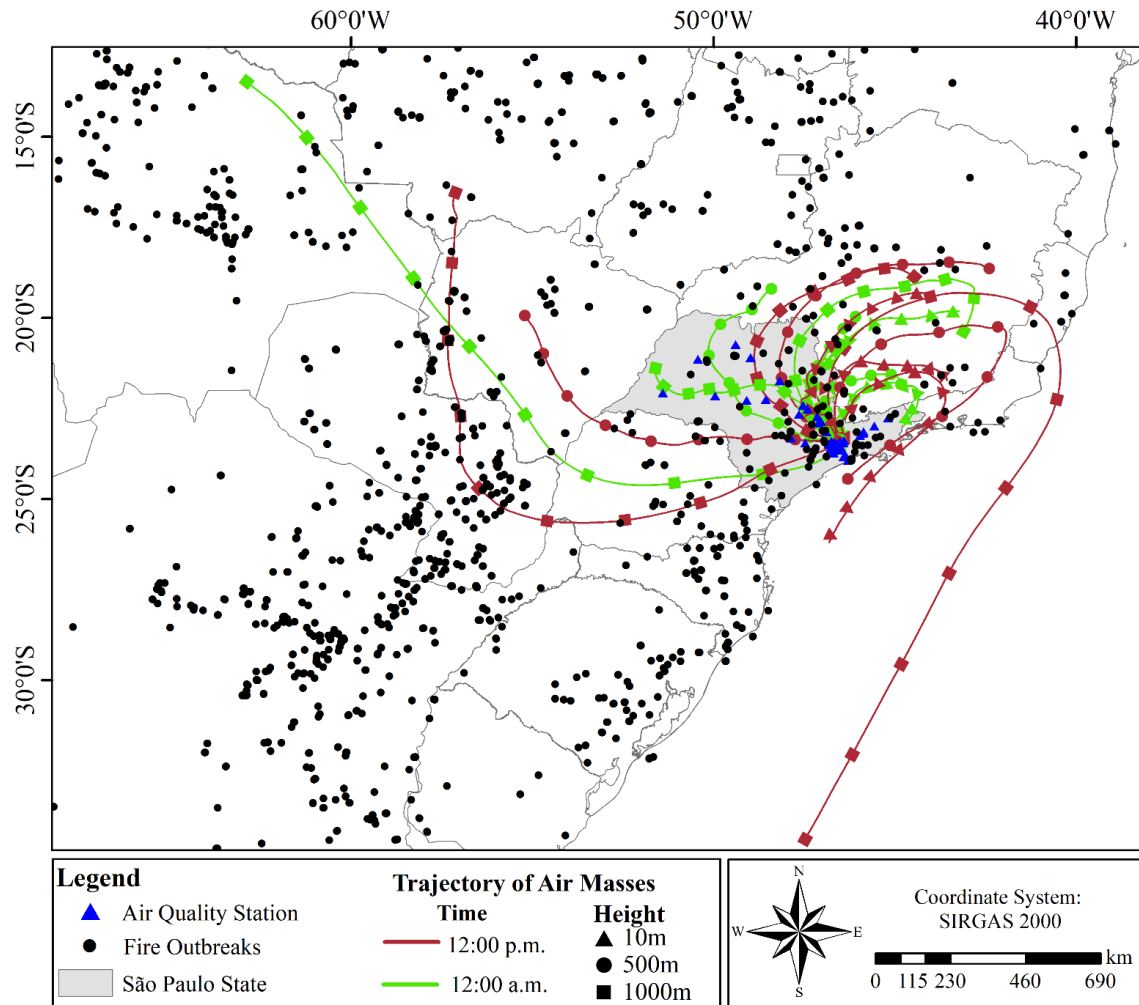


Figure B.9 - Trajectory of air masses arriving in the Itaim Paulista station, calculated by HYSPLIT. The trajectory of air masses and the fire outbreaks were acquired from May 30 to June 2.



**APPENDIX C – EVALUATING TROPOMI AND MODIS PERFORMANCE TO
CAPTURE THE DYNAMIC OF AIR POLLUTION IN SÃO PAULO STATE: A CASE
STUDY DURING THE COVID-19 OUTBREAK**

Data Summary

Table C.1 – CETESB air quality monitoring station for São Paulo state used in this research.

HUWRM	Station name	Lon	Lat	Station id	Classification	*Spatial representativity	PM ₁₀	PM _{2.5}	NO ₂
2	Guaratinguetá	-45.191	-22.802	289	Sub-urban traffic	Neighborhood	x		x
	Jacareí	-45.968	-23.294	259	Peri-urban	Neighborhood	x		
	S.José Campos	-45.871	-23.188	88	Sub-urban traffic	Neighborhood	x		x
	S.José Campos-Jd.Satelite	-45.891	-23.224	277	Sub-urban traffic	Neighborhood	x	x	x
	Taubaté	-45.576	-23.032	280	Sub-urban traffic	Neighborhood	x	x	x
	Total						5	2	4
5	Campinas-Centro	-47.057	-22.903	89	Urban traffic	Micro-scale	x		
	Campinas-Taquaral	-47.059	-22.875	276	Peri-urban	Urban			x
	Campinas-V.União	-47.119	-22.947	275	Sub-urban traffic	Urban		x	
	Jundiaí	-46.897	-23.192	109	Sub-urban traffic	Urban	x		
	Limeira	-47.414	-22.564	281	Sub-urban traffic	Neighborhood	x		
5	Paulínia	-47.155	-22.772	117	Sub-urban traffic	Neighborhood	x		
					Urban for NO ₂ and Neighborhood for others				
	Piracicaba	-47.65	-22.701	113	Peri-urban	Neighborhood	x	x	x
	Santa Gertrudes	-47.536	-22.46	273	Sub-urban traffic	Neighborhood	x	x	x
	Total						6	3	3
6	Capão Redondo	-46.78	-23.668	269	Sub-urban traffic	Neighborhood	x		
	Carapicuíba	-46.836	-23.531	263	Sub-urban traffic	Neighborhood	x		
	Cerqueira César	-46.673	-23.554	91	Urban traffic	Micro-scale	x		x
	Congonhas	-46.663	-23.616	73	Urban traffic	Micro-scale	x	x	x
	Diadema	-46.612	-23.686	92	Sub-urban traffic	Neighborhood	x		
	Grajaú-Parelheiros	-46.697	-23.776	98	Sub-urban traffic	Micro-scale	x	x	
	Guarulhos-Paço Municipal	-46.519	-23.456	264	Urban traffic	Urban	x	x	
	Guarulhos-Pimentas	-46.41	-23.44	279	Sub-urban traffic	Neighborhood			x
	Ibirapuera	-46.661	-23.592	83	Urban Park	Urban		x	x
	Interlagos	-46.675	-23.681	262	Sub-urban traffic	Neighborhood	x		
	Itaim Paulista	-46.421	-23.502	266	Urban traffic	Urban		x	x
	Marg.Tietê-Pte Remédios	-46.743	-23.519	270	Urban traffic	Micro-scale	x	x	x
	Mooca	-46.6	-23.55	85	Urban traffic	Medium scale		x	
	N.Senhora do Ó	-46.692	-23.48	96	Urban traffic	Medium scale	x		
	Osasco	-46.792	-23.527	120	Urban traffic	Micro-scale	x	x	x
	Parque D.Pedro II	-46.628	-23.545	72	Urban traffic	Neighborhood		x	x
	Pico do Jaraguá	-46.766	-23.456	284	Regional background	Urban		x	x
	Pinheiros	-46.702	-23.561	99	Urban traffic	Micro-scale			x
	S.André-Capuava	-46.492	-23.64	100	Urban traffic	Neighborhood	x		x
	S.Bernardo-Centro	-46.546	-23.699	272	Sub-urban traffic	Neighborhood		x	x
	S.Bernardo-Paulicéia	-46.585	-23.671	102	Sub-urban traffic	Neighborhood	x		
	Santana	-46.629	-23.506	63	Urban traffic	Medium scale		x	
	São Caetano do Sul	-46.556	-23.618	86	Urban traffic	Medium scale	x	x	x
	Taboão da Serra	-46.758	-23.609	103	Urban traffic	Micro-scale	x		
	Total						15	13	13
7	Cubatão-Centro	-46.418	-23.879	87	Industrial	Neighborhood	x		x
	Cubatão-V.Parisi	-46.389	-23.849	66	Industrial	Neighborhood	x		x
	Santos-Ponta da Praia	-46.301	-23.981	260	Port handling	Neighborhood	x	x	x
	Total						3	1	3
10	Sorocaba	-47.479	-23.502	67	Sub-urban traffic	Urban for NO ₂ and Neighborhood for PM ₁₀	x		x
	Tatuí	-47.871	-23.361	256	Sub-urban traffic	Urban for NO ₂ and Neighborhood for PM ₁₀	x		x
	Total						2	0	2
13	Araraquara	-48.186	-21.783	106	Sub-urban traffic	Neighborhood	x		x
	Bauru	-49.093	-22.327	108	Sub-urban traffic	Neighborhood	x		x
	Jaú	-48.567	-22.299	110	Sub-urban traffic	Urban for NO ₂ and Neighborhood for PM ₁₀	x		x
	Total						3	0	3
15	Catanduva	-48.983	-21.142	248	Sub-urban traffic	Urban	x		x
	São José do Rio Preto	-49.398	-20.785	116	Sub-urban traffic	Urban	x	x	x
	Total						2	1	2
19	Araçatuba	-50.439	-21.187	107	Peri-urban	Urban	x		
	Total						1	0	0
21	Marília	-49.96	-22.2	111	Sub-urban traffic	Urban for NO ₂ and Neighborhood for PM ₁₀	x		x
	Total						1	0	1
22	Presidente Prudente	-51.409	-22.12	114	Sub-urban traffic	Urban	x		x
	Total						1	0	1
TOTAL							39	20	32

Note: *Classification provided by (CETESB (COMPANHIA AMBIENTAL DO ESTADO DE SÃO PAULO), 2021), where: micro-scale is relative to the spatial representativeness of areas from a few meters to 100 meters; medium scale represents blocks of urban areas (few blocks with similar characteristics) with dimensions between 101 and 500 meters; neighborhood scale indicates urban neighborhoods with uniform activity and dimensions between 501 and 4,000 meters; and urban scale is relative to the spatial representation of cities or metropolitan regions, on the order of 4 to 50 km.

Table C.2 – Correlation between PM₁₀ measured at ground level by CETESB stations and AOD data measured by the MODIS sensor coupled to the Aqua satellite.

Name	AQUA Satellite - 2019 (QA - 0000)									AQUA Satellite - 2020 (QA - 0000)								
	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km
Araçatuba (UR)	-0.14	-0.12	0.07	-0.06	-0.06	0.05	81	57	42	-0.5***	-0.45***	-0.29*	-0.5***	-0.44***	-0.32*	81	64	57
Araraquara (NE)	-0.28*	-0.04	0.13	-0.21	-0.22	-0.03	53	26	21	-0.17	0	0.09	-0.27*	-0.1	-0.03	60	44	39
Bauru (NE)	0.07	0.21	0	0.13	0.12	-0.12	64	39	30	-0.21	-0.1	-0.03	-0.24	-0.12	-0.06	67	55	52
Campinas-Centro (MI)	0.08	0.3	0.3	0.1	0.21	0.24	56	38	30	-0.04	0.03	0.13	0	0.1	0.16	54	47	40
Capão Redondo (NE)	0.15	0.07	0.07	0.18	0.07	0.05	44	31	30	0.07	0.2	0.32	0.1	0.26	0.41*	46	36	32
Carapicuíba (NE)	0.12	-0.14	-0.23	0.16	-0.1	-0.18	42	25	23	0.16	0.17	0.21	0.2	0.19	0.23	51	42	36
Catanduva (UR)	-0.37***	-0.19	0.13	-0.34**	-0.15	-0.01	63	42	31	-0.27*	-0.22	-0.08	-0.27*	-0.2	-0.07	67	55	42
Cerqueira César (MI)	0.12	-0.04	0.24	-0.04	-0.21	-0.04	39	27	24	0.17	0.15	0.17	0.21	0.22	0.22	51	37	35
Congonhas (MI)	0.17	0.01	-0.05	0.09	-0.07	-0.14	37	24	22	0.3*	0.35*	0.21	0.23	0.3	0.16	56	39	34
Cubatão-Centro (NE)	-0.08	0.06	0.1	-0.07	-0.02	0.04	45	31	28	0.15	0.37*	0.31	0.17	0.28	0.24	44	28	27
Cubatão-V.Parisi (NE)	-0.07	-0.22	-0.11	-0.21	-0.29	-0.2	46	30	24	0.17	0.19	0.21	0.14	0.22	0.19	40	31	25
Diadema (NE)	-0.01	0.13	0.08	0.05	0.11	0.1	44	35	28	0.25	0.26	0.12	0.28*	0.3*	0.18	54	48	40
Grajaú-Paraisópolis (MI)	-0.01	0.05	-0.07	-0.05	0.01	-0.12	51	37	30	0.1	0.12	0.16	-0.01	-0.02	0.06	58	46	42
Guaratinguetá (NE)	0.08	0.11	0.15	0.21*	0.18	0.17	95	77	60	0.06	0	0.14	0.14	0.11	0.22	76	62	50
Guarulhos-Paço Municipal (UR)	0.24	0.18	-0.06	0.2	0.2	0.01	42	27	23	0.02	-0.04	0.03	0.13	0.06	0.11	48	41	35
Interlagos (NE)	-0.03	-0.07	-0.16	0.2	0.2	0.01	43	29	27	0.22	0.14	0.13	0.22	0.19	0.18	58	45	42
Jacaré (NE)	0.06	0.13	-0.05	0.04	0.08	-0.07	71	52	44	-0.08	-0.03	-0.03	-0.11	-0.07	-0.07	61	48	43
Jau (NE)	-0.18	0.02	0.03	-0.07	0.05	0	64	36	30	-0.09	0.06	0.1	-0.08	0.1	0.09	73	51	39
Jundiaí (UR)	0.02	0.08	-0.01	0	0.06	0.02	53	38	30	0.15	0.17	0.39**	0.15	0.12	0.34*	63	50	42
Limeira (NE)	-0.31*	-0.13	-0.17	-0.15	-0.01	-0.09	54	34	27	0.21	0.39**	0.37*	0.32*	0.44**	0.46**	51	39	35
Marg.Tietê-Pte Remédios (MI)	-0.16	0	-0.05	-0.21	-0.11	-0.2	44	29	28	0.36**	0.28	0.23	0.36**	0.32*	0.25	56	42	37
Marília (NE)	-0.19	-0.29	-0.1	-0.2	-0.38*	-0.23	59	40	29	-0.02	0.07	0.09	-0.09	-0.05	-0.04	72	61	57
N.Senhora do Ó (ME)	0.29	0.16	0.13	0.21	-0.02	-0.12	41	30	21	-0.02	-0.05	-0.2	-0.02	-0.01	-0.13	45	34	31
Osasco (MI)	0.04	-0.14	-0.32	-0.02	-0.21	-0.39	41	27	24	0.01	0.12	0.11	0.06	0.25	0.25	56	40	35
Paulínia (NE)	0.05	0.1	0.02	0.1	0.15	0.01	65	47	38	-0.16	-0.07	0.06	-0.04	0.09	0.23	69	58	51
Piracicaba (NE)	-0.11	0.01	0.12	-0.02	0.04	0.11	77	49	36	-0.07	0.09	0.11	-0.03	0.16	0.19	67	50	49
Presidente Prudente (UR)	0.03	0.18	0.13	0.04	0.13	-0.01	71	49	39	0.01	0.11	0.15	-0.18	-0.14	-0.09	79	66	59
S.André-Capuava (NE)	0.17	0.18	0.3	0.17	0.2	0.3	42	28	25	0.25	0.33*	0.37*	0.32*	0.38*	0.42**	50	38	36
S.Bernardo-Paulicéia (NE)	0.18	0.32	0.39	0.15	0.26	0.24	44	33	25	0.11	0.01	-0.06	0.18	0.15	0.2	57	46	40
S.José Campos (NE)	0.18	0.02	-0.07	-0.04	0.02	-0.01	89	63	47	-0.12	-0.07	-0.16	0.18	0.16	0.13	73	60	50
S.José Campos-Jd.Satelite (NE)	0	0.04	0.03	0.15	0.01	-0.06	81	54	45	0.08	0.07	0.03	-0.05	0.02	-0.03	70	52	48
Santa Gertrudes (NE)	-0.1	0.22	0.31	-0.03	0.18	0.22	71	45	36	0.28*	0.41***	0.48***	0.27*	0.37**	0.45**	62	51	40
Santos-Ponta da Praia (NE)	0.04	0.08	0.13	0.08	0.11	0.18	58	45	37	0.04	0	0.05	0.25	0.21	0.27	56	47	40
São Caetano do Sul (ME)	0.07	0.21	0.42	0.02	0.13	0.38	34	23	17	0.42***	0.28	0.19	0.42**	0.46*	0.4	45	28	24
São José do Rio Preto (UR)	-0.18	0.09	0.32	-0.1	0.05	0.2	62	39	33	-0.29*	0.04	0.14	-0.34**	-0.04	0.06	64	48	45
Sorocaba (NE)	-0.03	-0.06	-0.02	0	-0.03	-0.11	55	38	29	0.15	0.15	0.15	0.12	0.14	0.13	50	42	38
Taboão da Serra (MI)	0.02	0.11	0.1	0.03	0.08	0.07	47	29	28	0.21	0.17	0.19	0.23	0.2	0.19	52	48	38
Tatui (NE)	-0.12	-0.12	0	-0.04	-0.05	0.07	72	50	42	-0.05	0.08	0.23	0.01	0.12	0.23	64	56	47
Taubaté (NE)	0.18	0.26*	0.46***	0.21*	0.23	0.4**	96	72	49	0.05	0.11	0.27*	0.08	0.18	0.33*	89	65	52
Classification	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Industrial	-0.05	-0.08	0	-0.18	-0.16	-0.09	91	61	52	-0.01	0.1	0.04	0	0.1	0.04	84	59	52
Peri-urban	0.02	0.09	0.15	0.1	0.15	0.16	229	158	122	-0.19**	-0.17*	-0.1	-0.17**	-0.11*	-0.07	209	162	149
Port handling	0.04	0.08	0.13	0.08	0.11	0.18	58	45	37	0.25	0.21	0.27	0.22	0.21	0.27	56	47	40
Sub-urban traffic	-0.03	0.01	0.02	0.02	0.02	0	1435	979	779	0.02	0.07*	0.13***	0.05	0.1*	0.15***	1473	1178	1018
Urban traffic	0.1*	0.13*	0.14*	0.08	0.08	0.1	423	282	242	0.22***	0.26***	0.25***	0.25***	0.28***	0.26***	513	394	345
Spatial representativity	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Micro-scale	0.04	0.06	0.02	0.01	0	-0.05	315	211	186	0.15***	0.14*	0.12*	0.15**	0.17**	0.16**	383	299	261
Medium-scale	0.13	0.17	0.27	0.07	0.05	0.16	75	53	38	0.23*	0.16	0.03	0.23*	0.25	0.14	90	62	55
Neighborhood	0.02	0.06*	0.09**	0.06*	0.07*	0.08*	1474	1009	810	0.03	0.07*	0.1***	0.08**	0.11***	0.14***	1460	1155	1008
Urban	-0.1	0	0.06	-0.06	0.01	-0.01	372	252	198	-0.18***	-0.08	0.04	-0.2***	-0.12*	-0.01	402	324	280

Note: MI - micro-scale; ME - medium scale; NE - neighborhood scale; and UR - urban scale. Significance level: * - p-value<0.05; ** - p-value<0.01; *** - p-value<0.001.

Table C.3 – Correlation between PM₁₀ measured at ground level by CETESB stations and AOD data measured by the MODIS sensor coupled to the Terra satellite.

Name	TERRA Satellite - 2019 (QA - 0000)									TERRA Satellite - 2020 (QA - 0000)								
	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km
Araçatuba (UR)	0.07	0.21*	0.19	0.07	0.16	0.13	123	103	96	-0.34***	-0.32***	-0.21*	-0.29**	-0.26**	-0.15	109	103	97
Araraquara (NE)	-0.08	-0.07	0.01	-0.07	-0.09	-0.04	111	95	84	-0.14	-0.08	-0.1	-0.21*	-0.17	-0.2	105	95	90
Bauru (NE)	0.03	0.04	0.05	0.07	0.1	0.13	98	90	82	-0.05	-0.01	0.1	-0.04	0.01	0.12	99	91	84
Campinas-Centro (MI)	0.03	0.05	-0.02	0.07	0.08	0	104	85	68	-0.17	-0.04	-0.06	-0.08	0.03	-0.02	89	79	73
Capão Redondo (NE)	-0.06	-0.07	-0.07	-0.08	-0.1	-0.11	66	57	55	-0.13	-0.07	-0.05	-0.1	-0.05	-0.03	68	51	48
Carapicuíba (NE)	0.04	0.02	-0.08	0.06	0.04	-0.05	77	71	58	0.08	0.18	0.23	0.04	0.14	0.18	80	71	62
Catanduva (UR)	-0.01	0.04	0.16	0.01	0.06	0.15	112	100	86	-0.02	0.03	0.06	-0.05	0	0	98	92	82
Cerqueira César (MI)	0.01	-0.01	-0.07	-0.02	-0.05	-0.1	86	66	61	0.15	0.25*	0.2	0.13	0.19	0.12	79	67	64
Congonhas (MI)	-0.12	-0.12	-0.18	-0.16	-0.2	-0.26	72	63	53	0.09	0.1	0.11	0.07	0.08	0.07	71	64	58
Cubatão-Centro (NE)	-0.17	-0.14	-0.15	-0.21	-0.17	-0.18	66	54	48	0.15	0.26	0.37**	0.26	0.32*	0.41**	56	50	45
Cubatão-V.Parisi (NE)	-0.05	0.06	0.06	-0.04	0	-0.04	75	58	52	0.25*	0.22	0.21	0.27*	0.27*	0.28*	66	54	50
Diadema (NE)	0.03	0.08	0.07	0.01	0	0	82	66	61	0.06	0.07	0.08	0.1	0.11	0.12	74	62	58
Grajaú-Parelheiros (MI)	-0.2	-0.18	-0.18	-0.31**	-0.3*	-0.28*	81	73	64	0.05	0.06	0.07	0.07	0.08	0.1	66	58	53
Guaratinguetá (NE)	-0.03	-0.01	0.01	0.02	0.04	0.03	93	84	76	0.02	0.01	-0.02	0.11	0.1	0.08	82	73	66
Guarulhos-Paço Municipal (UR)	0.08	0.06	0.08	0.08	0.03	0.03	86	77	65	0.08	-0.03	-0.02	0.04	-0.04	-0.03	75	67	61
Interlagos (NE)	0.07	0	-0.04	-0.02	-0.06	-0.08	72	56	52	-0.04	0.02	-0.02	-0.01	0.04	-0.01	67	56	50
Jacareí (NE)	-0.04	-0.07	-0.07	-0.09	-0.15	-0.14	91	72	69	0.05	-0.05	-0.02	-0.02	-0.09	-0.06	71	60	53
Juá (NE)	0.05	0.05	0.04	0.12	0.12	0.13	107	90	73	-0.1	-0.01	0.01	-0.12	0	0.01	103	88	83
Jundiaí (UR)	0.1	0.09	0.01	0.05	0.05	-0.04	95	86	77	-0.06	0.08	0.04	-0.09	0.04	0.01	91	83	82
Limeira (NE)	0.13	0.19	0.25*	0.15	0.22*	0.27*	99	88	76	-0.18	-0.15	-0.07	-0.17	-0.13	-0.06	87	81	75
Marg,Tietê-Pte Remédios (MI)	0.02	-0.04	-0.06	-0.09	-0.15	-0.16	89	80	71	0.08	0.12	0.17	0	-0.01	0.02	87	75	71
Marília (NE)	-0.01	-0.01	0.12	-0.02	-0.01	0.11	105	94	82	-0.09	-0.08	-0.06	-0.17	-0.19	-0.19	113	99	92
N.Senhora do Ó (ME)	-0.01	-0.07	-0.16	-0.1	-0.16	-0.24	75	67	59	0.28*	0.29*	0.32*	0.23	0.23	0.25	66	61	58
Osasco (MI)	0.14	0.04	-0.05	0.1	0.03	-0.07	84	75	69	-0.14	-0.14	-0.05	-0.11	-0.13	-0.05	88	72	68
Paulínia (NE)	0.09	0.03	0.03	0.13	0.07	0.06	120	108	100	-0.11	-0.07	-0.05	-0.06	-0.02	0	110	99	97
Piracicaba (NE)	-0.03	0.04	-0.01	0.02	0.09	0.04	100	84	75	-0.14	-0.01	0.03	-0.05	0.1	0.16	102	90	82
Presidente Prudente (UR)	-0.02	-0.01	0.05	-0.06	-0.07	-0.04	121	110	102	0.01	0.03	0.09	-0.16	-0.15	-0.13	111	106	97
S.André-Capuava (NE)	0.06	0.07	0.06	0.02	0.02	0	82	65	58	0.13	0.08	0.09	0.12	0.08	0.07	74	62	56
S.Bernardo-Paulicéia (NE)	0	-0.04	-0.12	0	-0.05	-0.1	86	75	62	0.07	0.08	-0.07	0.08	0.09	-0.06	76	66	56
S.José Campos (NE)	0.07	0.04	0.03	0.07	0.05	0.03	102	90	84	0.05	0.1	0.18	0.03	0.21	0.16	91	75	69
S.José Campos-Jd.Satelite (NE)	0.08	0.05	0.02	0.09	0.07	0.04	99	86	77	0.02	0.2	0.15	0.13	0.21	0.3*	86	72	66
Santa Gertrudes (NE)	0.11	0.12	0.17	0.13	0.14	0.18	114	94	88	-0.01	-0.01	0.03	0.11	0.12	0.18	107	99	92
Santos-Ponta da Praia (NE)	-0.16	-0.18	-0.19	-0.01	-0.04	-0.05	71	61	53	-0.09	-0.05	-0.05	0.17	0.15	0.17	68	59	54
São Caetano do Sul (ME)	-0.2	-0.22	-0.21	-0.3*	-0.31*	-0.34*	69	64	49	0.23	0.21	0.13	0.22	0.19	0.11	70	59	55
São José do Rio Preto (UR)	-0.02	0.06	0.08	0.02	0.07	0.09	111	94	90	-0.17	-0.09	-0.09	-0.22*	-0.14	-0.14	104	93	93
Sorocaba (NE)	0.03	0.09	0.11	0.09	0.15	0.18	90	74	70	-0.13	-0.02	-0.03	-0.15	-0.04	-0.05	89	78	76
Taboão da Serra (MI)	-0.03	-0.07	-0.1	-0.11	-0.16	-0.17	79	66	59	0	0.01	0.05	0.01	0.01	0.04	79	65	63
Tatui (NE)	-0.03	0.03	0.05	0.03	0.04	0.05	98	87	82	0.15	0.2	0.2	0.19	0.23*	0.23*	100	91	85
Taubaté (NE)	0.18	0.14	0.08	0.2*	0.14	0.09	110	98	90	0.26**	0.37***	0.38***	0.27*	0.37***	0.37**	86	79	74
Classification	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Industrial	-0.08	-0.04	-0.04	-0.11	-0.1	-0.11	141	112	100	-0.11	-0.1	-0.11	0.19	0.19	0.23	122	104	95
Peri-urban	0.08	0.14*	0.11	0.07	0.1	0.08	314	259	240	0.07	0.1	0.08	-0.06	-0.03	0.03	282	253	232
Port handling	-0.16	-0.18	-0.19	-0.01	-0.04	-0.05	71	61	53	-0.01	-0.04	-0.05	0.16	0.15	0.16	68	59	54
Sub-urban traffic	0.01	0.01	0.02	0.02	0.02	0.02	2249	1966	1771	0.02	0.02	0.02	-0.03	0.02	0.02	2093	1858	1730
Urban traffic	0.01	-0.02	-0.05	-0.05	-0.08*	-0.12**	826	708	612	-0.05	-0.08*	-0.12**	0.05	0.07*	0.08**	778	671	627
Spatial representativity	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Micro-scale	-0.03	-0.05	-0.09	-0.08*	-0.11*	-0.14**	595	508	445	-0.01	0.03	0.04	0	0	0	559	480	450
Medium-scale	-0.07	-0.1	-0.14	-0.17*	-0.2*	-0.24*	144	131	108	0.3	0.31	0.29	0.28***	0.29**	0.26**	136	120	113
Neighborhood	0.04	0.03	0.03	0.06**	0.05*	0.05*	2214	1897	1707	0	0.05	0.05	0.05*	0.09***	0.08***	2060	1801	1663
Urban	0.01	0.05	0.07	0.02	0.04	0.04	648	570	516	-0.16	-0.13	-0.11	-0.19***	-0.17***	-0.15***	588	544	512

Note: MI - micro-scale; ME - medium scale; NE - neighborhood scale; and UR - urban scale. Significance level: * - p-value<0.05;

** - p-value<0.01; *** - p-value<0.001.

Table C.4 – Correlation between PM_{2.5} measured at ground level by CETESB stations and AOD data measured by the MODIS sensor coupled to the Aqua satellite.

Name	AQUA Satellite - 2019 (QA - 0000)									AQUA Satellite - 2020 (QA - 0000)								
	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km
Campinas-V.União (UR)	0.14	-0.05	-0.11	-0.03	-0.15	-0.2	38	22	14	0.07	0.03	0.04	0.36**	0.37*	0.28	45	38	32
Congonhas (MI)	0.27	0.21	0.06	0.26	0.18	0.03	37	25	23	0.42***	0.44**	0.37*	0.31*	0.25	0.24	55	38	34
Grajaú-Parelheiros (MI)	0.23	0.17	-0.04	0.29	0.21	0.03	46	35	28	0.3*	0.26	0.19	0.26	0.23	0.3	56	45	41
Guarulhos-Paço Municipal (UR)	0.39**	0.21	-0.19	0.4*	0.23	-0.15	40	25	21	0.24	0.17	0.24	0.39**	0.29	0.3	48	41	35
Ibirapuera (UR)	0.14	-0.01	0.06	0.12	-0.08	0.03	37	25	19	0.48***	0.43**	0.51***	0.15	0.21	0.22	55	39	34
Itaim Paulista (UR)	0	0.1	-0.01	-0.02	0.08	-0.03	43	27	22	0.28*	0.24	0.3	0.12	0.27	0.28	48	38	35
Marg.Tietê-Pte Remédios (MI)	-0.3*	-0.07	-0.11	-0.3*	-0.05	-0.11	44	29	28	0.42***	0.31*	0.31	0.37**	0.43**	0.47***	56	42	37
Mooça (ME)	0.04	-0.15	-0.04	0.01	-0.13	0.02	36	19	14	0.37**	0.5***	0.51***	0.61***	0.7***	0.78***	51	34	31
Osasco (MI)	-0.01	-0.03	-0.04	0.02	0	-0.05	42	29	26	0.09	0.07	0.05	0.5**	0.39*	0.37	57	41	36
Parque D.Pedro II (NE)	-0.08	0.14	0.39	-0.18	0	0.25	37	23	14	0.42***	0.35*	0.43**	0.59***	0.68***	0.59**	49	37	33
Pico do Jaraguá (UR)	-0.28	-0.3	-0.32	-0.25	-0.29	-0.3	30	24	20	0.18	0.23	0.3	-0.11	0.22	0.22	41	28	22
Piracicaba (NE)	0.15	0.19	0.22	0.15	0.12	0.21	82	51	36	0.09	0.24	0.24	0.09	0.04	0.03	67	50	49
S.Bernardo-Centro (NE)	-0.05	-0.03	0.07	-0.03	0.01	0.13	46	33	29	0.39***	0.42***	0.31	0.5***	0.51**	0.62***	55	47	38
S.José Campos-Jd.Satelite (NE)	0.1	0.2	0.21	0.07	0.08	0.09	81	54	45	0.37***	0.4***	0.43***	0.42**	0.48**	0.5**	71	53	49
Santa Gertrudes (NE)	0.17	0.39*	0.39*	0.22	0.36*	0.38*	55	36	30	0.5***	0.57***	0.61***	0.42**	0.6***	0.63***	44	33	25
Santana (ME)	0.28	0.12	0.28	0.22	0.07	0.11	40	23	19	0.3*	0.37*	0.38*	0.34*	0.28	0.43*	47	36	33
Santos-Ponta da Praia (NE)	-0.04	0.13	0.19	-0.11	0.04	0.07	49	36	30	0.59***	0.49***	0.5**	0.21	0.25	0.39	39	32	27
São Caetano do Sul (ME)	-0.06	0.05	0.12	-0.1	-0.07	0.02	33	22	16	0.54***	0.47**	0.33	0.37**	0.38**	0.28	44	27	23
São José do Rio Preto (UR)	-0.2	-0.05	-0.02	-0.16	-0.06	-0.09	62	39	33	-0.07	0.24	0.25	0.3*	0.41*	0.43*	62	47	45
Taubaté (NE)	0.18	0.26*	0.46***	0.21*	0.23	0.4**	96	72	49	0.05	0.11	0.27*	0.08	0.18	0.33*	89	65	52
Classification	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Peri-urban	0.15	0.19	0.22	0.15	0.12	0.21	82	51	36	0.09	0.24	0.24	0.12	0.27	0.28	67	50	49
Port handling	-0.04	0.13	0.19	-0.11	0.04	0.07	49	36	30	0.59***	0.49***	0.5**	0.5**	0.39*	0.37	39	32	27
Regional background	-0.28	-0.3	-0.32	-0.25	-0.29	-0.3	30	24	20	0.18	0.23	0.3	0.21	0.25	0.39	41	28	22
Sub-urban traffic	0.08	0.16**	0.16*	0.13**	0.17**	0.19**	424	291	228	0.23***	0.28***	0.28***	0.3***	0.35***	0.36***	422	328	282
Urban park	0.14	-0.01	0.06	0.12	-0.08	0.03	37	25	19	0.48***	0.43**	0.51***	0.5***	0.51**	0.62***	55	39	34
Urban traffic	0.06	0.08	0.06	0.04	0.06	0.04	352	222	183	0.33***	0.3***	0.3***	0.33***	0.35***	0.36***	455	334	297
Spatial representativity	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Micro-scale	0.03	0	-0.08	0.06	0.05	-0.03	169	118	105	0.36***	0.3***	0.27***	0.33***	0.29***	0.28***	224	166	148
Medium-scale	0.11	0.14	0.24	0.06	0.04	0.13	109	64	49	0.34***	0.37***	0.34***	0.35***	0.43***	0.42***	142	97	87
Neighborhood	0.05	0.16**	0.22***	0.06	0.12*	0.18**	446	305	233	0.24***	0.24***	0.25***	0.21***	0.2***	0.22***	414	317	273
Urban	0	-0.04	-0.12	-0.03	-0.09	-0.21*	210	137	108	0.16**	0.18**	0.22***	0.16**	0.21**	0.24***	251	190	168

Note: MI - micro-scale; ME - medium scale; NE - neighborhood scale; and UR - urban scale. Significance level: * - p-value<0.05;

** - p-value<0.01; *** - p-value<0.001.

Table C.5 – Correlation between PM_{2.5} measured at ground level by CETESB stations and AOD data measured by the MODIS sensor coupled to the Terra satellite.

Name	TERRA Satellite - 2019 (QA - 0000)									TERRA Satellite - 2020 (QA - 0000)								
	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km
Campinas-V.União (UR)	0.25*	0.19	0.13	-0.18	-0.17	-0.24	85	69	63	-0.13	-0.09	-0.11	-0.1	-0.08	-0.07	85	76	71
Congonhas (MI)	-0.11	-0.12	-0.17	-0.15	-0.17	-0.14	69	59	50	-0.02	-0.01	0.01	-0.06	-0.02	0	71	64	58
Grajaú-Parelheiros (MI)	-0.1	-0.12	-0.12	0.04	-0.01	0.03	68	61	54	-0.07	-0.03	-0.02	0.04	-0.02	0.02	57	51	46
Guarulhos-Paço Municipal (UR)	0.10	0.05	0.11	-0.05	-0.08	-0.14	84	75	63	0.10	0.04	0.08	0.01	0.04	0.06	76	68	62
Ibirapuera (UR)	0.02	0.03	-0.01	0.15	0.07	0.05	79	62	54	0.12	0.09	0.09	-0.06	-0.03	0.05	74	69	63
Itaim Paulista (UR)	0	-0.02	-0.06	0.05	0.07	0.04	79	67	63	0.02	0.11	0.07	0.02	0.14	0.2	76	67	62
Marg.Tietê-Pte Remédios (MI)	0.01	-0.02	-0.1	-0.07	-0.08	-0.1	88	79	70	0.06	0.11	0.15	0	0.16	0.15	87	75	71
Mooça (ME)	-0.07	-0.07	-0.06	0.15	0.15	0.17	80	68	56	0.12	0.15	0.19	0.17	0.12	0.23	80	68	63
Osasco (MI)	0.13	0.01	-0.02	0.05	0.03	0	87	78	72	-0.14	-0.08	0.01	0.23	0.16	0.23	87	71	67
Parque D.Pedro II (NE)	-0.19	-0.22	-0.14	-0.18	-0.2	-0.22	82	68	54	0.08	0.11	0.15	0.28*	0.21	0.15	75	65	59
Pico do Jaraguá (UR)	0.14	0.12	0.15	-0.01	0.03	0.05	58	50	41	0.23	0.24	0.27	-0.12	-0.05	-0.05	64	56	49
Piracicaba (NE)	0.04	0.09	0.04	0.21	0.17	0.11	104	89	80	-0.04	0.07	0.13	-0.17	-0.13	-0.14	104	91	83
S.Bernardo-Centro (NE)	-0.1	-0.02	0.02	-0.07	-0.06	-0.13	82	69	59	0.11	0.02	-0.04	0.03	-0.01	0.02	71	59	51
S.José Campos-Jd.Satelite (NE)	-0.05	-0.08	-0.11	-0.05	-0.08	-0.13	100	87	78	-0.06	0.06	0.05	0.04	0.12	0.07	86	72	66
Santa Gertrudes (NE)	0.04	0.02	0.07	-0.06	-0.08	-0.07	86	72	68	0.08	0.03	0.12	0.11	0.12	0.15	75	69	64
Santana (ME)	-0.16	-0.16	-0.14	-0.17	-0.19	-0.17	76	64	56	0.13	0.15	0.17	0.06	0.06	0.1	67	61	54
Santos-Ponta da Praia (NE)	-0.04	-0.08	-0.09	0.09	0.1	0.14	65	57	49	-0.02	0.02	0.07	0.22	0.23	0.25	64	55	50
São Caetano do Sul (ME)	-0.1	-0.13	-0.15	-0.16	-0.08	-0.07	69	64	49	0.27*	0.24	0.18	0.14	0.1	0.08	70	59	55
São José do Rio Preto (UR)	-0.09	-0.04	-0.03	-0.14	-0.13	-0.13	110	93	89	-0.11	-0.04	-0.04	0.03	0.06	0.07	104	93	93
Taubaté (NE)	0.18	0.14	0.08	0.2*	0.14	0.09	110	98	90	0.26*	0.37***	0.38***	0.27*	0.37***	0.37**	86	79	74
Classification	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Peri-urban	0.04	0.09	0.04	0.05	0.07	0.04	104	89	80	-0.04	0.07	0.13	0.02	0.14	0.2	104	91	83
Port handling	-0.04	-0.08	-0.09	0.05	0.03	0	65	57	49	-0.02	0.02	0.07	0.23	0.16	0.23	64	55	50
Regional background	0.14	0.12	0.15	0.09	0.1	0.14	58	50	41	0.23	0.24	0.27	0.22	0.23	0.25	64	56	49
Sub-urban traffic	-0.01	-0.01	-0.03	-0.01	-0.01	-0.01	641	549	501	0	0.06	0.05	0.02	0.06	0.07	564	499	465
Urban park	0.02	0.03	-0.01	-0.07	-0.1*	-0.11*	79	62	54	0.12	0.09	0.09	0.03	-0.01	0.02	74	69	63
Urban traffic	-0.04	-0.07	-0.07	-0.07	-0.06	-0.13	714	622	533	0.05	0.07	0.08	0.03	0.03	0.04	689	598	551
Spatial representativity	Without correction			PBLH - Corrected			n samples			Without correction			PBLH - Corrected			n samples		
Micro-scale	-0.06	-0.1	-0.14*	-0.08	-0.11	-0.13*	312	277	246	0.03	0.05	0.08	0.03	0.02	0.06	302	261	242
Medium-scale	-0.08	-0.08	-0.07	-0.1	-0.11	-0.1	225	196	161	0.16*	0.17*	0.17*	0.13*	0.12	0.12	217	188	172
Neighborhood	-0.03	-0.03	-0.03	-0.01	-0.01	-0.01	629	540	478	0.01	0.09	0.11*	0.09*	0.13**	0.16***	561	490	447
Urban	0.03	0.01	0	0.01	-0.02	-0.03	495	416	373	0	0.01	0.01	-0.03	-0.02	-0.01	479	429	400

Note: MI - micro-scale; ME - medium scale; NE - neighborhood scale; and UR - urban scale. Significance level: * - p-value<0.05;

** - p-value<0.01; *** - p-value<0.001.

Table C.6 – Correlation between PM₁₀/PM_{2.5} measured at ground level by CETESB stations and AOD data (observed and corrected) measured by the MODIS (Aqua and Terra satellites) for the HUWRMs of São Paulo State. The data shown represent only the smallest extraction radius (3 km).

		AOD	PM ₁₀	Correlation		AOD	PM _{2.5}	Correlation	
HUWRM		Mean±SD	Mean±SD	AOD	AOD/PBLH	Mean±SD	Mean±SD	AOD	AOD/PBLH
AQUA-TERRA combined	2	0.08±0.06	24.14±15.23	0.06*	0.11***	0.09±0.06	14.00±9.66	0.08*	0.1*
	5	0.07±0.05	27.47±16.42	0.02	0.1***	0.07±0.05	11.11±7.68	0.05	0.16***
	6	0.09±0.06	24.53±16.18	0.04*	0.02	0.09±0.06	12.38±9.42	0.06**	0.06**
	7	0.10±0.06	40.76±39.43	-0.08*	-0.14**	0.10±0.06	10.99±9.80	0.01	0.05
	10	0.07±0.05	21.52±17.79	0.04	0.11*	NA	NA	NA	NA
	13	0.06±0.04	21.78±11.91	-0.01	-0.02	NA	NA	NA	NA
	15	0.06±0.04	28.38±15.94	0.02	0.03	0.06±0.04	12.28±6.70	-0.03	0.02
	19	0.06±0.04	27.08±14.66	-0.02	-0.04	NA	NA	NA	NA
	21	0.07±0.03	18.32±12.17	0.05	-0.07	NA	NA	NA	NA
	22	0.06±0.04	18.50±8.69	0.08	-0.06	NA	NA	NA	NA
HUWRM		Mean±SD	Mean±SD	AOD	AOD/PBLH	Mean±SD	Mean±SD	AOD	AOD/PBLH
AQUA	2	0.09±0.05	23.37±16.8	0.08	0.11*	0.09±0.06	14.78±8.71	0.26***	0.24***
	5	0.08±0.05	24.26±12.18	0.16***	0.18***	0.08±0.05	9.78±8.26	0.27***	0.27***
	6	0.09±0.07	24.47±13.45	0.11**	0.12***	0.09±0.06	11.29±7.52	0.19***	0.23***
	7	0.11±0.08	32.8±31.25	-0.03	-0.17*	0.11±0.05	6.82±8.1	0.39**	0.27**
	10	0.08±0.05	18.14±10.8	0	0.01	NA	NA	NA	NA
	13	0.07±0.04	20.99±9.01	-0.02	-0.09	NA	NA	NA	NA
	15	0.06±0.04	24.65±10.68	0.13	0.05	0.06±0.04	10.51±5.45	0.12	0.08
	19	0.07±0.04	28.66±14.59	-0.13	-0.14	NA	NA	NA	NA
	21	0.07±0.03	18.25±9.09	0.03	-0.1	NA	NA	NA	NA
	22	0.07±0.03	18.58±7.88	0.13	-0.07	NA	NA	NA	NA
HUWRM		Mean±SD	Mean±SD	AOD	AOD/PBLH	Mean±SD	Mean±SD	AOD	AOD/PBLH
TERRA	2	0.08±0.06	24.66±14.06	0.06	0.08*	0.09±0.06	15.29±10.59	0.05	0.03
	5	0.07±0.05	30.38±18.15	0.01	0.07*	0.07±0.05	12.68±8.34	0.02	0.1*
	6	0.08±0.06	25.45±17.89	0.02	-0.03	0.09±0.06	12.93±10.09	0.02	-0.01
	7	0.09±0.06	52.01±48.51	-0.02	-0.09	0.09±0.06	13.98±9.85	-0.04	0.06
	10	0.07±0.05	23.18±20.18	0.08	0.11*	NA	NA	NA	NA
	13	0.06±0.04	22.12±12.94	-0.01	0.01	NA	NA	NA	NA
	15	0.06±0.04	29.98±17.51	0.04	0.01	0.06±0.05	13.03±7.05	-0.04	-0.02
	19	0.06±0.04	26.25±14.67	0.01	0.01	NA	NA	NA	NA
	21	0.06±0.03	18.35±13.46	0.04	-0.03	NA	NA	NA	NA
	22	0.06±0.05	18.47±9.08	0.06	-0.07	NA	NA	NA	NA

Note: AOD – correlation between PM and observed AOD; AOD/PBLH - correlation between PM and corrected AOD (AOD/PBLH). SD - standard deviation. Significance level: * - p-value<0.05; ** - p-value<0.01; *** - p-value<0.001.

Table C.7 – Correlation between NO₂ measured at ground level by CETESB stations and NO₂ tropospheric vertical column density (VCD) retrieved from TROPOMI (S-5P). The non-significant correlations (p-value > 0.05) are shown in bold.

Name	2019									2020								
	Spearman			p-value			n samples			Spearman			p-value			n samples		
	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km	10km	5km	3km
Araraquara (NE)	0.17	0.12	0.30	0.06	0.25	0.03	121	88	52	0.03	0.03	0.11	0.78	0.79	0.39	127	90	64
Bauru (NE)	-0.04	-0.06	-0.04	0.65	0.55	0.76	127	98	53	0.27	0.25	0.36	0.00	0.02	0.00	115	93	63
Campinas-Taquaral (UR)	0.44	0.46	0.44	0.00	0.00	0.00	126	98	52	0.38	0.30	0.18	0.00	0.03	0.32	68	54	32
Catanduva (UR)	0.14	0.13	0.26	0.11	0.19	0.04	130	103	64	0.03	0.13	0.22	0.75	0.18	0.08	130	100	64
Cerqueira César (MI)	0.48	0.44	0.52	0.00	0.00	0.00	84	64	34	0.47	0.56	0.58	0.00	0.00	0.00	83	57	37
Congonhas (MI)	0.51	0.61	0.68	0.00	0.00	0.00	94	61	30	0.25	0.26	0.23	0.02	0.03	0.11	94	72	48
Cubatão-Centro (NE)	0.72	0.78	0.73	0.00	0.00	0.00	69	54	35	0.66	0.75	0.81	0.00	0.00	0.00	74	50	36
Cubatão-V.Parisi (NE)	0.41	0.47	0.38	0.00	0.00	0.07	73	51	24	0.48	0.56	0.56	0.00	0.00	0.00	79	53	32
Guaratinguetá (NE)	0.21	0.24	0.19	0.02	0.02	0.13	115	101	61	0.31	0.38	0.46	0.00	0.00	0.00	102	87	61
Guarulhos-Pimentas (NE)	0.25	0.33	0.45	0.01	0.01	0.00	98	64	37	0.43	0.47	0.47	0.00	0.00	0.00	92	60	41
Ibirapuera (UR)	0.70	0.71	0.81	0.00	0.00	0.00	91	59	37	0.70	0.68	0.70	0.00	0.00	0.00	79	57	37
Itaim Paulista (UR)	0.55	0.63	0.63	0.00	0.00	0.00	86	54	34	0.61	0.69	0.71	0.00	0.00	0.00	84	64	40
Jau (UR)	0.06	-0.03	-0.04	0.49	0.77	0.77	134	95	58	0.01	-0.01	-0.08	0.92	0.95	0.53	132	103	67
Marg.Tietê-Pte																		
Remédios (MI)	0.40	0.39	0.40	0.00	0.00	0.01	98	69	43	0.33	0.33	0.41	0.00	0.00	0.00	98	77	48
Marília (UR)	-0.11	-0.04	-0.04	0.22	0.68	0.76	124	92	53	0.13	0.10	0.27	0.15	0.33	0.02	122	104	69
Osasco (MI)	0.66	0.72	0.59	0.00	0.00	0.00	102	72	35	0.74	0.76	0.81	0.00	0.00	0.00	105	66	39
Parque D.Pedro II (NE)	0.70	0.79	0.86	0.00	0.00	0.00	69	48	24	0.64	0.65	0.71	0.00	0.00	0.00	95	71	45
Pico do Jaraguá (UR)	0.45	0.62	0.71	0.00	0.00	0.00	69	54	30	0.64	0.72	0.74	0.00	0.00	0.00	102	72	46
Pinheiros (MI)	0.58	0.65	0.56	0.00	0.00	0.00	71	60	32	0.57	0.60	0.73	0.00	0.00	0.00	91	66	49
Piracicaba (UR)	0.14	0.21	0.38	0.12	0.03	0.00	135	108	64	0.19	0.23	0.31	0.06	0.04	0.02	100	85	61
Presidente Prudente (UR)	-0.14	-0.09	-0.18	0.13	0.37	0.18	125	94	57	0.11	0.17	0.24	0.23	0.08	0.03	129	103	81
S.André-Capuava (NE)	0.64	0.71	0.63	0.00	0.00	0.00	49	30	22	0.58	0.58	0.59	0.00	0.00	0.00	91	65	42
S.Bernardo-Centro (NE)	0.67	0.74	0.86	0.00	0.00	0.00	77	58	36	0.69	0.68	0.64	0.00	0.00	0.00	105	86	58
S.José Campos (NE)	0.45	0.45	0.50	0.00	0.00	0.00	126	98	54	0.40	0.47	0.41	0.00	0.00	0.00	115	84	56
S.José Campos-																		
Jd.Satelite (NE)	0.52	0.56	0.46	0.00	0.00	0.00	119	90	53	0.51	0.62	0.56	0.00	0.00	0.00	90	69	45
Santa Gertrudes (NE)	0.29	0.33	0.38	0.00	0.00	0.01	132	103	52	0.36	0.38	0.45	0.00	0.00	0.00	126	101	67
Santos-Ponta da Praia																		
(NE)	0.61	0.63	0.66	0.00	0.00	0.00	102	79	47	0.66	0.68	0.67	0.00	0.00	0.00	101	79	59
São Caetano do Sul (ME)	0.66	0.77	0.74	0.00	0.00	0.00	70	42	29	0.58	0.52	0.56	0.00	0.00	0.00	95	73	49
São José do Rio Preto																		
(UR)	0.04	0.05	0.06	0.65	0.64	0.68	132	91	48	0.22	0.23	0.28	0.02	0.03	0.03	119	89	59
Sorocaba (UR)	0.49	0.56	0.71	0.00	0.00	0.00	91	64	33	0.43	0.55	0.54	0.00	0.00	0.00	114	85	56
Tatuí (UR)	0.27	0.28	0.28	0.00	0.00	0.03	121	101	63	0.34	0.38	0.44	0.00	0.00	0.00	125	103	71
Taubaté (NE)	0.44	0.42	0.50	0.00	0.00	0.00	120	95	53	0.44	0.43	0.38	0.00	0.00	0.00	125	102	73
Classification	Spearman			p-value			n samples			Spearman			p-value			n samples		
Industrial	0.41	0.44	0.47	0.00	0.00	0.00	142	105	59	0.38	0.41	0.49	0.00	0.00	0.00	153	103	68
Peri-urban	0.26	0.31	0.38	0.00	0.00	0.00	261	206	116	0.05	0.08	0.12	0.50	0.33	0.26	168	139	93
Port handling	0.61	0.63	0.66	0.00	0.00	0.00	102	79	47	0.66	0.68	0.67	0.00	0.00	0.00	101	79	59
Regional background	0.45	0.62	0.71	0.00	0.00	0.00	69	54	30	0.64	0.72	0.74	0.00	0.00	0.00	102	72	46
Sub-urban traffic	0.40	0.42	0.44	0.00	0.00	0.00	1892	1435	827	0.45	0.48	0.49	0.00	0.00	0.00	1868	1459	995
Urban park	0.70	0.71	0.81	0.00	0.00	0.00	91	59	37	0.70	0.68	0.70	0.00	0.00	0.00	79	57	37
Urban traffic	0.34	0.33	0.30	0.00	0.00	0.00	723	500	283	0.35	0.38	0.43	0.00	0.00	0.00	836	611	397
Spatial representativity	Spearman			p-value			n samples			Spearman			p-value			n samples		
Micro-scale	0.38	0.38	0.35	0.00	0.00	0.00	449	326	174	0.33	0.37	0.41	0.00	0.00	0.00	471	338	221
Medium-scale	0.66	0.77	0.74	0.00	0.00	0.00	70	42	29	0.58	0.52	0.56	0.00	0.00	0.00	95	73	49
Neighborhood	0.63	0.65	0.68	0.00	0.00	0.00	1397	1057	603	0.62	0.63	0.64	0.00	0.00	0.00	1437	1090	742
Urban	0.37	0.39	0.43	0.00	0.00	0.00	1364	1013	593	0.42	0.45	0.48	0.00	0.00	0.00	1304	1019	683

Note: MI - micro-scale; ME - medium scale; NE - neighborhood scale; and UR - urban scale.

Table C.8 – Correlation between NO₂ measured at ground level by CETESB stations and NO₂ tropospheric vertical column density (VCD) retrieved from TROPOMI (S-5P) for the HUWRMs of São Paulo State. The data shown represent only the smallest extraction radius (3 km). The non-significant correlations (p-value > 0.05) are shown in bold.

HUWRM	NO ₂ trop. VDC (Pmolec/cm ²)	NO ₂ concentration (µg/m ³)	Correlation	<i>p</i> -value
	Mean±SD	Mean±SD		
2	2.31±1.69	7.44±5.80	0.57	0.000
5	2.33±1.45	8.12±6.38	0.14	0.000
6	6.45±5.10	25.53±21.97	0.51	0.000
7	7.22±7.68	24.27±21.67	0.68	0.000
10	1.81±1.43	6.91±3.90	0.55	0.000
13	1.17±0.76	4.94±2.28	0.09	0.083
15	1.33±0.80	5.12±3.06	0.21	0.001
21	1.03±0.82	5.14±2.78	0.18	0.042
22	1.05±0.68	3.45±1.81	0.07	0.422

Figure C.1 - Evolution in the displacement of cell phone users during the first half of 2020. The vertical dashed line indicates the beginning of mobility restrictions. Evolution in the displacement of cell phone users during the first half of 2020. The vertical dashed line indicates the beginning of mobility restrictions.

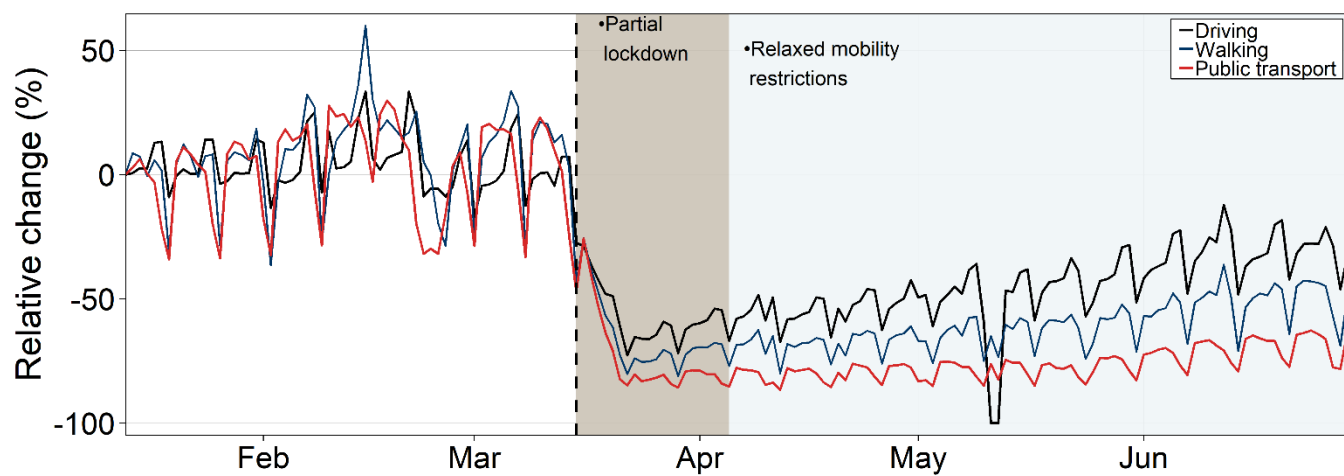


Figure C.2 - Violin plots of MODIS-AOD (Aqua and Terra satellites) and intervals of ground-based PM₁₀ for the periods: (A) before the beginning of mobility restriction in 2019; (B) during the mobility restriction in 2019; (C) before the mobility restriction in 2020; and (D) during the mobility restriction in 2020. The data shown represent only the smallest extraction radius (3 km).

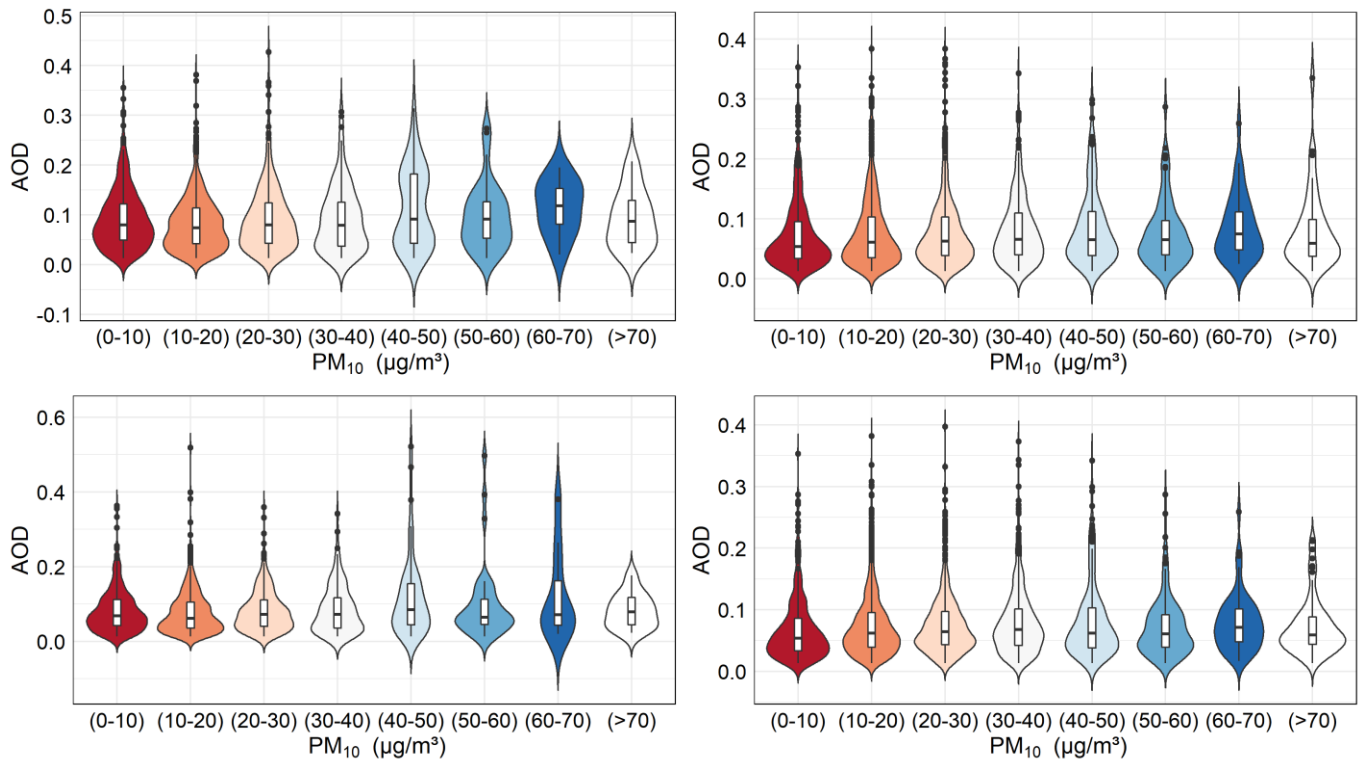


Figure C.3 - Violin plots of MODIS-AOD (Aqua and Terra satellites) and intervals of ground-based PM_{2.5} for the periods: (A) before the beginning of mobility restriction in 2019; (B) during the mobility restriction in 2019; (C) before the mobility restriction in 2020; and (D) during the mobility restriction in 2020. The data shown represent only the smallest extraction radius (3 km).

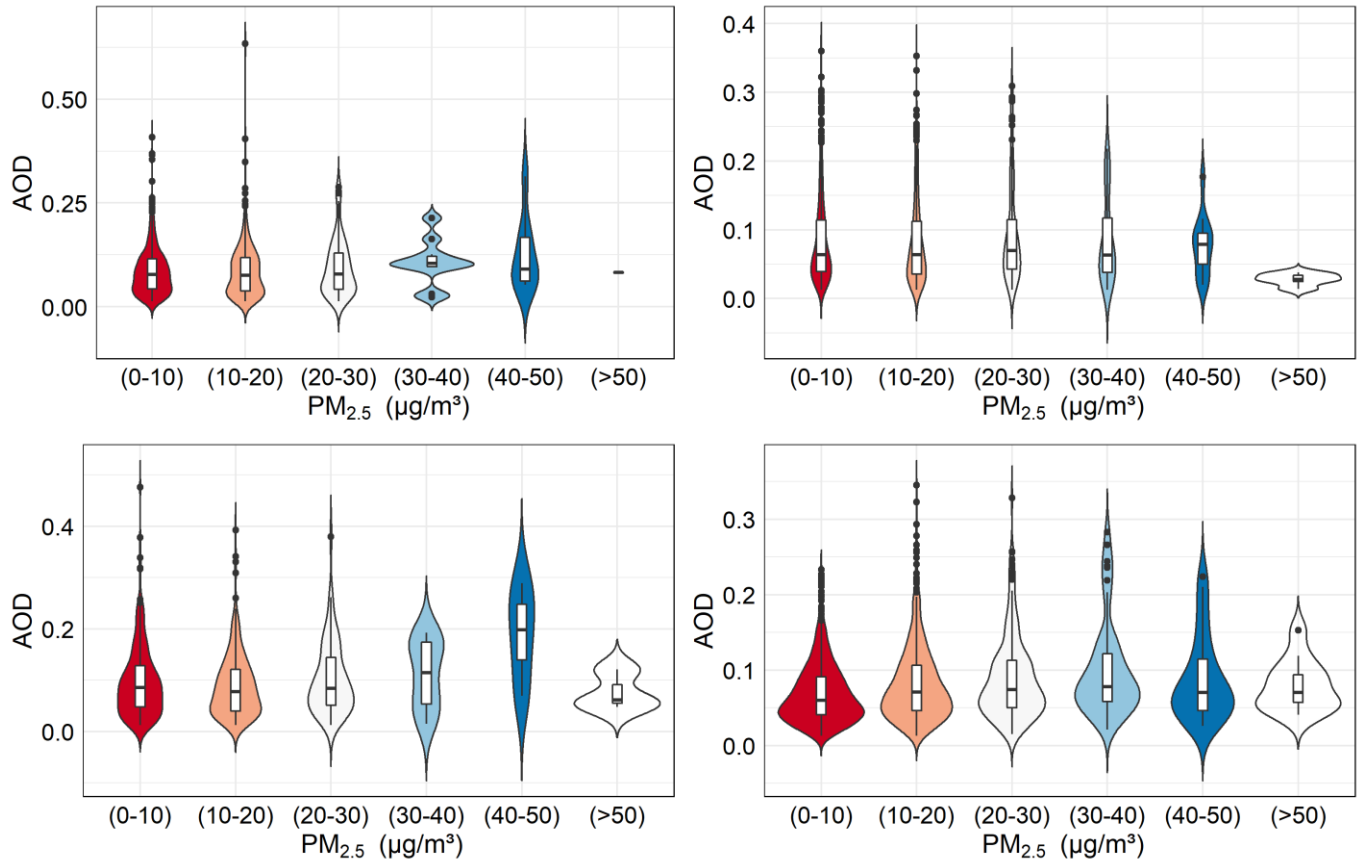
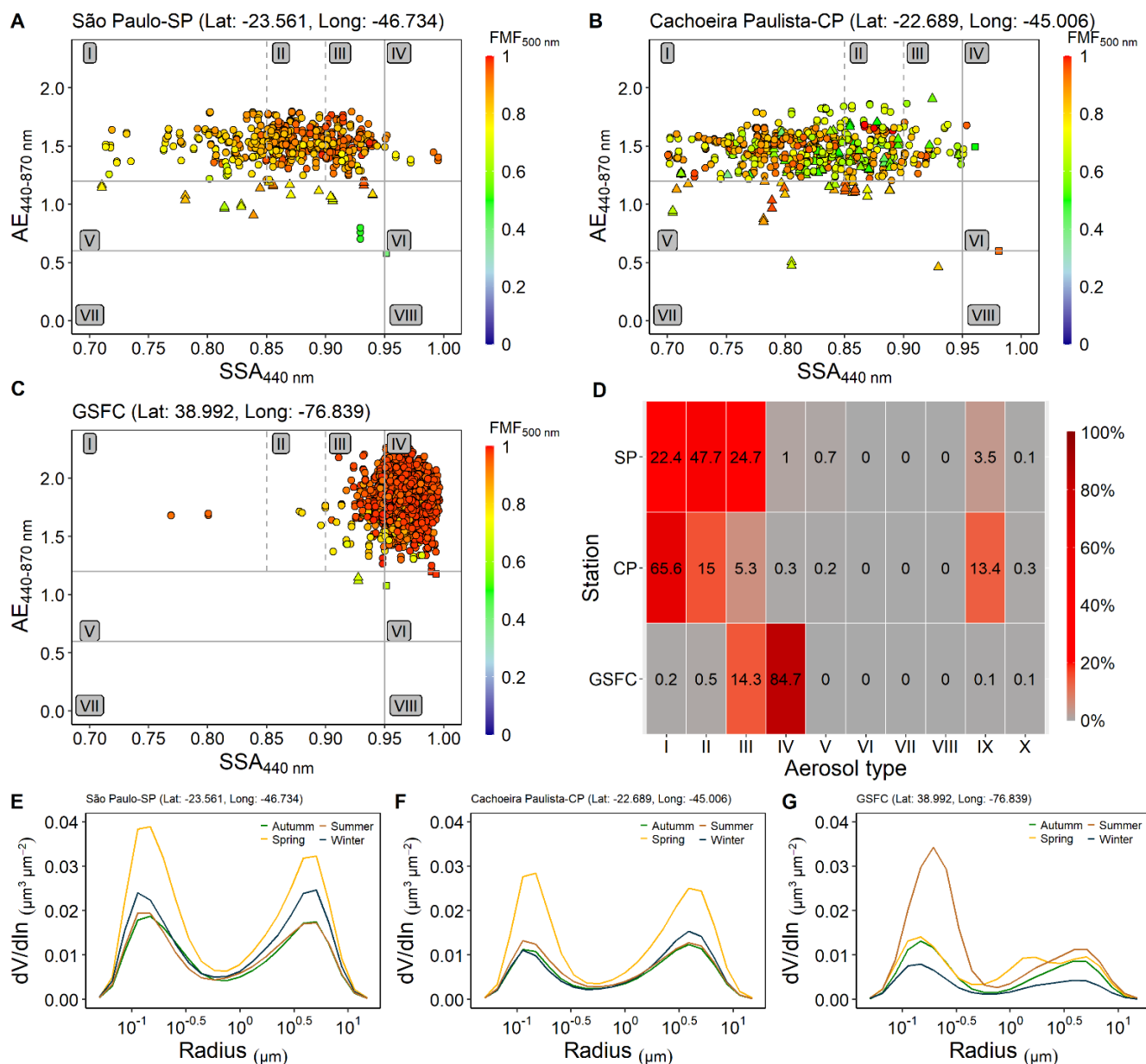


Figure C.4 - Aerosol classification based on (LOGOTHETIS; SALAMALIKIS; KAZANTZIDIS, 2020).



Note: The graphs were developed with all the data available for the AERONET sites at São Paulo (A, 2001-2017), Cachoeira Paulista (B, 2016-2020), and GSFC (C, 1993-2016). Heatmap with the percentages of aerosol type at every station is shown in (D), and seasonal variation in the aerosol volume size distribution for São Paulo (E), Cachoeira Paulista (F), and GSFC (G) are shown at the bottom. The solid and dashed lines separate the aerosol type. The dots are showing the Aerosol Type between I and VIII, triangles Aerosol Type IX, and squares Aerosol Type X. Aerosol classification: I. Fine highly absorbing ($SSA \leq 0.85$, $FMF > 0.6$ and $AE > 1.2$). II. Fine moderately absorbing ($0.85 < SSA \leq 0.9$, $FMF > 0.6$ and $AE > 1.2$). III. Fine slightly absorbing ($0.9 < SSA \leq 0.95$, $FMF > 0.6$ and $AE > 1.2$). IV. Fine non-absorbing ($SSA > 0.95$, $FMF > 0.6$ and $AE > 1.2$). V. Mixed absorbing ($SSA \leq 0.95$, $0.4 \leq FMF \leq 0.6$ and $0.6 \leq AE \leq 1.2$). VI. Mixed non-absorbing ($SSA > 0.95$, $0.4 \leq FMF \leq 0.6$ and $0.6 \leq AE \leq 1.2$). VII. Coarse absorbing ($SSA \leq 0.95$, $FMF < 0.4$ and $AE < 0.6$). VIII. Coarse non-absorbing ($SSA > 0.95$, $FMF < 0.4$ and $AE < 0.6$). IX. Other absorbing (divergences between the intervals in FMF and AE and $SSA \leq 0.95$); X. Other non-absorbing (divergences between the intervals in FMF and AE and $SSA > 0.95$). FMF (500nm) was obtained considering the Aerosol Optical Depth version 3 and Spectral Deconvolution Algorithm (SDA) at level 2.0 for AERONET sites at São Paulo and GSFC and at level 1.5 for Cachoeira Paulista. SSA (440nm) were obtained through the data set Inversions version 2 and considering the same data levels of FMF (500nm). The seasons consider typical northern weather seasons for AERONET – GSFC ("Winter": Dec, Jan, Feb; "Spring": Mar, Apr, May; "Summer": Jun, Jul, Aug; "Autumn": Sep, Oct, Nov) and southern weather seasons for AERONET sites at São Paulo and Cachoeira Paulista ("Summer": Dec, Jan, Feb; "Autumn": Mar, Apr, May; "Winter": Jun, Jul, Aug; "Spring": Sep, Oct, Nov).

Figure C.5 - Comparison between the AERONET data for AOD at 550 nm and those derived from the MAIAC algorithm for the Aqua satellite (left) and Terra satellite (right) in the MASP for the period between 2000 and 2020. Black dashed and solid lines indicate the MAIAC expected error (EE) envelope, defined as $\pm (0.05 + 0.15 \times \text{AOD})$ and 1:1 lines, respectively. The solid red line displays the regression line. r is the linear correlation between MAIAC AODs and AERONET AODs; n is the number of samples; RMSE is the root mean square error; MAE is the mean absolute error; and within EE is the fraction of MAIAC retrievals that fell within the EE envelope. The data colocation was carried out through the spatial window of 10x10km and temporal window of ± 60 min.

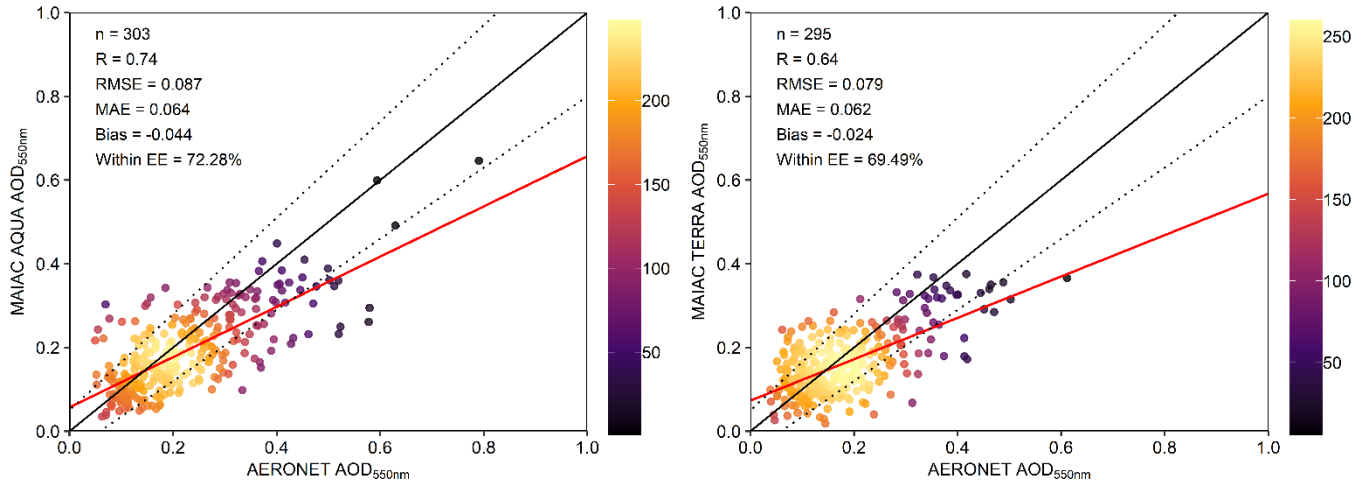


Figure C.6 - Violin plots of satellite-based NO₂ tropospheric vertical column density (VCD) derived from TROPOMI (S-5P) and intervals of ground-based NO₂ for the periods: (A) before the beginning of mobility restriction in 2019; (B) during the mobility restriction in 2019; (C) before the mobility restriction in 2020; and (D) during the mobility restriction in 2020. The data shown represent only the smallest extraction radius (3 km).

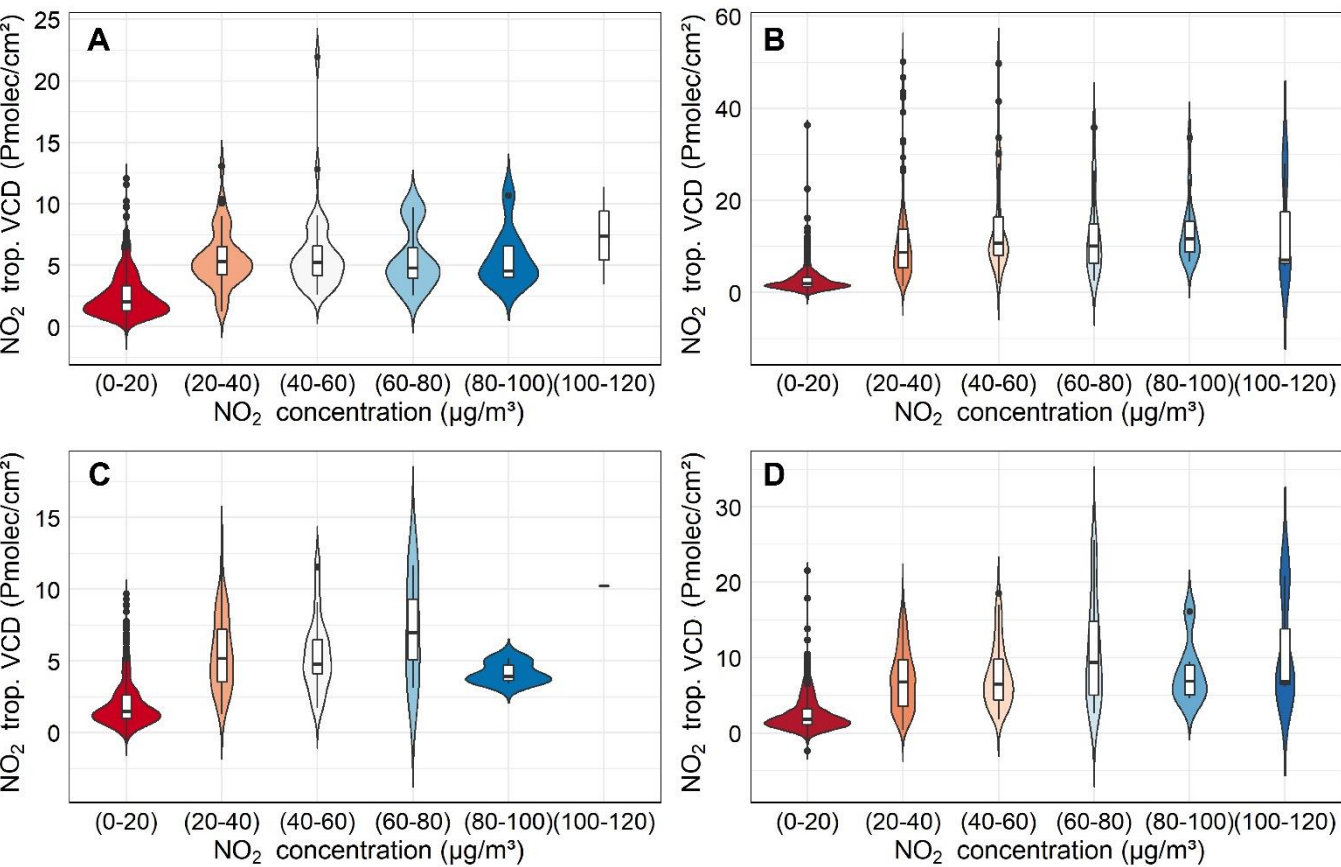


Figure C.7 - Correlation between ground-based measurements of PM_{2.5} and NO₂ and tropospheric NO₂ vertical column densities for the periods: (A) before the beginning of mobility restriction in 2019; (B) during the mobility restriction in 2019; (C) before the mobility restriction in 2020; and (D) during the mobility restriction in 2020. The distribution of each variable is shown on the main diagonal. Bellow the diagonal the bivariate scatter plots with a fitted smooth line (loess) are displayed. Above the diagonal the value of the Spearman correlation coefficient plus the significance level as stars are displayed (*) $p \leq 0.001$).**

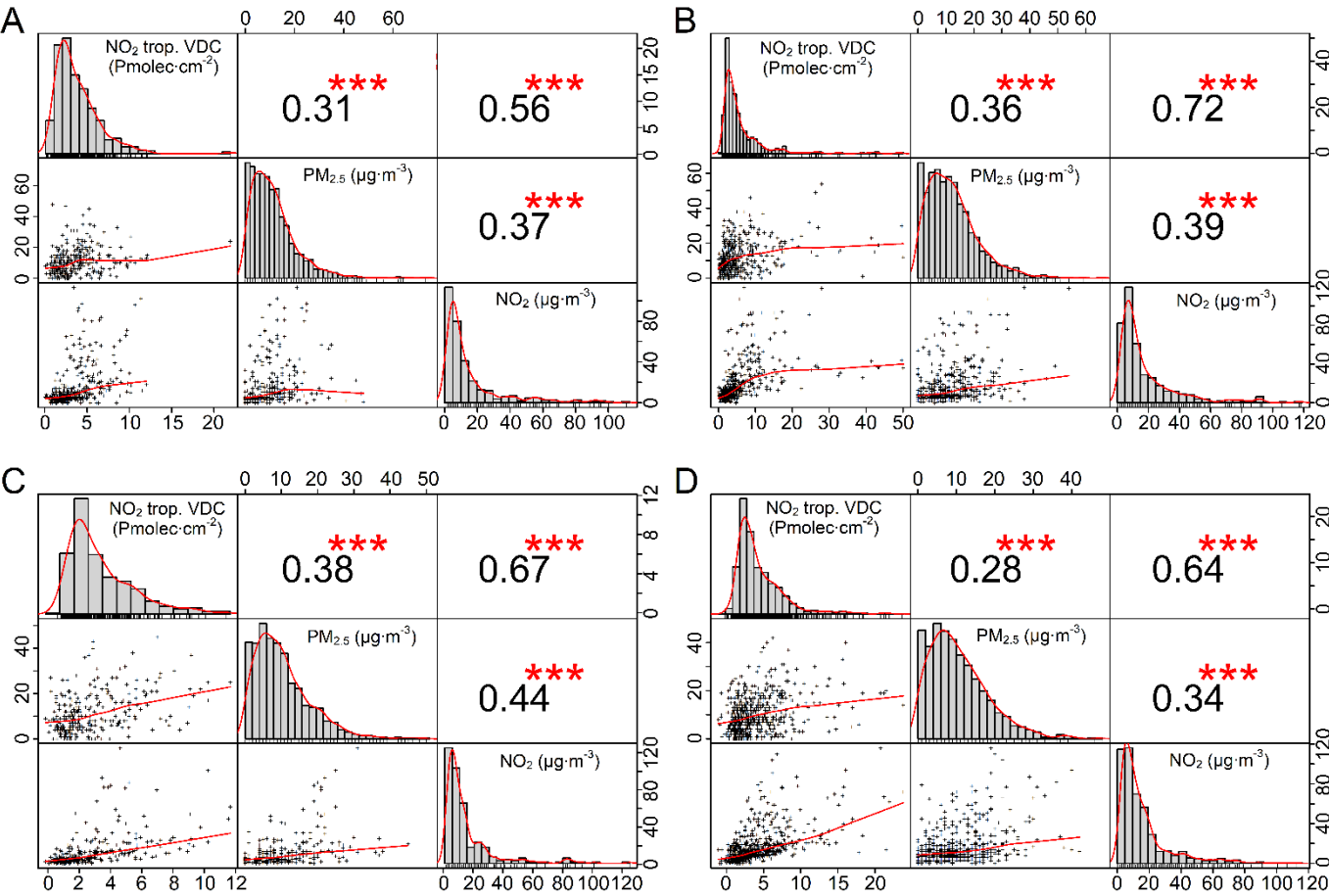


Figure C.8 - NO₂ tropospheric vertical column density averaged over 30 days: (A) before March 15, 2019; (B) after March 15, 2019; (C) before March 15, 2020; and (D) after March 15, 2020 (mobility restrictions).

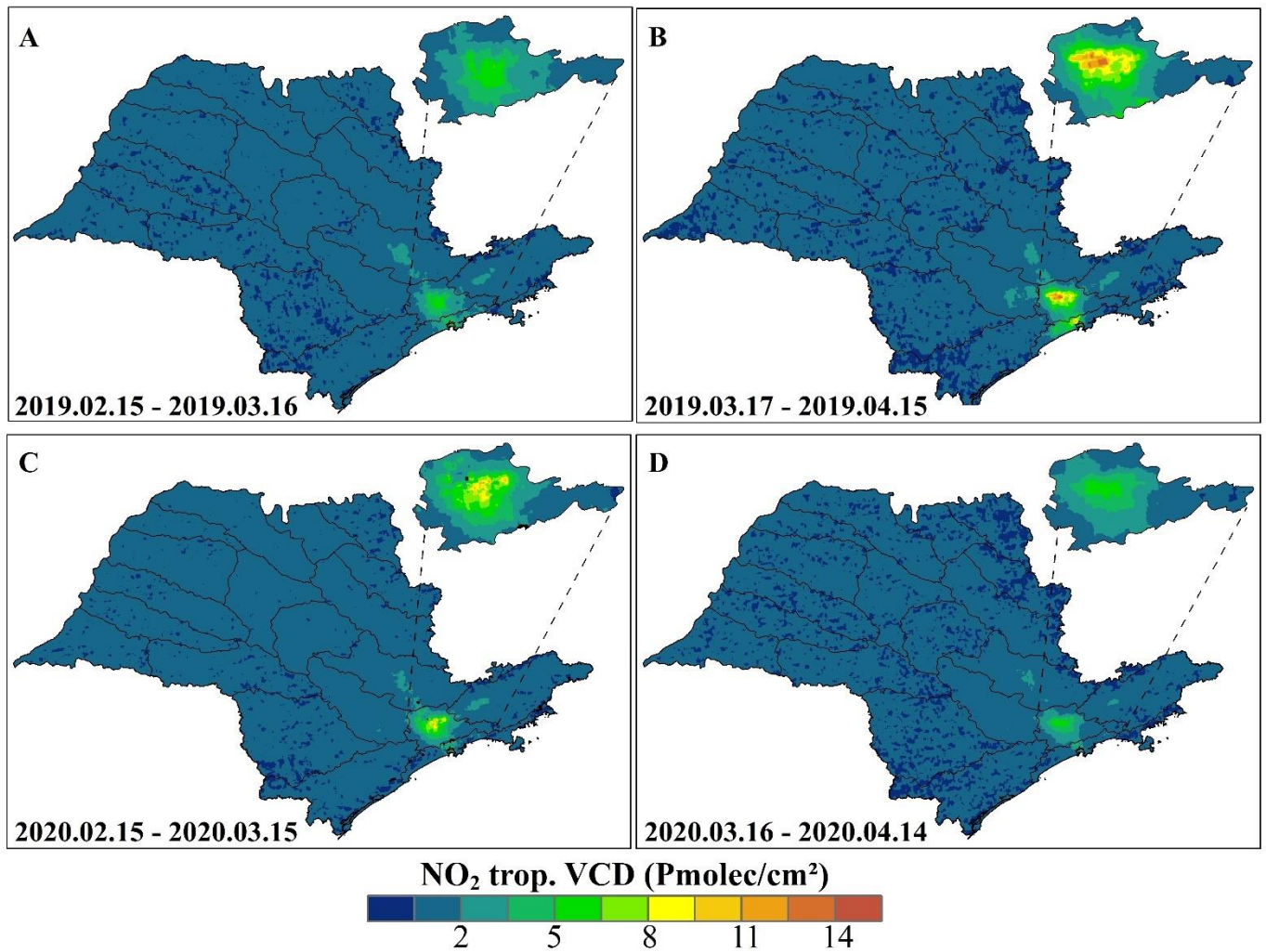


Figure C.9 - NO₂ tropospheric vertical column density data point sample sizes over 30 days: (A) before March 15, 2019; (B) after March 15, 2019; (C) before March 15, 2020; and (D) after March 15, 2020 (mobility restrictions).

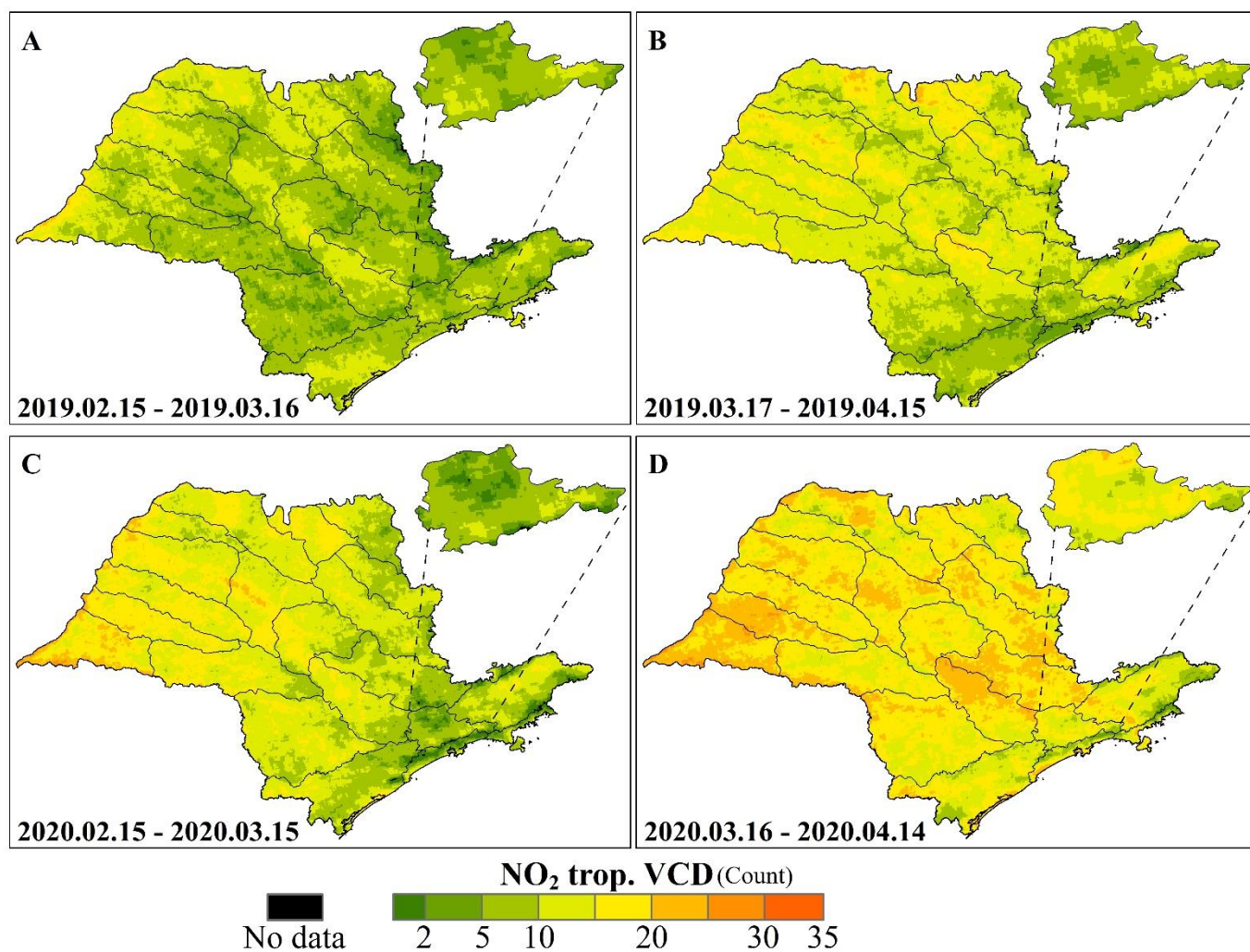


Figure C.10 - Cloudiness in the dry (left column) and rainy seasons (middle column) and Annual average (right column) for the State of São Paulo. The images were generated through interpolation (Inverse distance weighted - IDW) of the normal climatological data for Brazil provided by the National Institute of Meteorology (INMET, in portuguese Instituto Nacional de Meteorologia) (Available in: <https://portal.inmet.gov.br/normais>).

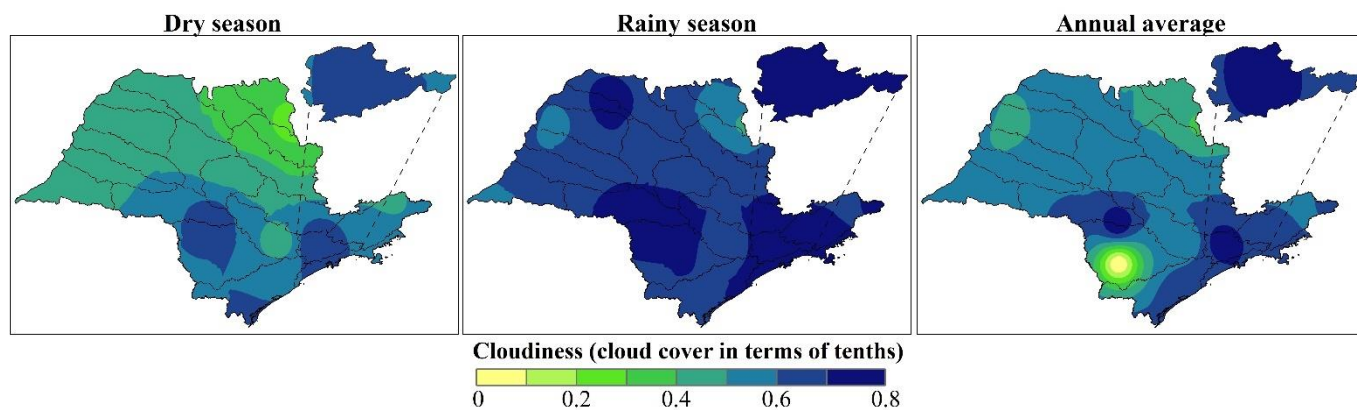
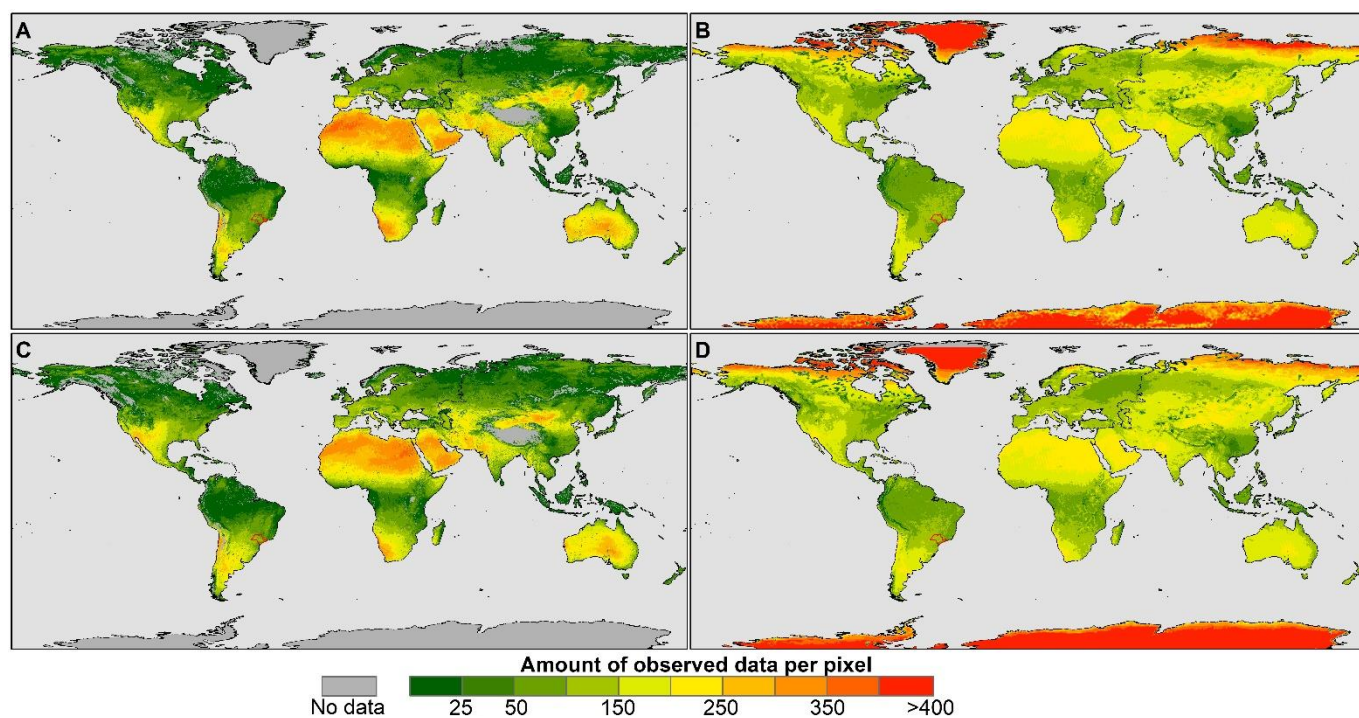


Figure C.11 - Global availability of AOD satellite data in the periods 12-01-2018 to 06-30-2019 (A) and 12-01-2019 to 06-30-2020; (C) and NO₂ tropospheric vertical column density in the periods of 12-01-2018 to 06-30-2019 (B) and 12-01-2019 to 06-30-2020 (D).

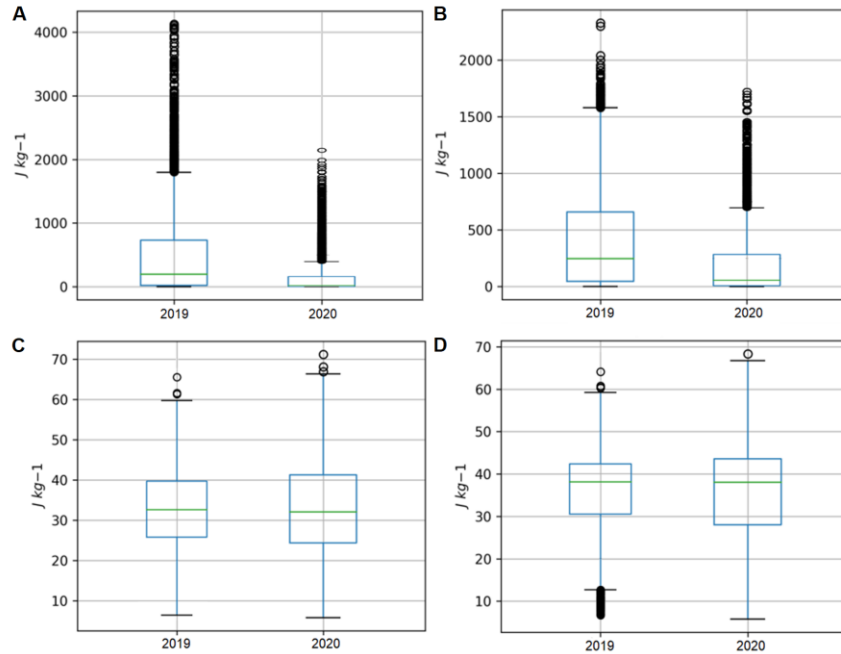


Section C1 - Atmospheric conditions and impact of fire emissions on measurements

Using ERA5 reanalysis hourly data, two important atmospheric quantities were analyzed to assess whether the overall and local atmospheric conditions for São Paulo (city and state) were significantly similar or different among the first six months of 2019 and 2020: Convective Available Potential Energy (CAPE) and Precipitable Water (PWater). Greater CAPE values imply greater energy availability for deep convection activity in that period, while greater values of PWater imply greater availability of moisture to feed that convective activity. When both values are simultaneously high, it means that the atmospheric conditions in the period are prone to mixing air masses, so that pollutants can be better dissipated. Moreover, both quantities are vertically integrated, so that they both represent a tropospheric condition without a clear distinction between the Planetary Boundary Layer and the free troposphere.

The box plots and the t-Student tests, in general, indicated that the energy available for deep convection was statistically different between January and June 2019 and 2020. This is valid for both the São Paulo city and state (see Figure C.12). However, São Paulo city and state show statistically similar conditions with respect to moisture for the first six months of the two years. These two variables indicate that different atmospheric conditions may have predominated in the region in the two periods analyzed in this study. However, they are diagnostic variables and additional elements must be analyzed to understand what happened in the atmosphere in both years. Therefore, we analyzed the synoptic conditions found in the two years.

Figure C.12 - Comparisons for surface data between Jan-Jun, 2019 and Jan-Jun, 2020 - Average areal surface CAPE hourly data for São Paulo city (A) and State (B); Surface-300 hPa mean areal Integrated Precipitable Water (PWater) hourly data for São Paulo city (C) and State (D). The blue square represents the interval between the first and third quartiles; the green line indicates the second quartile (median); the black circles indicate the outliers; and the black lines indicate the maximum (top line) and minimum (bottom line) values excluding outliers.

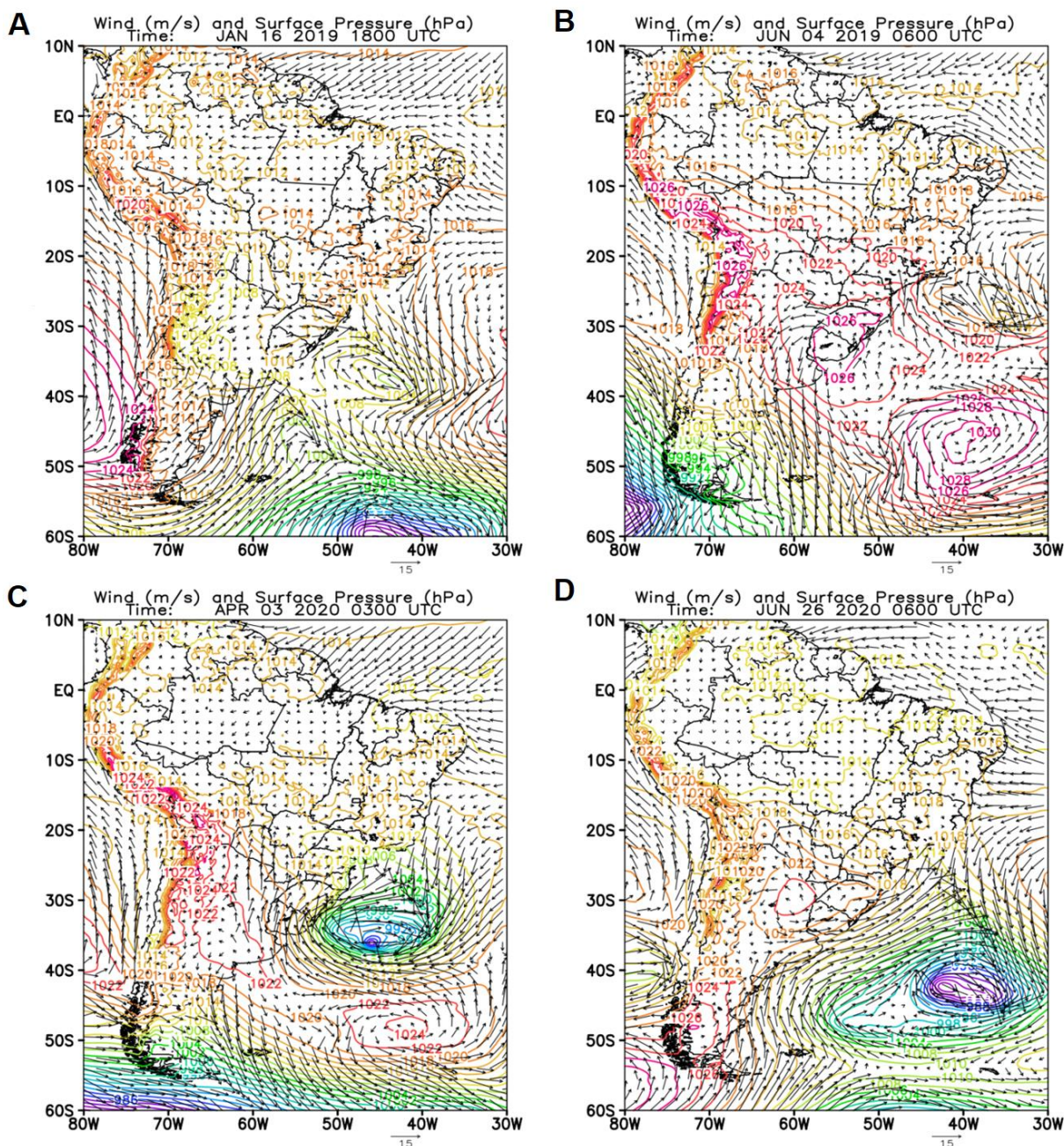


A visual estimate of the number of synoptic cyclones and associated cold fronts observed in the regions around the state of São Paulo, considering the specific period of January 01 to June 30 in the years of 2019 and 2020, was made through ERA5 reanalysis, in a temporal interval of 3 hours. Two types of systems were observed: i) Classical systems consisting of well-defined extratropical cyclones that, in general, develop at the coast of the continent in latitudes around Uruguay/Southern Brazil with a deepening central low pressure. The trajectory of these moving synoptic-scale low-pressure systems was frequently observed to be eastward/southeastward in the Atlantic Ocean, which is the climatologic behavior of these phenomena. Around surface low-pressure systems, low-level winds turn clockwise in the Southern Hemisphere and, therefore, bring cold air from the South in the continent, and warm air from the North in the ocean. As the cold air advances in the continent, cold fronts form when and where these different air masses meet, which happens largely in the continent as a function of the prevalent warm air mass in Brazil. The cold front then intensifies and displaces in direction to the North/Northeast in the continent, normally reaching São Paulo State after one to two days. Only a few of these systems were born over the continent, acquiring a cyclonic circulation before reaching the warm oceanic water; ii) Subtropical vortices that develop over the ocean along the

coast of São Paulo. These vortices are less deep than the extratropical ones. However, they produce a disturbance in the winds in the east region of those states that resembles cold fronts.

After the passage of the above-described systems through São Paulo state, the main circulation observed affecting the region is that associated with transient high-pressure systems which, depending on the position of its pressure center, produces northeast, east, or southeast dominant winds on the coast of São Paulo that, sometimes, can reach São Paulo city. In terms of the number of systems, our visual estimate indicates 24 synoptic cyclonic vortices and associated cold fronts affecting more the eastern part of the state of São Paulo than the interior, against 31 such systems passing in the same region in 2020. This is a large difference and shows the predominance of different atmospheric conditions for the two periods. In addition, it should be noted that 2020 presented 12 intense extratropical cyclones among the 31 events, while the same six-month period of 2019 showed only two intense vortices. These data suggest that the first half of 2020 was much more dynamic than the same period in 2019, at least along with the state of São Paulo's coast. Figure C.13 shows examples of 1000 hPa cyclonic vortices and associated cold fronts. Figure C.13a shows an extratropical cyclone born on the coast of Uruguay on January 16, 2019, with a relatively weak low-pressure center (about 998 hPa at its minimum). Figure C.13b shows a subtropical cyclone that developed during June 04, 2019, disturbing the atmosphere in São Paulo. Figure C.13c shows an extratropical cyclone developed at the coast of Uruguay, that reached a low center of 984 hPa when its associated cold front was crossing over São Paulo, being classified as an explosive extratropical cyclone (which occurs when the cyclonic surface pressure falls 24 hPa or more in 24 hours). Figure C.13d shows an intense extratropical cyclone, observed during May 26, 2020, whose low center showed 990 hPa.

Figure C.13 - (A) Extratropical cyclone born on the coast of Uruguay on January 16, 2019; (B) Subtropical cyclone affecting São Paulo on June 04, 2019; (C) Extratropical cyclone born on the coast of Uruguay on April 03, 2020, classified as an explosive extratropical vortex; (D) Intense extratropical cyclone, observed on May 26, 2020, whose low center showed 990 hPa.



To verify the impact of meteorological variables on air pollutant concentrations, the correlations between NO_2 , PM_{10} , and $\text{PM}_{2.5}$ measured at ground level and temperature, wind speed, precipitation, planetary boundary layer height (PBLH), and relative humidity (RH) were evaluated (Table C.9 – Correlation between meteorological variables and PM_{10} concentrations measured at ground level. Table C.9 to Table C.11). As expected, precipitation and relative humidity significantly impact atmospheric pollutant concentrations, especially particulate matter. This is due to wet deposition, which is considered an essential pathway for removing

several air pollutants (GUO et al., 2014; PAN et al., 2017; YOO et al., 2014). (YOO et al., 2014) indicate that scavenging of air pollutants is directly related to rain intensity and suggest that precipitation is especially associated with PM removal, followed by SO₂, NO₂, CO, and O₃.

PBLH also showed a negative correlation at many monitoring stations, once PBLH is an essential factor related to atmospheric diffusion, determining the vertical dispersal range of pollutants (SU; LI; KAHN, 2018; WANG et al., 2019). With greater PBLH, there is a greater dispersion of pollutants to higher layers, reducing the concentrations at the surface level. On the other hand, when PBL is low, the pollutants tend to have higher concentrations, as they are trapped closer to the surface. It is also possible to observe that several stations presented significant and negative correlations with horizontal speed of air moving towards the north. This is due to the fact that, to the south of HUWRMs 6 and 7 (where most of the stations are concentrated), the ocean is responsible for the transport of pollutants caused by the sea breeze coming from the southeast (CHIQUELLO et al., 2019). This is more evident when observing the Santos - Ponta da Praia station. The station is located near the coast of São Paulo and presented one of the highest negative correlations with V-wind for all pollutants. On the other hand, it is possible to observe that stations to the north of MASP (e.g., Pico do Jaraguá station) have a significant but positive correlation, as the local circulation coming from the south brings more pollution to those stations (BOIAN; ANDRADE, 2012). For temperature, the correlations were more significant for NO₂. In this case, negative correlations were observed, possibly related to the photochemical reactions that occur in the atmosphere and that favor the formation of other chemical elements, such as O₃.

Comparisons between the period studied for 2019 and 2020 do not show significant differences in the correlations of meteorological variables and pollutant concentrations measured at ground level. However, it is possible to observe that, in general, the 2020 correlations were more significant than those of 2019. This fact may be related to reducing very localized sources as a function of mobility restrictions, especially on high-traffic roads. The reduction in localized sources favored meteorological phenomena to have more impact on pollutant concentrations. For precipitation, it is possible to notice that, in 2020, the correlations were generally less significant and smaller when compared to 2019, as an increase in air pollutant concentrations is expected during the dry season, which was partially altered due to mobility restrictions. For 2020, more significant correlations are observed for the horizontal speed of air moving towards

the east. This fact may be related to the more substantial number of fire outbreaks observed in 2020 (Figure C.14), which originated in the northwest of the study area, possibly increasing concentrations of pollutants when air masses came from these directions, as mentioned by (RUDKE et al., 2021a; SQUIZZATO et al., 2021). The impact of biomass burning in the region is already well known and certainly had a more significant effect in 2020 due to the record number of fires observed for the year. In 2020, fire outbreaks were also observed in regions less favorable to fires, such as the Pantanal (known for its flooded plains) (DA SILVA JUNIOR et al., 2020).

Table C.9 – Correlation between meteorological variables and PM₁₀ concentrations measured at ground level.

Name	U-Wind		V-Wind		Temperature		Total precipitation		PBLH		RH	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Araçatuba	-0.15*	-0.34***	-0.07	-0.08	-0.2**	-0.26***	-0.39***	-0.43***	-0.01	-0.02	-0.66***	-0.7***
Araraquara	0.01	-0.17*	-0.09	-0.05	-0.09	-0.37***	-0.39***	-0.4***	0.07	-0.04	-0.58***	-0.63***
Bauru	0.12	0	-0.23***	-0.28***	-0.13	-0.26***	-0.34***	-0.35***	-0.12	-0.29***	-0.61***	-0.58***
Campinas-Centro	0.14	0.11	-0.21**	-0.15*	0	-0.19**	-0.34***	-0.17*	-0.01	-0.13	-0.5***	-0.39***
Capão Redondo	0.23***	0.38***	-0.5***	-0.56***	0.12	-0.02	-0.44***	-0.34***	-0.03	-0.15*	-0.6***	-0.7***
Carapicuíba	0.27***	0.33***	-0.46***	-0.45***	-0.03	-0.06	-0.37***	-0.26***	-0.17*	-0.3***	-0.54***	-0.65***
Catanduva	-0.15*	-0.26***	-0.12	-0.14*	-0.31***	-0.45***	-0.41***	-0.42***	-0.09	-0.05	-0.64***	-0.7***
Cerqueira César	0.15*	0.28***	-0.42***	-0.43***	0.28***	0.04	-0.39***	-0.29***	-0.08	-0.28***	-0.53***	-0.57***
Congonhas	0.07	0.3***	-0.25***	-0.37***	0.03	0	-0.43***	-0.21**	-0.11	-0.26***	-0.47***	-0.53***
Cubatão-Centro	0.21**	0.24***	-0.55***	-0.52***	0.2**	0.1	-0.39***	-0.35***	-0.14*	-0.05	-0.45***	-0.48***
Cubatão-V.Parisi	0.2**	0.16*	-0.42***	-0.37***	0.29***	0.12	-0.39***	-0.34***	0.13	0.16*	-0.57***	-0.48***
Diadema	0.16*	0.27***	-0.43***	-0.45***	0.17*	0.04	-0.42***	-0.3***	0	-0.07	-0.52***	-0.51***
Grajaú-Parelheiros	0.32***	0.5***	-0.62***	-0.66***	0.12	0.03	-0.43***	-0.32***	0.07	0.04	-0.66***	-0.75***
Guaratinguetá	-0.02	0.11	-0.34***	-0.22**	-0.12	-0.33***	-0.34***	-0.37***	0.06	-0.11	-0.53***	-0.48***
Guarulhos-Paço Municipal	0.06	0.24***	-0.31***	-0.34***	-0.03	-0.1	-0.4***	-0.33***	-0.15*	-0.24***	-0.5***	-0.64***
Interlagos	0.22**	0.31***	-0.56***	-0.48***	0.08	0.04	-0.47***	-0.32***	-0.13	-0.08	-0.48***	-0.56***
Jacareí	0.16	0.28***	-0.52***	-0.45***	0.15	-0.03	-0.41***	-0.31***	-0.04	-0.09	-0.64***	-0.5***
Jau	0.07	0.01	-0.15*	-0.15*	-0.1	-0.28***	-0.37***	-0.34***	-0.04	-0.24***	-0.59***	-0.58***
Jundiaí	0.28***	0.25***	-0.41***	-0.35***	0.07	-0.16*	-0.36***	-0.32***	-0.08	-0.26***	-0.49***	-0.58***
Limeira	0.07	0	-0.05	-0.2**	-0.16*	-0.35***	-0.32***	-0.34***	-0.26***	-0.25***	-0.19*	-0.52***
Marg.Tietê-Pte Remédios	0.27***	0.43***	-0.47***	-0.55***	0.12	-0.02	-0.33***	-0.28***	-0.14*	-0.27***	-0.51***	-0.67***
Marília	0.03	-0.08	-0.18**	-0.32***	-0.06	-0.08	-0.31***	-0.42***	-0.02	-0.02	-0.62***	-0.62***
N.Senhora do Ó	0.11	0.23**	-0.31***	-0.38***	0.12	-0.07	-0.41***	-0.33***	-0.05	-0.28***	-0.53***	-0.62***
Osasco	0.17*	0.33***	-0.43***	-0.52***	0.03	0.02	-0.32***	-0.26***	-0.19**	-0.32***	-0.5***	-0.59***
Paulínia	0.07	0.12	-0.14*	-0.27***	-0.04	-0.33***	-0.38***	-0.29***	0.03	-0.27***	-0.51***	-0.43***
Piracicaba	0.05	-0.29	-0.18**	0.06	0	0.56**	-0.39***	-0.51**	0.02	0.5**	-0.46***	-0.59***
Presidente Prudente	-0.09	-0.18**	-0.09	-0.21**	0.1	-0.03	-0.43***	-0.41***	0.15*	0.14*	-0.73***	-0.7***
S.André-Capuava	0.07	0.25***	-0.32***	-0.38***	0.11	0.01	-0.43***	-0.31***	-0.17*	-0.22**	-0.45***	-0.52***
S.Bernardo-Paulicéia	0.1	0.25***	-0.4***	-0.44***	0.02	0.09	-0.45***	-0.32***	-0.13	-0.12	-0.47***	-0.51***
S.José Campos	0.09	0.25***	-0.41***	-0.46***	0.02	-0.15*	-0.4***	-0.35***	-0.12	-0.2**	-0.5***	-0.56***
S.José Campos-Jd.Satelite	0.1	0.25***	-0.36***	-0.34***	-0.05	-0.23***	-0.42***	-0.37***	-0.14	-0.23***	-0.51***	-0.5***
Santa Gertrudes	-0.12	-0.2**	-0.16*	-0.04	-0.23***	-0.37***	-0.39***	-0.41***	-0.09	-0.24***	-0.47***	-0.54***
Santos-Ponta da Praia	0.01	0.15*	-0.63***	-0.63***	0.13	-0.05	-0.44***	-0.31***	-0.6***	-0.51***	-0.09	-0.26***
São Caetano do Sul	0.23**	0.22**	-0.45***	-0.37***	0.1	0.03	-0.4***	-0.28***	-0.19*	-0.21**	-0.41***	-0.42***
São José do Rio Preto	-0.3***	-0.34***	-0.02	-0.06	-0.26***	-0.35***	-0.4***	-0.48***	-0.06	0.16*	-0.7***	-0.76***
Sorocaba	0.36***	0.34***	-0.54***	-0.51***	0.19*	0.02	-0.25***	-0.24***	0.02	-0.21**	-0.58***	-0.64***
Taboão da Serra	0.25***	0.38***	-0.49***	-0.54***	-0.02	0.1	-0.39***	-0.29***	-0.17*	-0.14*	-0.48***	-0.65***
Tatui	0.25***	0.32***	-0.5***	-0.63***	0.06	0	-0.28***	-0.21**	-0.12	-0.32***	-0.51***	-0.52***
Taubaté	0.13	0.22**	-0.41***	-0.35***	-0.01	-0.27***	-0.39***	-0.37***	0.03	-0.11	-0.56***	-0.52***

Note: U-Wind - horizontal speed of air moving towards the east; V-Wind - horizontal speed of air moving towards the north; PBLH - planetary boundary layer height; RH – relative humidity. Significance level: * - p-value<0.05; ** - p-value<0.01; *** - p-value<0.001.

Table C.10 – Correlation between meteorological variables and PM_{2.5} concentrations measured at ground level.

Name	U-Wind		V-Wind		Temperature		Total precipitation		PBLH		RH	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Campinas-V.União	0.15*	0.22**	-0.23***	-0.24**	-0.25***	-0.02	-0.35***	-0.28***	-0.23***	-0.01	-0.34***	-0.49***
Congonhas	0.06	0.28***	-0.25***	-0.35***	0.1	0.05	-0.39***	-0.17*	-0.18**	-0.36***	-0.4***	-0.45***
Grajaú-Parelheiros	0.24**	0.41***	-0.56***	-0.57***	0.15*	0.06	-0.41***	-0.29***	0.04	-0.07	-0.61***	-0.63***
Guarulhos-Paço Municipal	0.06	0.24***	-0.27***	-0.3***	0.03	-0.04	-0.36***	-0.26***	-0.23***	-0.32***	-0.41***	-0.49***
Ibirapuera	0.08	0.24***	-0.32***	-0.36***	0.12	0.13	-0.43***	-0.25***	-0.16*	-0.23***	-0.44***	-0.47***
Itaim Paulista	0.19*	0.35***	-0.33***	-0.39***	-0.01	-0.14*	-0.33***	-0.25***	-0.14	-0.3***	-0.46***	-0.57***
Marg,Tietê-Pte Remédios	0.23***	0.46***	-0.43***	-0.58***	0	0.1	-0.31***	-0.21**	-0.19**	-0.33***	-0.47***	-0.6***
Mooca	0.12	0.23***	-0.34***	-0.33***	-0.02	0.06	-0.42***	-0.22**	-0.21**	-0.35***	-0.48***	-0.5***
Osasco	0.1	0.28***	-0.34***	-0.45***	-0.03	-0.02	-0.36***	-0.26***	-0.19**	-0.35***	-0.43***	-0.59***
Parque D.Pedro II	0.11	0.22**	-0.32***	-0.35***	0.17*	0.07	-0.32***	-0.25***	-0.28***	-0.32***	-0.38***	-0.51***
Pico do Jaraguá	0.01	0.12	0.01	-0.12	0.27***	0.37***	-0.18*	0.09	0.22**	-0.03	-0.26***	-0.08
Piracicaba	0.04	0	-0.19**	-0.22***	0.04	-0.25***	-0.39***	-0.32***	0.01	-0.22**	-0.5***	-0.51***
S.Bernardo-Centro	0.09	0.22**	-0.45***	-0.4***	0.34***	0.21**	-0.42***	-0.24***	-0.14	-0.18**	-0.4***	-0.37***
S.José Campos-Jd.Satelite	0.1	0.27***	-0.36***	-0.37***	-0.02	-0.25***	-0.44***	-0.3***	-0.15*	-0.31***	-0.51***	-0.45***
Santa Gertrudes	-0.1	-0.19*	-0.07	0.04	-0.06	-0.12	-0.33***	-0.26***	-0.06	-0.27***	-0.41***	-0.39***
Santana	-0.03	0.11	-0.18*	-0.24***	0	-0.01	-0.38***	-0.26***	-0.21**	-0.29***	-0.32***	-0.46***
Santos-Ponta da Praia	0.05	0.15*	-0.63***	-0.56***	0.01	0.14*	-0.47***	-0.29***	-0.65***	-0.65***	-0.01	-0.05
São Caetano do Sul	0.29***	0.19**	-0.45***	-0.34***	0.15*	0.04	-0.35***	-0.24***	-0.23**	-0.31***	-0.33***	-0.33***
São José do Rio Preto	-0.29***	-0.32***	0.06	0.06	-0.21**	-0.36***	-0.43***	-0.47***	-0.05	0.04	-0.71***	-0.74***
Taubaté	0.14*	0.24***	-0.39***	-0.33***	0.04	-0.26***	-0.36***	-0.31***	0.01	-0.18**	-0.51***	-0.45***

Note: U-Wind - horizontal speed of air moving towards the east; V-Wind - horizontal speed of air moving towards the north; PBLH - planetary boundary layer height; RH – relative humidity. Significance level: * - p-value<0.05; ** - p-value<0.01; *** - p-value<0.001.

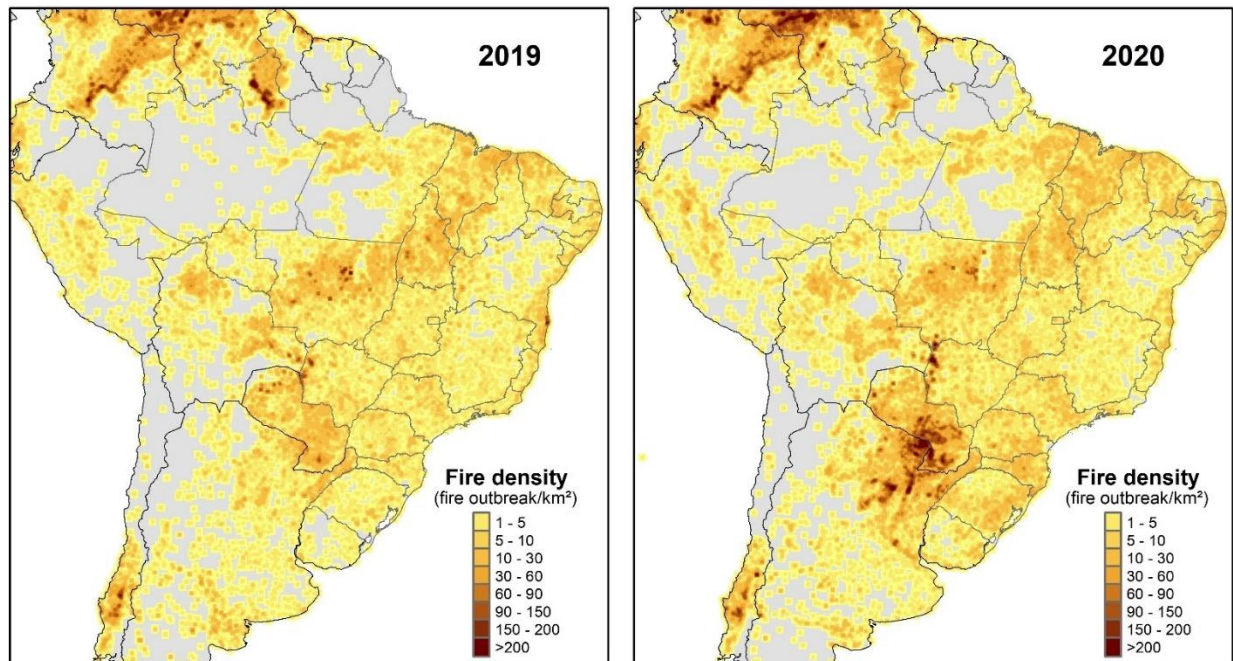
Table C.11 – Correlation between meteorological variables and NO₂ concentrations measured at ground level.

Name	U-Wind		V-Wind		Temperature		Total precipitation		PBLH		RH	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Araraquara	-0.09	-0.13	-0.05	-0.08	-0.32***	-0.44***	-0.32***	-0.28***	-0.19**	-0.33***	-0.4***	-0.46***
Bauru	0.03	-0.02	-0.09	-0.26***	-0.32***	-0.44***	-0.29***	-0.32***	-0.23***	-0.43***	-0.5***	-0.46***
Campinas-Taquaral	0.26***	0.34***	-0.31***	-0.34***	-0.37***	-0.33***	-0.23***	-0.07	-0.35***	-0.53***	-0.25***	-0.21**
Catanduva	-0.22**	-0.24***	-0.05	0	-0.4***	-0.52***	-0.41***	-0.38***	-0.23**	-0.19**	-0.61***	-0.63***
Cerqueira César	0.33***	0.38***	-0.56***	-0.53***	0.19**	0.08	-0.2**	-0.05	-0.18**	-0.44***	-0.47***	-0.36***
Congonhas	0.11	0.19**	-0.14*	-0.15*	0.13	0.06	-0.33***	-0.03	-0.08	-0.4***	-0.43***	-0.3***
Cubatão-Centro	0.2**	0.25***	-0.45***	-0.31***	-0.25***	-0.11	-0.22**	-0.1	-0.33***	-0.34***	-0.27***	-0.21**
Cubatão-V.Parisi	0.23***	0.21**	-0.41***	-0.29***	-0.16*	-0.29***	-0.13	-0.14*	-0.2**	-0.28***	-0.24***	-0.16*
Guaratinguetá	0.05	0.17*	-0.37***	-0.52***	-0.41***	-0.31***	-0.29***	-0.22**	-0.12	-0.32***	-0.28***	-0.32***
Guarulhos-Pimentas	0.32***	0.4***	-0.31***	-0.38***	0.01	-0.09	-0.11	-0.01	-0.27***	-0.44***	-0.27***	-0.4***
Ibirapuera	0.19**	0.34***	-0.38***	-0.44***	-0.07	0.08	-0.25***	-0.07	-0.28***	-0.4***	-0.36***	-0.44***
Itaim Paulista	0.46***	0.57***	-0.54***	-0.57***	-0.06	-0.07	-0.08	0	-0.26***	-0.35***	-0.33***	-0.49***
Jau	0.29***	0.29***	-0.34***	-0.45***	-0.02	-0.33***	-0.23***	-0.17*	-0.12	-0.54***	-0.54***	-0.28***
Marg,Tietê-Pte Remédios	0.38***	0.54***	-0.6***	-0.67***	0.22**	0.16*	-0.24***	-0.03	-0.1	-0.38***	-0.51***	-0.45***
Marília	0.08	0.17*	-0.12	-0.45***	-0.25***	-0.29***	-0.22**	-0.14*	-0.23***	-0.41***	-0.5***	-0.27***
Osasco	0.05	0.09	-0.31***	-0.34***	0.11	0.06	-0.3***	-0.08	-0.09	-0.46***	-0.44***	-0.32***
Parque D.Pedro II	0.26***	0.39***	-0.39***	-0.47***	0.12	0.06	-0.2**	0	-0.22**	-0.5***	-0.39***	-0.39***
Pico do Jaraguá	-0.15	-0.37***	0.17*	0.33***	-0.08	-0.23**	0.03	0.01	-0.11	-0.36***	0.04	0.26***
Pinheiros	0.32***	0.39***	-0.46***	-0.43***	0.07	0.25***	-0.22**	0.14*	-0.22**	-0.45***	-0.44***	-0.21**
Piracicaba	-0.07	-0.03	-0.06	-0.19*	-0.29***	-0.3***	-0.31***	-0.15	-0.13	-0.4***	-0.33***	-0.15*
Presidente Prudente	0.07	0.17*	0.08	-0.1	-0.27***	-0.25***	-0.25***	-0.24***	-0.31***	-0.27***	-0.34***	-0.35***
S.André-Capuava	0.26**	0.37***	-0.5***	-0.53***	-0.1	-0.03	-0.3***	-0.12	-0.31***	-0.28***	-0.38***	-0.48***
S.Bernardo-Centro	0.44***	0.5***	-0.66***	-0.6***	0.12	0.1	-0.27***	-0.08	-0.1	-0.15*	-0.45***	-0.48***
S.José Campos	0.14*	0.28***	-0.29***	-0.34***	-0.39***	-0.36***	-0.13	-0.05	-0.39***	-0.47***	-0.12	-0.24***
S.José Campos-Jd.Satelite	0.2**	0.43***	-0.47***	-0.53***	-0.31***	0.02	-0.13	0.09	-0.42***	-0.3***	-0.27***	-0.21*
Santa Gertrudes	-0.07	-0.18**	0.02	0.09	-0.16*	-0.51***	-0.44***	-0.36***	0	-0.35***	-0.47***	-0.38***
Santos-Ponta da Praia	-0.12	0.09	-0.68***	-0.63***	-0.15*	0.03	-0.33***	-0.25***	-0.52***	-0.57***	-0.14	-0.16*
São Caetano do Sul	0.39***	0.47***	-0.56***	-0.55***	0.14	0.17*	-0.21**	-0.04	-0.15*	-0.26***	-0.52***	-0.46***
São José do Rio Preto	-0.39***	-0.36***	0.13	0.07	-0.39***	-0.43***	-0.39***	-0.41***	-0.15*	-0.02	-0.68***	-0.72***
Sorocaba	0.38***	0.45***	-0.57***	-0.65***	-0.18*	-0.08	-0.19*	-0.02	-0.4***	-0.6***	-0.27***	-0.34***
Tatui	0.31***	0.37***	-0.62***	-0.75***	-0.1	-0.02	-0.15*	-0.02	-0.3***	-0.5***	-0.29***	-0.24***
Taubaté	0.24**	0.42***	-0.38***	-0.45***	-0.47***	-0.32***	-0.1	-0.02	-0.37***	-0.42***	-0.15*	-0.29***

Note: U-Wind - horizontal speed of air moving towards the east; V-Wind - horizontal speed of air moving towards the north; PBLH - planetary boundary layer height; RH – relative humidity. Significance level: * - p-value<0.05; ** - p-value<0.01; *** - p-value<0.001.

As mentioned, biomass burning significantly impacts air pollutant emissions in Brazil. Figure C.1 shows the density of fire outbreaks observed in South America during the first semester of 2019 and 2020. It is observed that the fire outbreaks have the same behavior for both years, but with a more expressive density mapped for 2020. In both years, it was observed that fire outbreaks are concentrated in Paraguay, Venezuela, and Colombia, in the Brazilian state of Roraima, and in the states on the axis between Maranhão and Mato Grosso do Sul.

Figure C.14 - Spatial distribution of the fire outbreaks density observed for the studied period in 2019 and 2020.

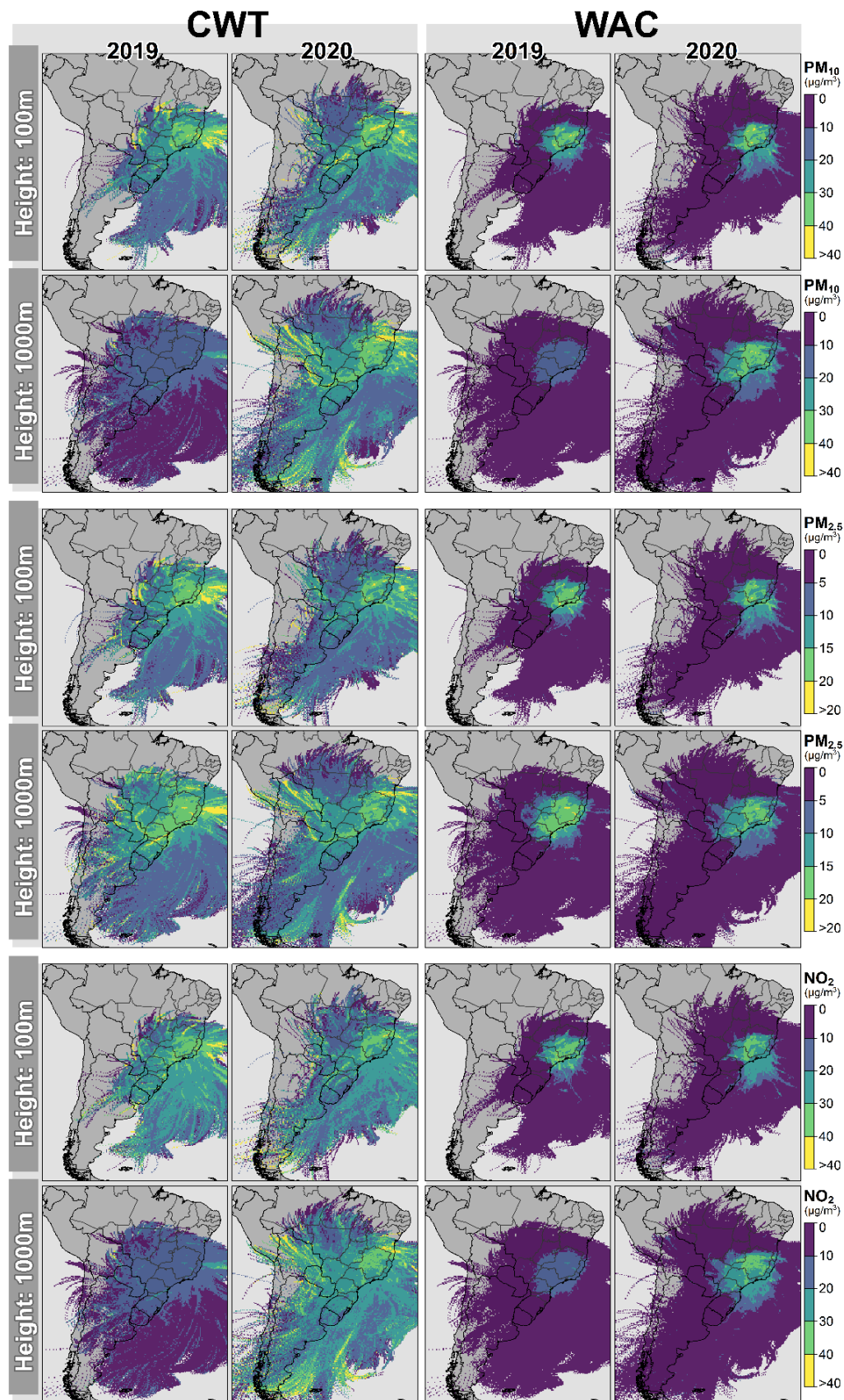


Due to the importance of fire emission in the air quality of the study region, the HYSPLIT model (STEIN et al., 2015) of the National Oceanic and Atmospheric Administration (NOAA) was used to assess the origin of pollutants and long-range transport. In this sense, 3-day backward atmospheric trajectories were used to verify wildfire smoke originating in the north and central region of South America. Seeking to evaluate the impacts on the surface observed by the air quality stations, trajectories of air masses reaching São Paulo at 100 m and 1000m Above Ground Level (AGL) were considered. The trajectories obtained through HYSPLIT were used together with the Concentration Weighted Trajectory (CWT) algorithm (BRERETON; JOHNSON, 2012; HSU; HOLSEN; HOPKE, 2003) to locate significant sources of pollutants degrading air quality in São Paulo. The Weighted Average Concentration (WAC) (BRERETON; JOHNSON, 2012; DIMITRIOU et al., 2015) was also used to locate the sources of pollutants that had the most impact for the period. In these stages, the location of the monitoring stations was considered the place of arrival of the air masses. The equations and more information about CWT and WAC can be obtained from (BRERETON; JOHNSON, 2012; DIMITRIOU et al., 2015).

The CWT algorithm was used to evaluate the main sources of the analyzed pollutants (PM_{10} , $PM_{2.5}$, and NO_2) and the effect of biomass burning on their concentrations. The results of the model that uses air masses trajectories at 100 m and 1000m height from Hysplit are shown in Figure C.15. It is observed that regions with high biomass burning emissions (Figure C.14)

impacted the concentrations of pollutants analyzed for São Paulo. High air pollutant concentrations related to biomass burning are mainly attributed to northwest and west airflows through the Pantanal biome, besides Bolivia and Paraguay. The high number of fire outbreaks in the Pantanal was reported worldwide due to the unprecedented devastation of the Biome (DA SILVA JUNIOR et al., 2020; FERREIRA BARBOSA et al., 2022; MARQUES et al., 2021). In addition, fires in this region were associated with deteriorating air quality in several Brazilian states, such as São Paulo, Rio de Janeiro, and Paraná (BERINGUI et al., 2022; LIBONATI et al., 2020; RUDKE et al., 2021a). These findings corroborate previous studies that demonstrate the impacts of long-range transport of pollutants emitted from burning biomass areas over South America, especially those originating in the Amazon and central-west region of Brazil, and their effects on southern and southeastern regional air quality (MARTINS et al., 2018; PEREIRA et al., 2017a; TARGINO et al., 2019; VARA-VELA et al., 2018). For example, (MARTINS et al., 2018; PEREIRA et al., 2017a; TARGINO et al., 2019; VARA-VELA et al., 2018) show that fires have significant impacts in São Paulo, doubling the concentrations of PM₁₀, PM_{2.5}, and NO_x, compared to the period without fire episodes in 2015.

Figure C.15 - Concentration Weighted Trajectory (CWT) and Weighted Average Concentrations (WAC) of PM₁₀ (top row), PM_{2.5} (middle row), and NO₂ (bottom row) at 100 m and 1000 m for the analyzed period of 2019 and 2020.

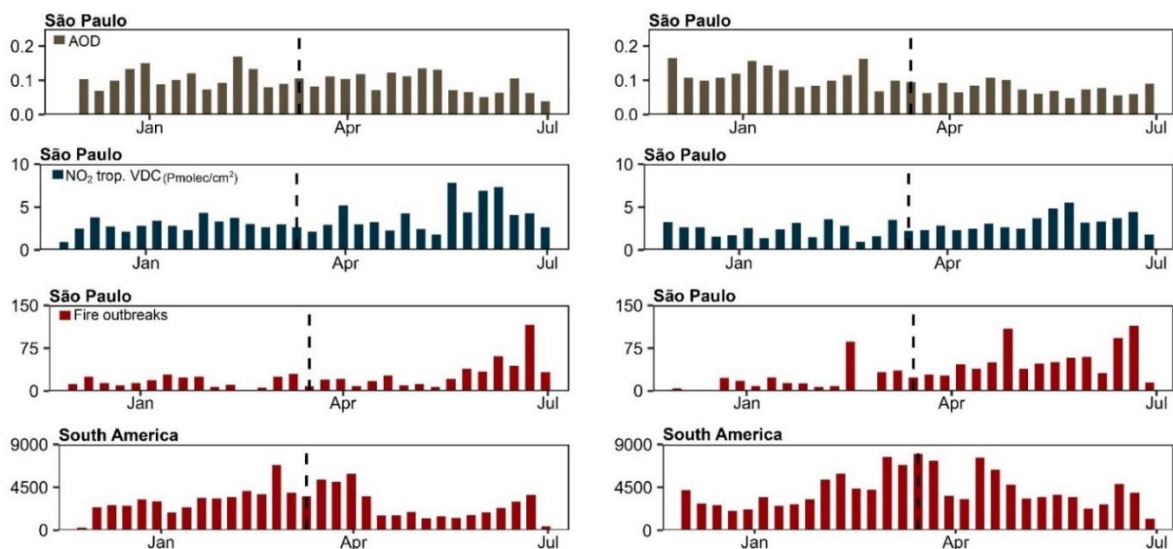


Although the impact of the fires is evident, when evaluating the primary sources of pollutants for the period, WAC (CWT weighted by the number of trajectories in each cell of the grid), it is noticed that local sources have more impact on the concentration (including biomass burning in proximate areas) than fire outbreaks in remote areas. Analyzing WAC (Figure C.15), it is observed that the inflow of air masses traveling over Minas Gerais, Rio de Janeiro, and São Paulo are primarily responsible for the transport of pollutants to the receiver site, which is added on top of those locally emitted (PEDRUZZI, 2020).

Section C2 - Impact of fire emissions on satellite measurements

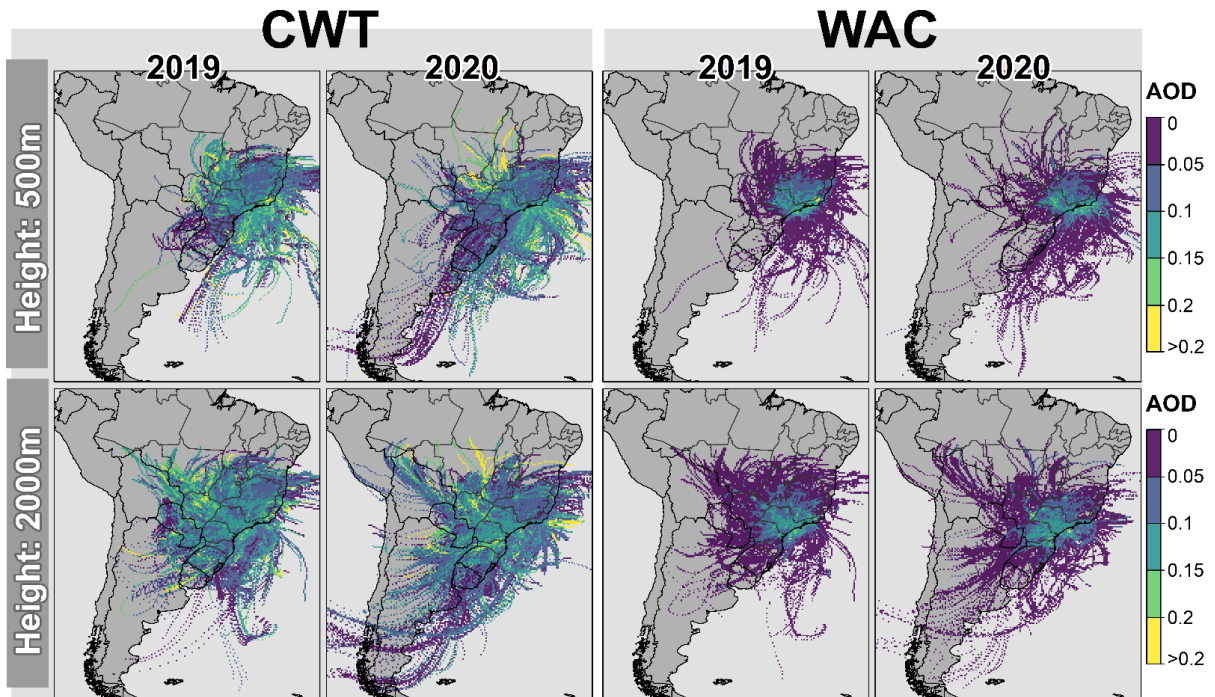
As previously demonstrated, biomass burning emissions in northern and central South America significantly impact ground-level air pollutant concentrations in São Paulo. Thus, to verify the impact of fires on satellite measurements, Figure C.16 presents the weekly profile of AOD and tropospheric NO₂ measures concerning the number of fire outbreaks observed in São Paulo and South America. It is observed that tropospheric NO₂ data has a similar response to fire data for the state of São Paulo, with more significant numbers of outbreaks occurring after March 16 (beginning of mobility restrictions in 2020 and reference period for 2019), which is a drier period in the region. For AOD, a correlation trend is not clear, with the highest values recorded in the rainy season (before March 16, Figure 7.3). As NO₂ is a reactive chemical species, its residence time in the atmosphere is shorter than other pollutants, so the impact of more localized fires (São Paulo state, for example) may be more evident than biomass burning emissions far away. NO_x and VOC emitted during biomass burning events provide the formation of O₃ downwind of fire emissions (BREY; FISCHER, 2016; SQUIZZATO et al., 2021; WENTWORTH et al., 2018). During the day, O₃ is formed approximately one or two hours after the emission of its precursors (CHIQUELTO et al., 2019), driving higher concentrations of this secondary pollutant over longer distances rather than NO_x. On the other hand, the long-distance transport of aerosols is well known (MARTINS et al., 2018; MORAN-ZULOAGA et al., 2018; PEREIRA et al., 2017a; VARA-VELA et al., 2018). Thus, the impact of fires that occur in locations other than the state of São Paulo may have more effect on AOD than on tropospheric NO₂.

Figure C.16 - Weekly profile of measurements of AOD (top row) and tropospheric NO₂ (second row) and number of fire outbreaks for São Paulo and South America (two lower rows). The vertical dashed black line indicates the 76th day of 2019 and 2020, when the first restrictions started in 2020.



To verify the effect of biomass burning emissions on AOD measurements, CWT and WAT were also calculated for AOD (Figure C.17). For the Satellite data, heights of 500 m and 2000 m AGL were used, altitudes where the impact of fires is still quite evident (GONZALEZ-ALONSO; VAL MARTIN; KAHN, 2019; MARTINS et al., 2009). Through Figure C.17, it can be observed, in general, that high values of AOD are obtained when airflows cross regions impacted by biomass burning (see Figure C.14). This effect is observed for heights closer to the surface (500 m) and higher altitudes (2000 m). For WAC, the highest values observed for airflows coming from Minas Gerais and Rio de Janeiro at the height of 500 m are similar to those observed for PM concentrations (see Figure C.15 for WAC at 100 m – ground level). However, for heights of 2000 m, high values are also observed for airflows coming from the state of Paraná, which are not so evident for concentrations measured at ground level. As for the measurements at ground level, it is observed that more local emissions have more substantial effects on the analyzed period than places where the highest number of fires were observed (e.g., Pantanal). Studies carried out in the region with the most intense density of fire outbreaks (Figure C.14) show that more than 85% of the smoke emitted by biomass burning is located at altitudes below 2000 m (GONZALEZ-ALONSO; VAL MARTIN; KAHN, 2019). In addition, there is the dominance of injection close to the surface; altitudes lower than 500 m correspond to more than 50% of the heights observed by (GONZALEZ-ALONSO; VAL MARTIN; KAHN, 2019). According to (VAL MARTIN; KAHN; TOSCA, 2018), most emissions related to biomass burning in Brazil (between 75 and 100%) occur at altitudes below the PBLH. As it occurs below PBLH, it is expected that variations in the measurements captured by the satellite will also be observed in the air quality monitoring stations, even if to a lesser extent. Thus, although fires substantially affect satellite measurements, it is noteworthy that similar air quality deterioration patterns are expected at the surface (see Section C1 - Atmospheric conditions and impact of fire emissions on measurements).

Figure C.17 - AOD Concentration Weighted Trajectory (CWT) and Weighted Average Concentrations (WAC) for 500 m (top row) and 2000 m (bottom row) height considering the analyzed period of 2019 and 2020.



Although, in many cases, fire emissions can similarly affect surface PM and satellite-measured AOD, studies show that some fire events emit aerosols in layers above the PBLH and thus have different effects on PM and AOD. (DE ARRUDA MOREIRA et al., 2021) studied fire events and their effects on PM_{2.5} levels over MASP. They indicated that aerosol plume decoupled from the PBLH does not affect the lower regions of the atmosphere. Thus, the impacts of fires are not evident in measurements performed by air quality stations. During the fire emission event analyzed, the authors observed that, although the aerosol layer and the PBL were disconnected, the influence of the decoupled layer gradually affected the PBL region. Therefore, a significant increase in PM_{2.5} concentration was observed during the night or early in the following morning, causing the levels of PM_{2.5} in all air quality stations evaluated on the day after fire emissions to increase.

**APPENDIX D – EVALUATING TROPOMI AND MODIS PERFORMANCE TO
CAPTURE THE DYNAMIC OF AIR POLLUTION IN SÃO PAULO STATE: A CASE
STUDY DURING THE COVID-19 OUTBREAK**

Table D.1 - Summary of the statistical analysis for the MAIAC, DT (3 and 10 km), and DB at each AERONET sites. For each station, the first row represents the correlation, the second row indicates number of matches/within EE, and third row indicate RMSE/Bias.

AERONET sites	MAIAC		DT (3km)		DT (10km)		DB	
	Terra	Aqua	Terra	Aqua	Terra	Aqua	Terra	Aqua
Abracos_Hill	0.97	0.96	0.97	0.9	0.97	0.94	0.98	0.98
	431 / 84.5	154 / 70.1	346 / 64.7	110 / 38.2	252 / 76.2	47 / 59.6	238 / 93.7	39 / 89.7
	0.111 / -0.027	0.129 / 0.002	0.283 / 0.12	0.533 / 0.231	0.238 / 0.09	0.352 / 0.12	0.083 / 0.005	0.121 / -0.047
Alta_Floresta	0.97	0.96	0.97	0.95	0.97	0.96	0.98	0.97
	3079 / 87.4	2126 / 71.3	1469 / 65.6	1205 / 53	1313 / 66.3	552 / 54	990 / 88.7	479 / 91
	0.122 / -0.027	0.157 / -0.001	0.198 / 0.043	0.218 / 0.048	0.154 / 0.011	0.194 / -0.001	0.098 / -0.003	0.126 / -0.03
Amazon_ATTO_Tower	0.87	0.76	0.85	0.88	0.94	0.95	0.92	0.96
	238 / 79.8	233 / 73	175 / 46.3	203 / 13.3	137 / 65	55 / 38.2	52 / 75	19 / 47.4
	0.06 / -0.014	0.064 / 0.038	0.128 / 0.089	0.155 / 0.135	0.08 / 0.051	0.139 / 0.108	0.067 / 0.033	0.081 / 0.063
ARM_Manacapuru	0.76	0.86	0.95	0.96	0.96	0.96	0.72	0.93
	83 / 77.1	103 / 73.8	81 / 61.7	79 / 51.9	54 / 77.8	30 / 56.7	5 / 100	12 / 83.3
	0.123 / -0.054	0.103 / 0.002	0.131 / 0.078	0.121 / 0.087	0.064 / 0.031	0.082 / 0.058	0.035 / 0.007	0.082 / 0.03
Balbina	0.76	0.74	0.79	0.77	0.93	0.87	-	-
	149 / 81.9	16 / 75	158 / 25.3	18 / 27.8	59 / 66.1	5 / 100	-	-
	0.06 / -0.014	0.062 / 0.015	0.156 / 0.13	0.173 / 0.141	0.083 / 0.061	0.06 / 0.037	0.89	0.98
Belterra	0.6	0.84	0.9	0.9	0.9	0.98	0.89	0.98
	101 / 60.4	110 / 60.9	103 / 36.9	84 / 48.8	26 / 80.8	20 / 90	37 / 91.9	5 / 80
	0.131 / -0.013	0.11 / 0.038	0.16 / 0.118	0.119 / 0.087	0.083 / 0.031	0.056 / 0.039	0.047 / 0.018	0.038 / 0.021
Brasilia_SONDA	0.45	0.77	0.7	0.79 /	0.8	0.66	0.93	0.92
	114 / 78.9	110 / 59.1	103 / 68	64.6	72 / 68.1	35 / 62.9	22 / 68.2	26 / 76.9
	0.067 / 0.023	0.079 / 0.053	0.089 / -0.007	0.089 / 0.003	0.07 / -0.03	0.088 / -0.048	0.102 / 0.067	0.066 / -0.054
Campo_Grande_SONDA	0.96	0.93	0.95	0.93	0.94	0.95	0.93	0.89
	1217 /	1117 / 75.5	836 / 42.3	940 / 32.3	846 / 29	613 / 22.3	536 / 76.1	557 / 77.2
	0.072 / -0.01	0.077 / 0.004	0.098 / -0.063	0.117 / -0.068	0.111 / -0.086	0.124 / -0.093	0.109 / -0.041	0.132 / -0.041
CUIABA-MIRANDA	0.94	0.94	0.91	0.91	0.92	0.94	0.96	0.96
	1911 / 77	1799 / 71.5	1471 / 51.3	1531 / 43.8	1482 / 44	1044 / 36.2	1110 / 71.5	950 / 81.5
	0.14 / -0.04	0.129 / -0.018	0.166 / -0.051	0.153 / -0.052	0.142 / -0.077	0.139 / -0.086	0.137 / -0.063	0.114 / -0.049
Itajuba	0.81	0.69	0.82	0.71	0.81	0.81	0.89	0.89
	563 / 79.2	541 / 71.5	454 / 67.8	454 / 55.3	396 / 73.5	266 / 52.6	240 / 70.4	226 / 71.7
	0.072 / 0.009	0.069 / 0.021	0.087 / -0.002	0.08 / -0.016	0.064 / -0.021	0.077 / -0.049	0.076 / -0.056	0.074 / -0.058
Ji_Parana_SE	0.97	0.96	0.96	0.95	0.97	0.95	0.98	0.98
	1053 / 88.9	985 / 63.2	852 / 72.1	853 / 50.9	908 / 73.6	449 / 57.2	552 / 90.8	362 / 91.4
	0.124 / -0.026	0.119 / 0.03	0.242 / 0.063	0.202 / 0.067	0.177 / 0.019	0.256 / 0.015	0.107 / -0.01	0.098 / -0.013
Manaus_EMBRAPA	0.87	0.93	0.94	0.92	0.95	0.9	0.78	0.87
	496 / 80	441 / 66	207 / 45.4	258 / 23.6	166 / 64.5	57 / 47.4	95 / 67.4	26 /
	0.064 / -0.014	0.084 / 0.043	0.169 / 0.105	0.175 / 0.126	0.136 / 0.075	0.096 / 0.063	0.096 / 0.053	0.101 / 0.06
Petrolina_SONDA	0.51	0.46	0.47	0.5	0.49	0.55	0.61	0.71
	835 / 67.3	1041 / 63.1	625 / 40.2	671 / 36.8	311 / 23.2	289 / 27.7	233 / 83.7	271 / 90.8
	0.091 / 0.043	0.082 / 0.042	0.097 / -0.068	0.093 / -0.063	0.108 / -0.094	0.099 / -0.086	0.051 / 0.01	0.038 / -0.002
Rio_Branco	0.97	0.94	0.95	0.94	0.97	0.96	0.97	0.97
	2424 / 75.6	1939 / 60.7	1078 / 79.6	856 / 52.3	81.7	362 / 67.7	715 / 89	271 / 88.6
	0.108 / -0.047	0.13 / 0.011	0.175 / 0.03	0.205 / 0.076	0.133 / 0.007	0.202 / 0.022	0.076 / 0.004	0.103 / -0.022
Sao_Martinho_SONDA	0.88	0.77	0.93	0.92	0.94	0.95	0.93	0.96
	417 / 85.6	484 / 82.2	309 / 75.7	434 / 62.4	321 / 77.3	306 / 58.2	326 / 94.8	294 / 96.3
	0.055 / 0.013	0.059 / 0.012	0.136 / -0.006	0.079 / -0.033	0.057 / -0.021	0.071 / -0.046	0.044 / -0.012	0.045 / -0.018
Sao_Paulo	0.79	0.78	0.83	0.85	0.87	0.89	0.78	0.88
	1442 / 59.8	1141 / 64.9	1097 / 65.9	786 / 71.5	775 / 60.6	434 / 65.4	615 / 35.4	418 / 37.3
	0.098 / -0.058	0.098 / -0.04	0.091 / -0.014	0.096 / -0.015	0.09 / -0.041	0.092 / -0.057	0.135 / -0.1	0.13 / -0.105
SP-EACH	0.81	0.78	0.92	0.89	0.93	0.94	0.88	0.95
	291 / 63.9	238 / 76.9	196 / 74.5	188 / 85.1	159 / 64.2	95 / 83.2	98 / 44.9	93 / 61.3
	0.087 / -0.04	0.084 / -0.01	0.082 / 0.01	0.071 / 0.012	0.088 / -0.033	0.06 / -0.026	0.151 / -0.079	0.086 / -0.073

Table D.2 - Comparison of the retrieval accuracy of the MAIAC, DT (3 and 10 km), and DB for different seasons. The bold number is the highest performance among seasons by each indicator.

		MAIAC		DT (3km)		DT (10km)		DB	
		Terra	Aqua	Terra	Aqua	Terra	Aqua	Terra	Aqua
R	Summer	0.77	0.70	0.82	0.85	0.89	0.84	0.58	0.89
	Fall	0.46	0.41	0.38	0.51	0.54	0.38	0.42	0.32
	Winter	0.97	0.94	0.92	0.95	0.96	0.95	0.95	0.94
	Spring	0.96	0.95	0.94	0.94	0.95	0.94	0.95	0.94
R ²	Summer	0.59	0.48	0.67	0.72	0.79	0.71	0.34	0.79
	Fall	0.21	0.17	0.14	0.26	0.29	0.14	0.18	0.10
	Winter	0.94	0.89	0.84	0.91	0.91	0.90	0.90	0.88
	Spring	0.92	0.90	0.88	0.89	0.91	0.88	0.91	0.88
RMSE	Summer	0.083	0.099	0.140	0.108	0.095	0.094	0.075	0.054
	Fall	0.062	0.081	0.100	0.078	0.065	0.077	0.062	0.048
	Winter	0.070	0.080	0.143	0.139	0.117	0.118	0.077	0.080
	Spring	0.167	0.171	0.226	0.232	0.188	0.234	0.163	0.168
MAE	Summer	0.059	0.080	0.098	0.077	0.066	0.075	0.050	0.039
	Fall	0.043	0.063	0.075	0.056	0.050	0.063	0.042	0.035
	Winter	0.048	0.052	0.084	0.073	0.072	0.088	0.047	0.048
	Spring	0.092	0.095	0.128	0.125	0.114	0.124	0.102	0.099
Bias	Summer	0.031	0.064	0.056	0.017	-0.013	0.003	0.005	0.000
	Fall	0.004	0.042	0.020	0.002	-0.023	-0.032	-0.010	-0.016
	Winter	-0.032	-0.009	-0.026	-0.009	-0.030	-0.062	-0.025	-0.035
	Spring	-0.053	-0.017	0.033	0.033	-0.013	-0.021	-0.056	-0.071
Within EE	Summer	67.9	46.8	43.6	55.5	59.1	49.4	75.7	82.7
	Fall	80.0	57.9	49.5	67.2	67.8	52.1	80.3	86.4
	Winter	85.3	78.6	47.4	64.2	60.0	41.7	82.5	82.6
	Spring	72.1	67.5	51.6	53.9	55.9	55.1	67.0	68.2

Table D.3 - Seasonal comparison of the retrieval accuracy of the MAIAC, DT, and DB for different land cover classes.
The bold number is the highest performance among the four seasons by each indicator.

			Forest	Savanna	Grassland	Cropland	Urban	Mixed
R ²	MAIAC	Summer	0.81	0.29	0.69	0.17	0.22	0.22
		Fall	0.09	0.27	0.10	0.35	0.08	0.34
		Winter	0.54	0.03	0.94	0.91	0.87	0.62
		Spring	0.67	0.21	0.92	0.59	0.90	0.72
	DT _{3km}	Summer	0.88	0.42	0.75	0.42	0.36	0.54
		Fall	0.07	0.23	0.19	0.51	0.27	0.14
		Winter	0.65	0.09	0.92	0.89	0.60	0.78
		Spring	0.85	0.23	0.91	0.77	0.79	0.86
	DT _{10km}	Summer	0.89	0.32	0.84	0.49	0.48	0.41
		Fall	0.19	0.08	0.32	0.36	0.20	0.16
		Winter	0.84	0.19	0.95	0.93	0.59	0.80
		Spring	0.85	0.21	0.92	0.83	0.86	0.89
	DB	Summer	0.54	0.38	0.56	0.52	0.25	0.28
		Fall	0.01	0.29	0.28	0.45	0.26	0.18
		Winter	0.81	0.28	0.96	0.93	0.55	0.73
		Spring	0.67	0.19	0.93	0.81	0.72	0.83
RMSE	MAIAC	Summer	0.083	0.086	0.094	0.066	0.100	0.087
		Fall	0.075	0.098	0.074	0.042	0.058	0.079
		Winter	0.061	0.081	0.077	0.061	0.065	0.085
		Spring	0.089	0.084	0.195	0.054	0.187	0.119
	DT _{3km}	Summer	0.222	0.094	0.146	0.058	0.092	0.092
		Fall	0.142	0.073	0.100	0.054	0.088	0.073
		Winter	0.114	0.082	0.157	0.172	0.076	0.144
		Spring	0.173	0.114	0.308	0.067	0.107	0.183
	DT _{10km}	Summer	0.187	0.105	0.097	0.057	0.090	0.073
		Fall	0.094	0.080	0.054	0.046	0.078	0.075
		Winter	0.067	0.088	0.121	0.080	0.087	0.131
		Spring	0.123	0.123	0.274	0.062	0.097	0.170
	DB	Summer	0.084	0.039	0.111	0.029	0.089	0.055
		Fall	0.101	0.036	0.060	0.025	0.088	0.046
		Winter	0.069	0.037	0.057	0.062	0.106	0.095
		Spring	0.100	0.054	0.169	0.034	0.170	0.242
MAE	MAIAC	Summer	0.060	0.062	0.076	0.044	0.076	0.068
		Fall	0.058	0.074	0.055	0.030	0.043	0.056
		Winter	0.048	0.055	0.050	0.035	0.041	0.067
		Spring	0.061	0.060	0.107	0.035	0.099	0.093
	DT _{3km}	Summer	0.144	0.080	0.111	0.050	0.071	0.065
		Fall	0.117	0.058	0.070	0.042	0.062	0.059
		Winter	0.093	0.070	0.074	0.055	0.057	0.094
		Spring	0.142	0.098	0.155	0.054	0.073	0.126
	DT _{10km}	Summer	0.109	0.094	0.073	0.048	0.072	0.060
		Fall	0.076	0.068	0.040	0.039	0.060	0.064
		Winter	0.054	0.080	0.069	0.045	0.069	0.101
		Spring	0.094	0.109	0.134	0.052	0.074	0.135
	DB	Summer	0.064	0.029	0.078	0.020	0.075	0.039
		Fall	0.090	0.026	0.038	0.019	0.069	0.034
		Winter	0.054	0.025	0.034	0.034	0.088	0.058
		Spring	0.065	0.043	0.102	0.025	0.136	0.142
Bias	MAIAC	Summer	0.025	0.042	0.065	0.034	0.062	0.006
		Fall	0.042	0.064	0.035	0.018	0.012	-0.020
		Winter	0.002	0.040	-0.021	-0.007	-0.028	-0.058
		Spring	-0.007	0.035	-0.040	0.010	-0.055	-0.071
	DT _{3km}	Summer	0.136	-0.055	0.099	-0.029	0.017	0.001
		Fall	0.111	-0.024	0.053	-0.029	-0.001	-0.027
		Winter	0.087	-0.062	0.007	0.003	-0.036	-0.068
		Spring	0.139	-0.091	0.108	-0.033	0.009	-0.054
	DT _{10km}	Summer	0.090	-0.094	0.050	-0.039	-0.044	-0.036
		Fall	0.056	-0.067	0.010	-0.031	-0.032	-0.052
		Winter	0.045	-0.078	-0.017	-0.014	-0.056	-0.087
		Spring	0.088	-0.109	0.060	-0.043	-0.036	-0.104
	DB	Summer	0.041	-0.006	0.059	-0.004	-0.067	-0.001
		Fall	0.079	0.004	0.017	-0.014	-0.057	-0.021
		Winter	0.045	-0.001	-0.009	-0.027	-0.078	-0.051
		Spring	0.044	0.002	-0.035	-0.014	-0.110	-0.095

Figure D.1 - Seasonal averaged AOD distributions for MAIAC, DT (3 and 10km), and DB, retrieved over Brazil from 2000 to 2020.

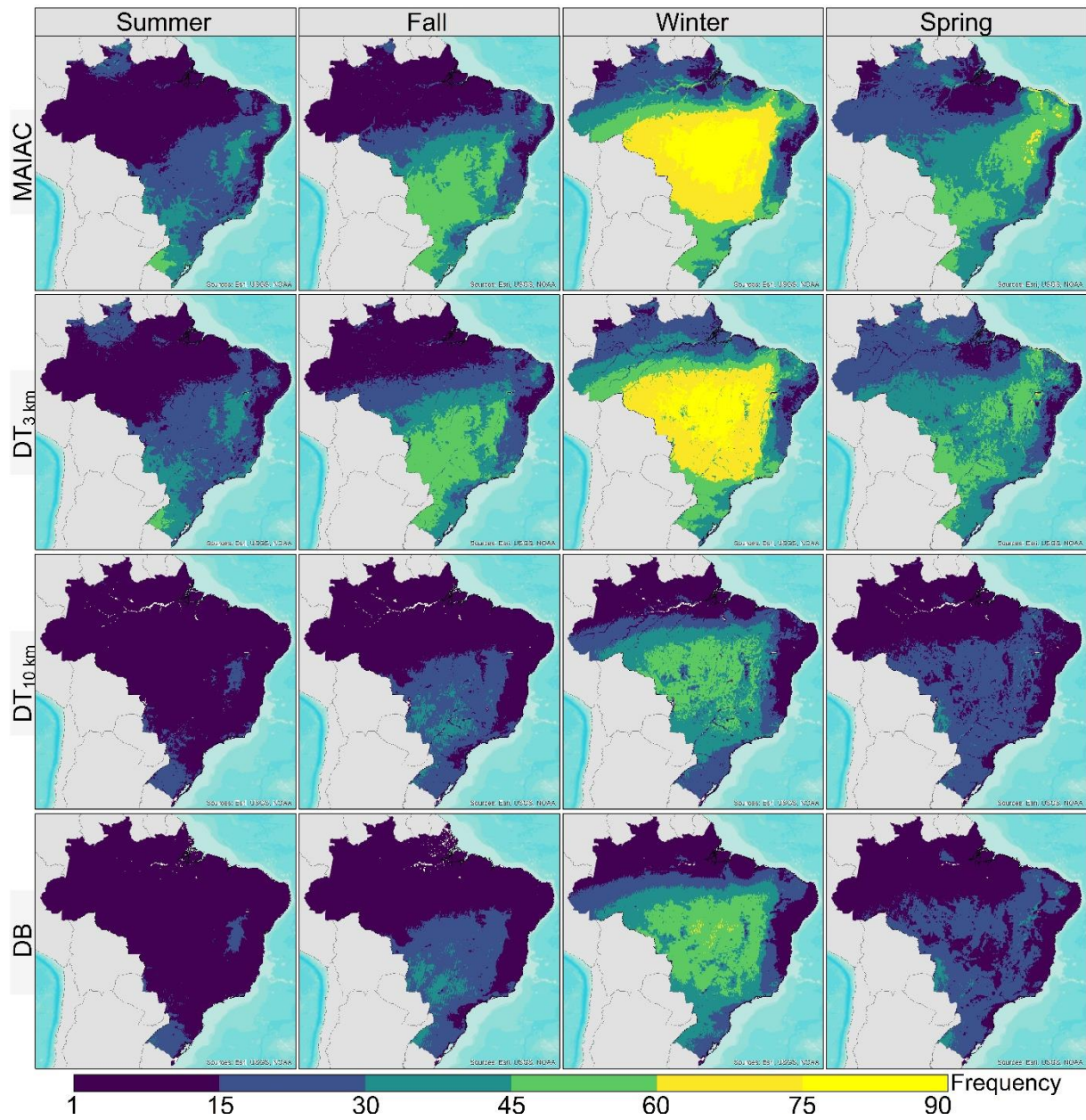


Figure D.2 – Correlation between AOD retrievals obtained from the MAIAC, DT (3 and 10km), and DB algorithms. This analysis considers data from: (A) both Aqua and Terra satellites, (B) only the Terra satellite, and (C) only the Aqua satellite.

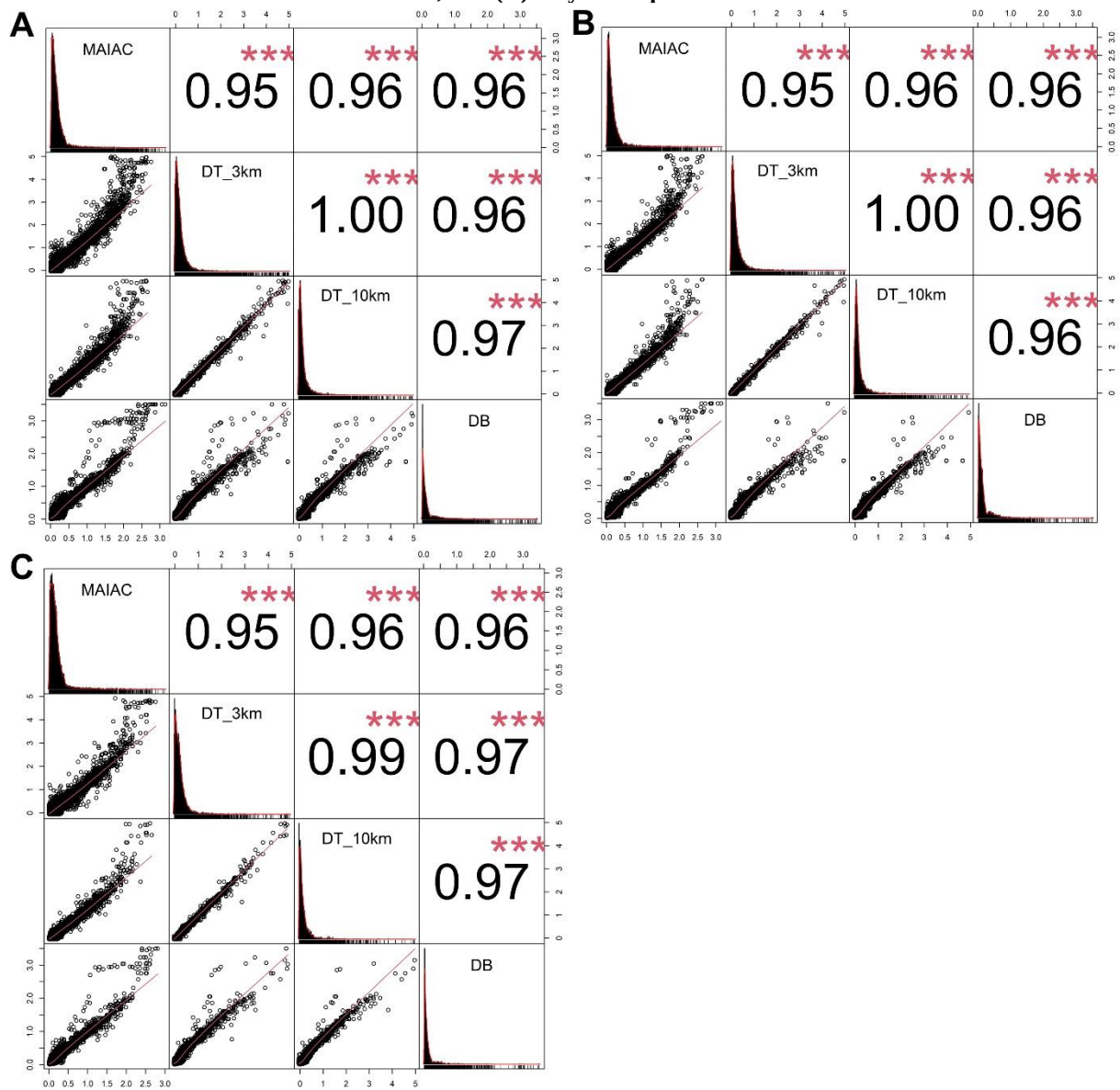


Figure D.3 - Accuracy evaluation of MODIS (Aqua and Terra) AOD algorithms [MAIAC, DT (3 and 10 km), and DB] against AERONET sites classified as Forest across Brazil).

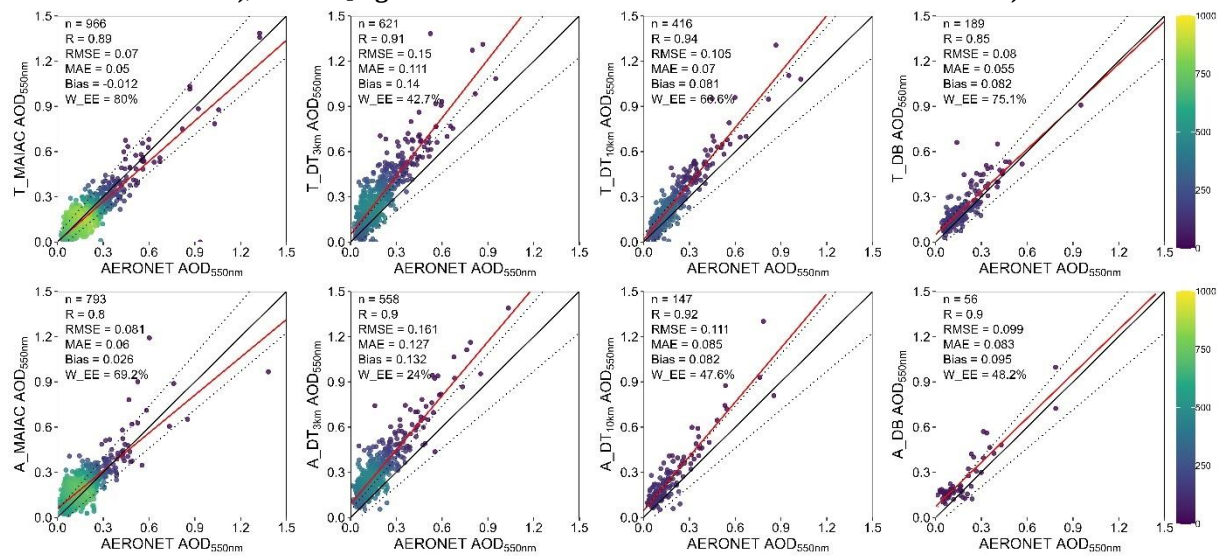


Figure D.4 - Same as Figure D.3, but for Savanna.

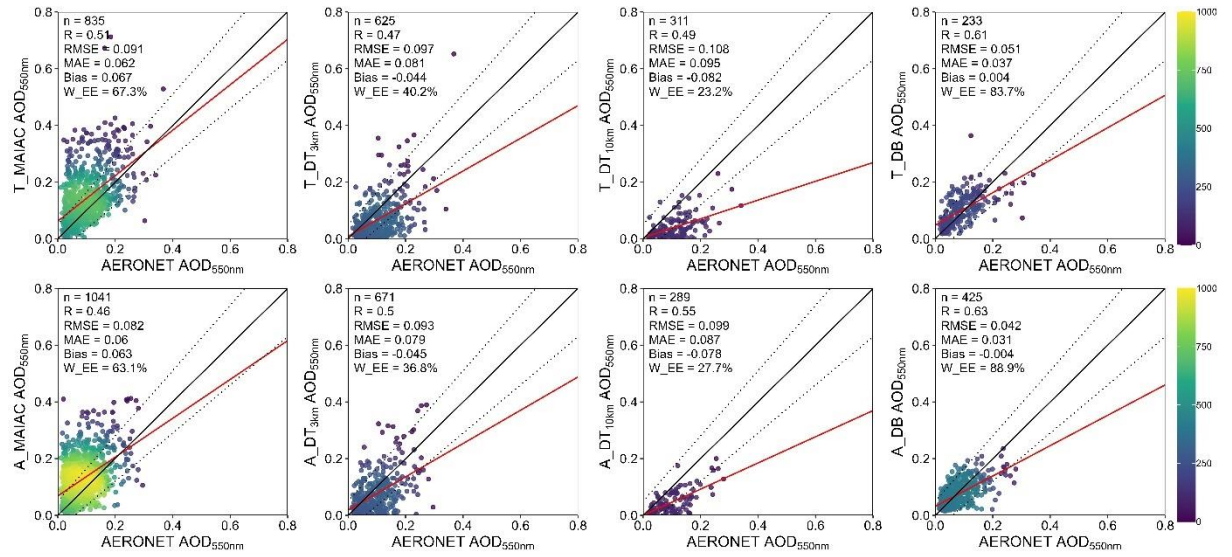


Figure D.5 - Same as Figure D.3, but for Grassland.

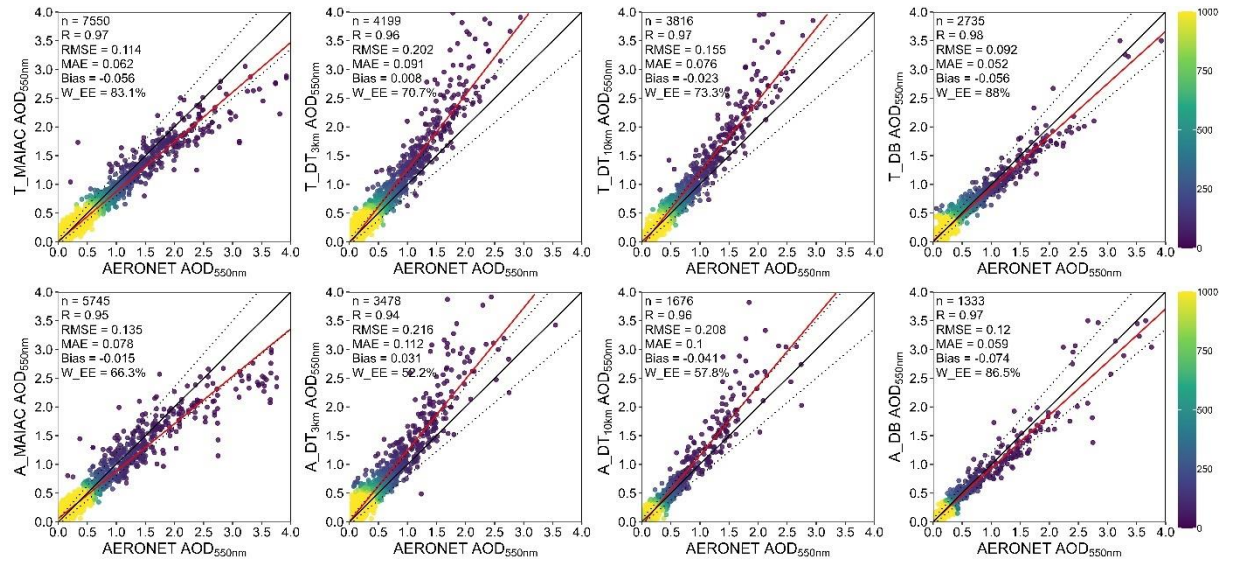


Figure D.6 - Same as Figure D.3, but for Cropland.

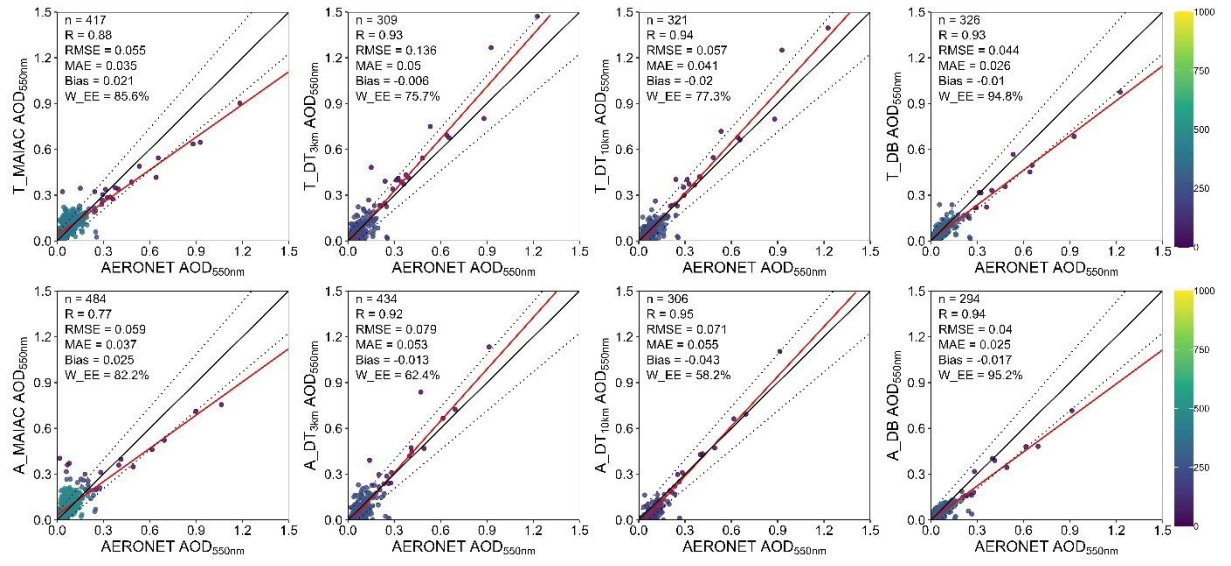


Figure D.7 - Same as Figure D.3, but for Urban.

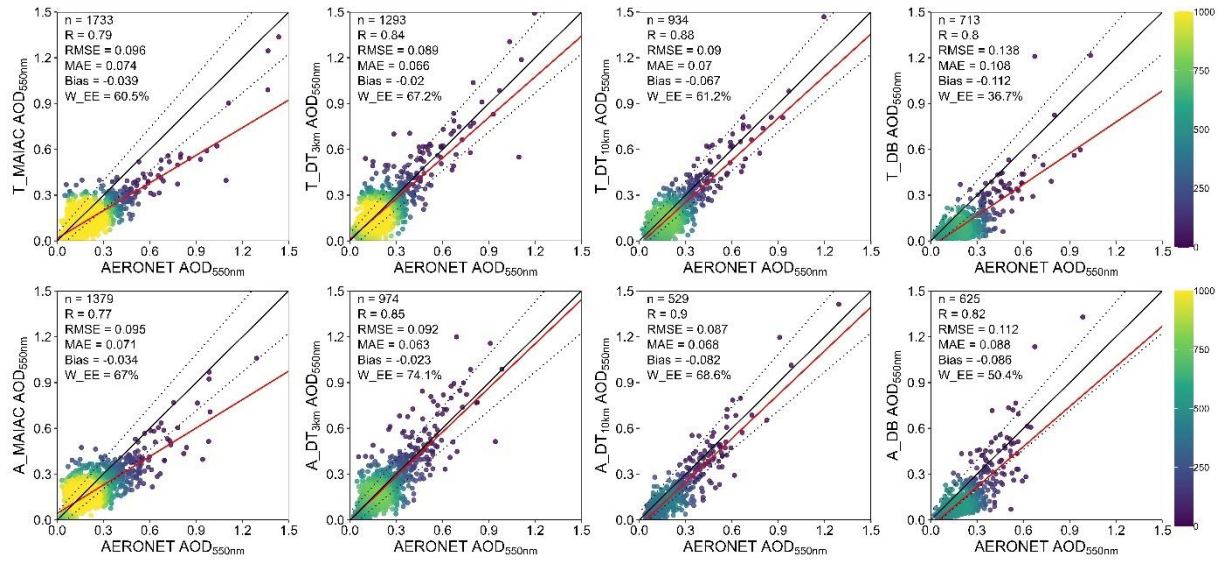


Figure D.8 - Same as Figure D.3, but for Mixed.

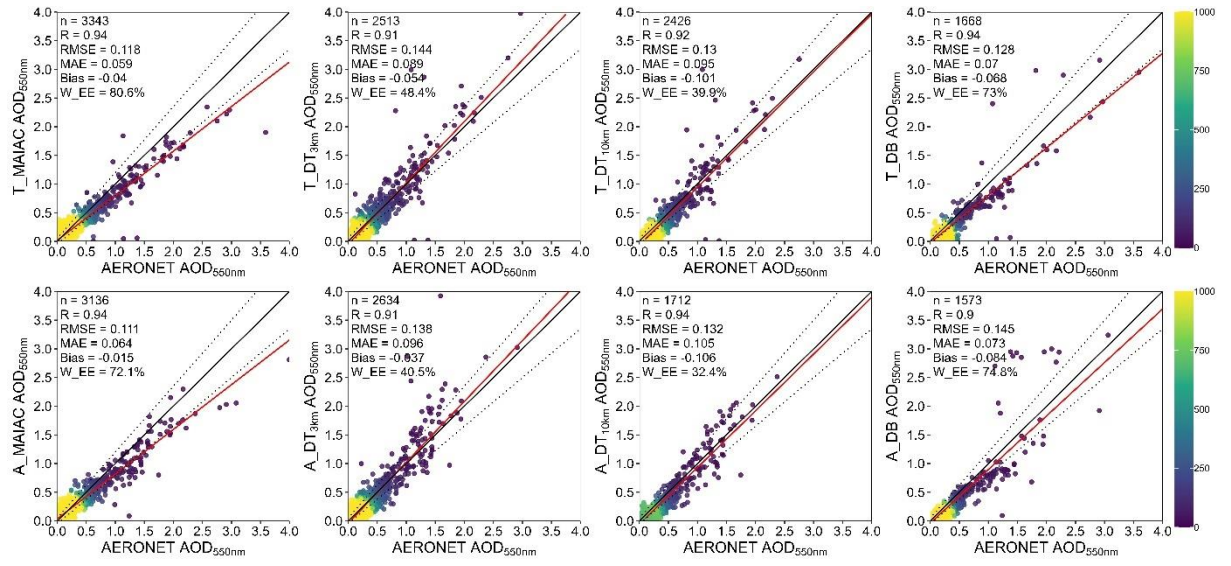


Figure D.9 - Seasonal variation in the aerosol volume size distribution for the different land cover classes in Brazil.

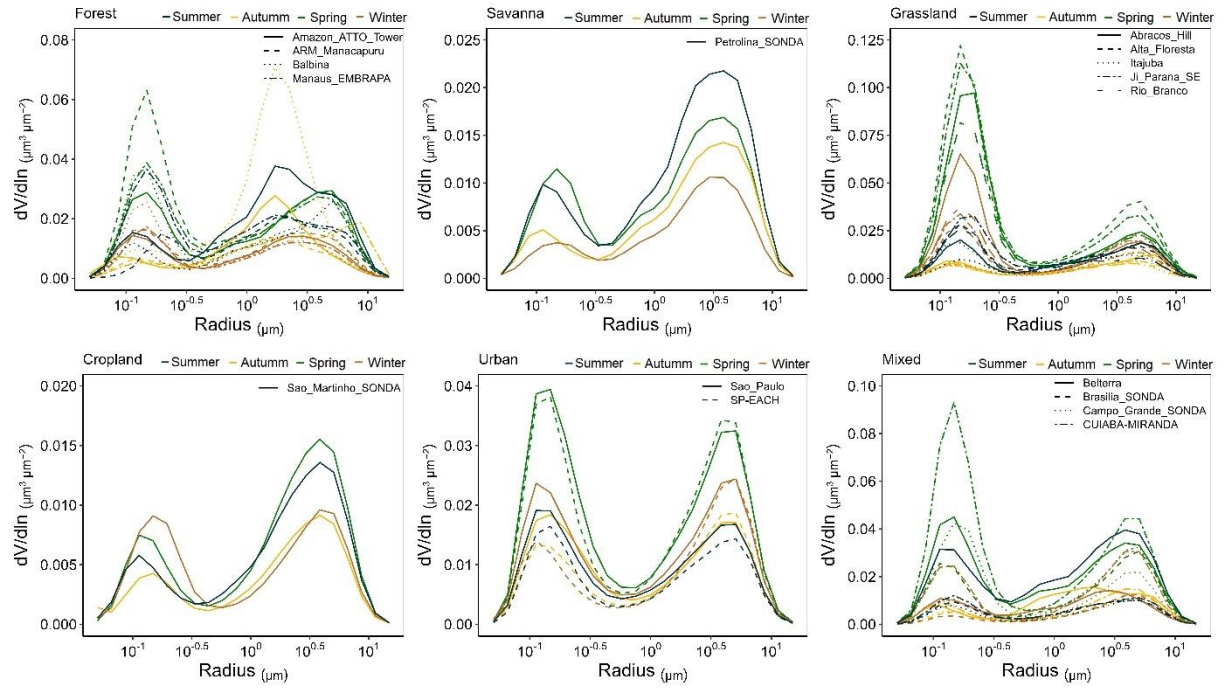


Figure D.10 - Aerosol classification for each AERONET site.

	Aerosol Type										
	I	II	III	IV	V	VI	VII	VIII	IX	X	
Amazon_ATTO_Tower	7	0	60.1	17.1	0	0.4	15.4	0	0	0	Forest
ARM_Manacapuru	0	20.9	44.6	26.2	8.3	0	0	0	0	0	
Balbina	0	0	48.9	51.1	0	0	0	0	0	0	
Manaus_EMBRAPA	1.2	0	51.1	33.8	0	1	0	0	0.5	12.5	
Petrolina_SONDA	32.6	58.7	8.7	0	0	0	0	0	0	0	Savanna
Abracos_Hill	0	2.8	83.6	13.6	0	0	0	0	0	0	Grassland
Alta_Floresta	0.5	7.7	74	17.8	0	0	0	0	0.1	0	
Itajuba	0	47.7	50	2.3	0	0	0	0	0	0	
Ji_Parana_SE	0	2.1	66.5	31.4	0	0	0	0	0	0	
Rio_Branco	2.5	22.7	65.2	9.6	0	0	0	0	0	0	Cropland
Sao_Martinho_SONDA	0	16.2	80	3.8	0	0	0	0	0	0	Urban
SP-EACH	0	16.1	78.4	5.2	0	0	0	0	0.3	0	
Sao_Paulo	14.6	40.4	37.7	4.3	0.5	0	0	0	2.5	0.1	
Belterra	0.4	45.3	39.3	12	0.4	0.4	0	0	2.2	0	Mixed
Brasilia_SONDA	5.2	94.8	0	0	0	0	0	0	0	0	
Campo_Grande_SONDA	0.5	26.9	70	2.7	0	0	0	0	0	0	
CUIABA-MIRANDA	2.6	36.7	50.8	10	0	0	0	0	0	0	

Figure D.11 - Bias, RMSE, and percentage of retrievals within EE envelope for MAIAC, DT, and DB algorithms in each AERONET site.

