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Exploring the relationship between high-resolution aerosol optical depth values and ground-level particulate matter concentrations in the Metropolitan Area of São Paulo

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HIGHLIGHTS

- MAIAC AOD retrievals correlated weakly with PM concentrations in the MASP.
- We evaluated MAIAC AOD retrievals over the MASP.
- The MAIAC aerosol model is not compatible with the aerosol properties in the MASP.
- There is a wide variety of aerosol types in the MASP.

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ABSTRACT

The spatiotemporal pattern of particulate matter (PM) concentrations is an important factor in predicting health issues in inhabitants of urban areas. The integration of satellite-derived aerosol optical depth (AOD) data with ground-level PM concentration data, obtained from monitoring networks, has contributed to better characterization of the spatiotemporal variability of aerosols worldwide. However, before using satellite AOD data as a proxy for PM in epidemiological and air quality studies in specific regions, the applicability of that strategy must be evaluated. In this study, we evaluate the use of the high-resolution AOD, derived from Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm, as a predictor of surface PM concentrations in the Metropolitan Area of São Paulo (MASP). We found relatively weak or negative correlations between PM concentrations and MAIAC AOD, even after vertical correction by planetary boundary layer height and the hygroscopic growth factor. The weak correlations reported in this study are mainly due to the mismatch between the current MAIAC aerosol model and the properties of local aerosols in the MASP. Our results suggest that sources of aerosol particles in the MASP are quite diverse and that there is therefore no single optical model suitable for use with satellite-derived AOD.

1. Introduction

Particulate matter (PM) is the solid component of atmospheric aerosol, which is defined as a suspension of solid or liquid particles in a gas (Seinfeld and Pandis, 2016). These particles play a central role in regulating climate and air quality, by directly and indirectly affecting the radiative balance on Earth. Depending on their size and composition, these particles can scatter or absorb solar radiation, changing the surface

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temperature (direct effect) or altering the cloud lifetime and albedo (indirect effect) (IPCC, 2013). In addition, the impact of PM on human health is well known, the degree of harm being inversely related to the particle size. A literature review correlating PM_{10} (particles with aerodynamic diameter of less than 10 μm) and $PM_{2.5}$ (particles with aerodynamic diameter of less than 2.5 μm) with various morbidity and mortality outcomes is presented in a World Health Organization (WHO) report (World Health Organization, 2006). In this article, PM refers only to $PM_{2.5}$ and PM_{10} and no other size of particulate matter.

The most harmful air pollutants, such as PM, are typically measured in air quality monitoring networks, which are sparse in low- and middle-income countries (Fajersztajn et al., 2014; World Health Organization, 2016). In Brazil, there are 27 "federative units" (26 states and the Federal District of Brasília), of which only eight have air pollutant monitoring networks: Bahia, Espírito Santo, Federal District of Brasília, Minas Gerais, Paraná, Rio de Janeiro, Rio Grande do Sul and São Paulo. Of those eight, only the states of São Paulo and Rio de Janeiro have monitoring networks somewhat comparable to those employed in the United States and Europe (Alves et al., 2014; IEMA, 2014). In view of that, the integration of high-resolution satellite data with ground-level monitoring network data can contribute to better characterization of the spatiotemporal variability of aerosols at local and regional scales (Chudnosvsky et al., 2014a).

The satellite-retrieved parameter related to PM is the Aerosol Optical Depth (AOD), which is a quantitative measure of the attenuation of the electromagnetic radiation by the presence of aerosols in the atmospheric column, at a specific wavelength (Wallace and Hobbs, 2006). The AOD is one of the atmospheric products of the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor aboard the Terra and Aqua satellites. However, the coarse spatial resolution of the AOD (10×10 km^2 and 3×3 km^2) has prohibited its applicability in local scale studies. Recently, a higher spatial resolution (1×1 km^2) MODIS AOD was derived from the Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm (Lyapustin et al., 2018), which can have beneficial applications in epidemiological and air quality studies, mainly in urban regions.

In the past 10–15 years, satellite-retrieved AOD data have been successfully applied in several approaches, in order to retrospectively predict the concentrations of PM, which can be used by health studies. Those approaches can be classified as statistical or geophysical (Jin et al., 2019). Chu et al. (2016) presented a review of the studies that used different methodologies for the retrospective prediction of $PM_{2.5}$ using satellite-retrieved AOD. The authors included 116 studies and found that four types of model were the most frequently used in the predictions: Multiple Linear, Linear Mixed Model, Geographically Weighted Regression and Chemical Transport Model. The last is a geophysical approach while the others are statistical. Examples of the application of these approaches to retrospectively predict PM_{10} can be found in the works of, amongst others, Stafoggia et al. (2017) and Kumar et al. (2016). Recently, in order to retrospectively predict the PM concentrations, there have also been approaches that combine the geophysical and statistical approaches (e.g., Knibbs et al., 2018; Beloconi et al., 2018; Meng et al., 2019; van Donkelaar et al., 2019).

The populations most affected by air pollution are those living in urban areas, in which pollutant levels often exceed the standards established by the WHO (World Health Organization, 2016). In 2019, about 56% of the global population resided in urban areas (World Urbanization Prospects, 2019a), while in the South America that percentage was higher than 85% (World Urbanization Prospects, 2019b). The Metropolitan Area of São Paulo (MASP) is the most populous region in Latin America, and vehicle emissions constitute the main source of air pollution in the area (CETESB, 2017). Although the effects of air pollution levels on human heath in the MASP have been described in many epidemiological studies (Braga et al., 2001; Ribeiro and Cardoso, 2003; Saldiva et al., 1994, 1995; Ribeiro et al., 2019; Takano et al., 2019), few studies have attempted to retrospectively predict surface PM concentrations (Habermann and Gouveia, 2012), and only one study has investigated the applicability of satellite-retrieved AOD (Natali, 2008), in this area.

Before using satellite AOD data as a proxy for PM in epidemiological studies and air quality studies in specific regions, the applicability of the strategy must be evaluated. Therefore, the aim of this study was to evaluate the use of high-resolution MAIAC AOD data to predict surface PM concentrations in the MASP in the 2012–2017 period. To that end, we integrated ground-level measurements with satellite data, describing the data processing procedures. In addition, we presented an evaluation of the MAIAC-AODs data performance over the MASP.

2. Methods

This study was conducted in the MASP, which comprises the city of São Paulo and 39 surrounding cities (Fig. 1). It is an urban region with a total area of 7946 km^2 and over 21 million inhabitants (https://emplasa.sp.gov.br/RMSP).

The climate of the MASP is characterized by two seasons (Andrade et al., 2012a): a wet season (between October and March) and a dry season (between April and September). During the dry season, when the planetary boundary layer (PBL) is low and atmospheric conditions do not favor air pollutant dispersion, episodes of high PM concentrations are common in the MASP (de Almeida Castanho and Artaxo, 2001; Albuquerque et al., 2012; de Miranda et al., 2012; Carvalho et al., 2015; CETESB, 2017).

2.1. Particulate matter surface data at MASP

Hourly PM mass concentrations data, in the 2012–2017 period, were obtained from automatic air quality monitoring stations operated by the São Paulo State *Companhia de Tecnologia de Saneamento Ambiental* (CETESB, Environmental Protection Agency). The CETESB measures PM concentrations using an automated beta attenuation monitor (CETESB, 2017; Xavier, 2011; Thermo Fisher Scientific Inc., 2010), with a minimum detectable concentration limit of 4 μg/m^3 and a 1-h time resolution.

In the MASP, CETESB stations are impacted mainly by vehicular emissions. The difference is in the fleet type, although some of the stations are located near industrial areas (Mauá and Santo André-Capuava) or inside parks (Ibirapuera, Cidade Universitária - USP and Pico do Jaraguá). The stations at Congonhas, Marginal Tietê-Ponte dos Remédios, Osasco, Pinheiros e São Caetano do Sul are impacted by light-duty and heavy-duty vehicles. The others are affected mainly by light-duty vehicles and buses (Carvalho et al., 2015).

Considering the recommendations of the "*Code of Federal Regulations - Title 40 – Protection of Environment/Part 58 – Ambient Air Quality Surveillance/Appendix E to Part 58 – Probe and Monitoring Path Siting Criteria for Ambient Air Quality Monitoring*" developed by the United States Environmental Protection Agency (USEPA), CETESB classifies the monitoring stations according to their spatial representativeness. The classification takes into account the pollutant, the characteristics of the monitored region (e.g., topographic and meteorological characteristics), as well as the impact of emissions from fixed and mobile sources (e.g., proximity and intensity) in the stations (CETESB, 2016).

Regarding the PM concentrations, since in the MASP the main source of these concentrations is vehicular emission, the distance from the major traffic lanes is one of the characteristics considered in the spatial representativeness classification. The stations are classified on the micro, medium, suburban and urban scales. The former two are highly impacted by the local emissions while the latter are representative of areas of dimensions varying between 501 m and 50 km (CETESB, 2016).

In the present study, we considered the stations classified as suburban and urban, since their spatial representativeness is close to or higher than that of the satellite data. The number of CETESB stations measuring PM concentrations, which met the criteria for spatial representativeness in the present study, varied during the study period (Table S1 in the

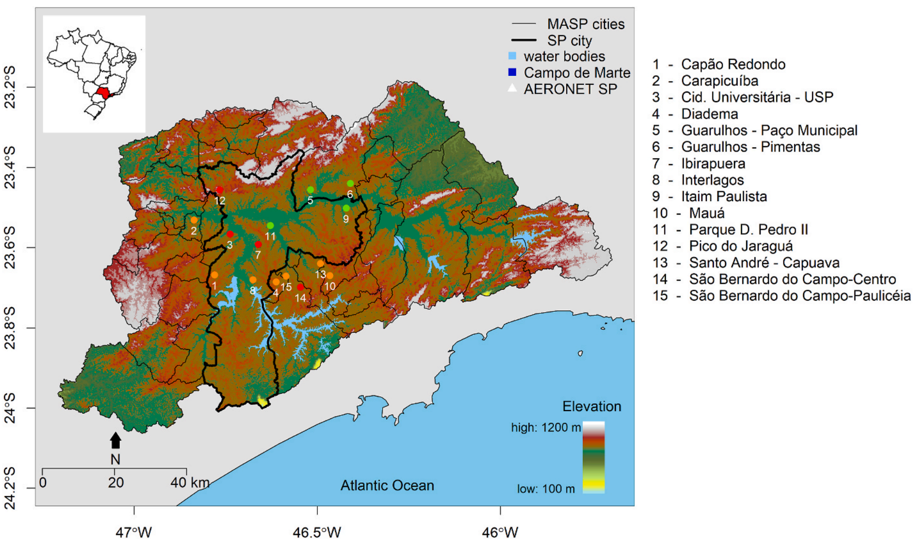

Fig. 1. Topography map of the study area showing the city of São Paulo (SP city), as well as the other cities in the MASP, and the locations of the CETESB monitoring stations used in this study in 2017, the AERONET site, and the Campo de Marte Airport. Red and orange dots represent stations measuring $PM_{2.5}$ and PM_{10} only, respectively. Green dots represent stations measuring both pollutants. Inset shows the state of São Paulo (in red) within a map of Brazil. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Supplementary Information (SI)). Fig. 1 shows the location of the ground-level monitoring stations used in the present study for the year 2017.

For each CETESB station, we considered three distinct time intervals for calculating the mean PM concentrations: the hour of the satellite overpasses; diurnal (average of measurements obtained over 6–18 h periods); and daily (average of 24-h measurements). Diurnal and daily PM mean concentrations were included because they are used in epidemiological studies.

2.2. MODIS–MAIAC AOD

In this study, we used the MODIS–MAIAC AOD (MCD19A2 product), with 1 × 1 km^2 of spatial resolution, at a wavelength of 550 nm, retrieved from the Terra and Aqua satellites for the 2012–2017 period. The data are available on Earthdata website (https://earthdata.nasa.gov), and we used only the high-quality retrievals (those with a Quality Assurance bit 0000, which represents "Best quality"; https://modis-land.gsfc.nasa.gov/pdf/MCD19_UserGuide_final_Feb-6-2018.pdf). The data were processed by using the HDF-EOS to GeoTIFF Conversion Tool (HEG, https://newsroom.gsfc.nasa.gov/sdptoolkit/HEG/HEGHome.html). During the study period, the Terra satellite passed over the MASP from 9:00 to 11:30 (local time), whereas the Aqua satellite passed over the area from 12:45 to 15:05 (local time).

A detailed description of the MAIAC algorithm can be found in the studies conducted by Lyapustin et al. (2011a, b; 2012; 2018). In brief, it is a generic algorithm based on image-processing and time series analysis. Validation against AErosol RObotic NETwork (AERONET, https://aeronet.gsfc.nasa.gov; Holben et al., 1998) AOD data has shown that the MAIAC algorithm is capable of retrieving aerosol data over dark (vegetated) surfaces with accuracy comparable to that of the MODIS level 2 aerosol product from the Terra satellite (10 × 10 km^2 resolution) and that its accuracy increases over brighter surfaces, such as deserts and urban areas in the United States (Lyapustin et al., 2011b). The validation study conducted by Zhang et al. (2019) over China, showed that the MAIAC performed better in urban/cropland regions than in other regions (arid, semiarid, general vegetation, and water).

The MAIAC AOD data (Terra and Aqua retrievals) have been validated across South America. The validation procedure consisted of comparing the average MAIAC AOD retrievals collocated spatially and temporally with the average AOD from AERONET sites in the 2000–2016 period. The Pearson's correlation coefficient (R) for the relationships that the AOD data from the São Paulo AERONET station, located atop the tallest building of the Physics Institute on the main campus of the University of São Paulo (latitude: −23.561°, longitude: −46.735°), had with the Terra and Aqua retrievals of MAIAC AOD data was $R = 0.90$ and $R = 0.82$, respectively. Details about the validation study of MAIAC data for South America can be found in Martins et al. (2017).

2.3. Corrections by PBL height and by the hygroscopic growth factor

The relationship between AOD and PM concentrations (AOD–PM relationship) is not straightforward and is affected by a variety of factors, including the PBL height, the relative humidity (RH), and the vertical distribution of aerosols (Chu et al., 2003; Li et al., 2015). Because AOD is a value associated with the atmospheric column, it is affected by the PBL height, which has an impact on the vertical profile of aerosols. In general, aerosols are well mixed under the PBL (Gupta et al., 2006; Koelemeijer et al., 2006; Guinot et al., 2006; Zhang et al., 2009a). However, when there is aerosol transport at different heights, the AOD–PM relationship is altered, because ground-level monitoring stations do not capture the PM concentrations at higher levels. In addition, disconnected aerosol layers in the atmospheric column can differ in terms of their physical and chemical composition (Slater and Dibb, 2004). Therefore, the PBL height and the vertical distribution of aerosols are key variables for determining the AOD–PM relationship.

Assuming that aerosols are well mixed under the PBL, previous studies have corrected satellite-derived AOD using the vertical correction depicted in equation (1) (Koelemeijer et al., 2006; Schaap et al., 2009; Boyouk et al., 2010; Chu et al., 2013; Tsai et al., 2011; Chew et al., 2016; Kong et al., 2016; Zheng et al., 2017; Gong et al., 2017):

$$ASH = \frac{AOD}{PBLH} \tag{1}$$

where ASH is the aerosol scale height and $PBLH$ is the PBL height. Given that we are not integrating the aerosol extinction in the total atmospheric column, the term "AOD" is incorrect and we will use the term "ASH". That correction eliminates aerosols at higher levels, taking into account only those under the PBL, thereby typically improving the AOD–PM relationship (Koelemeijer et al., 2006).

We calculated morning and afternoon estimates of the PBL height using balloon sounding data from Campo de Marte Airport (latitude: −23.515°, longitude: −46.647°). Atmospheric balloon sounding data for stations around the world are available in http://weather.uwyo.edu/upperair/sounding.html, balloons being launched daily at 12:00 UTC and 00:00 UTC. Following the methodology employed by Sánchez et al. (2020), who studied the dynamic and thermodynamic properties of the

PBL in the MASP, we estimated morning and afternoon PBL height by the parcel and air temperature gradient methods, respectively.

We obtained morning PBL height estimates by virtual potential temperature profiles from 12:00 UTC balloon sounding data using the parcel method, in which the estimated PBL height is that at which the virtual potential temperature is the same as the surface virtual potential temperature. Due to the fact that balloon soundings are not carried out during the afternoon, we estimated the residual-mixing layer (RML) height by determining the air temperature profiles at 00:00 UTC and, as suggested by Sánchez et al. (2020), used the RML height as an approximation of the maximum daytime PBL height. The RML height was obtained, by using the air temperature gradient method, as the height at which the air temperature gradient is equal to or higher than zero. Using that methodology, we were able to correct the MAIAC AOD data by PBL height: we used morning PBL height estimates to correct the data retrieved from the Terra satellite and afternoon RML height estimates to correct the data retrieved from the Aqua satellite. Additional details regarding those methods can be found elsewhere, including the studies conducted by Stull (1988) and by Seidel et al. (2010).

The RH is also a key variable for the AOD–PM relationship. In the automated beta attenuation monitor employed by the CETESB (CETESB, 2017; Xavier, 2011), the PM concentrations are determined after heating the sample tube above ambient temperature to remove condensation, as well as to allow the temperature and humidity to be standardized in all measurements (Thermo Fisher Scientific Inc., 2010). Therefore, PM measured in this way are a dry aerosol mass, whereas AOD is measured under environmental conditions and is influenced by the RH. Depending on the hygroscopic nature of the aerosols, an increase in RH can modify their chemical composition, size distribution, and other characteristics; when the RH is higher, aerosol particles scatter more light, affecting the AOD loadings (Liu et al., 2013). The ratio between the aerosol scattering in an ambient air parcel with a specific RH and the aerosol scattering in dry air conditions is known as the hygroscopic growth factor—f(RH)—which depends on the hygroscopic characteristics of the aerosol, the time of day, and the location (Day and Malm, 2001). Another classical correction, displayed in equation (2), accounts for the increase in aerosol loading due to an increase in RH:

$$AOD_{dry} = \frac{AOD}{f(RH)} \tag{2}$$

where AOD_{dry} is the AOD corrected by hygroscopic growth factor.

The f(RH) is defined by performing specific experiments (Li et al., 2005). In the present study, we adopted the f(RH) functions obtained from experiments performed by Rodrigues (2014a, b) between 2013 and 2014 in São Paulo. She fitted the f(RH) function shown in equation (3) (Hänel, 1976) to the data generated by Lidar Raman techniques and found rates of growth of the aerosol under increasing RH values (γ)(standard deviation) equal to 0.11 (0.03), 0.48 (0.12) and 0.78 (0.16) on specific days in the fall, spring and summer seasons, respectively, when considering $RH_0 = 40\%$. In the period of our study, we used those values in the functions to correct the AOD retrieved in the fall, spring and summer, respectively; we used the value obtained in the fall in the function to correct the AOD retrieved in the winter.

$$f(RH) = \left(\left(1 - \frac{RH}{100}\right) \Big/ \left(1 - \frac{RH_0}{100}\right)\right)^{-\gamma} \tag{3}$$

We obtained hourly RH values from 14 of the CETESB stations and 29 monitoring stations operated by the São Paulo Municipal Climate Emergency Management Center (https://www.cgesp.org/v3/). For sites at which RH measurements were unavailable, the RH values obtained at the nearest station were used.

In this study, we evaluated the correlation between PM concentrations and MAIAC AOD values, considering the corrections depicted in equations (1) and (2), as well as the combination of both corrections, as displayed in equation (4):

$$ASH_{dry} = \frac{AOD}{f(RH) \times PBLH} \tag{4}$$

where ASH_{dry} is the ASH corrected by hygroscopic growth factor.

2.4. Integration data and intercomparison

All statistical analyses were carried out with the R statistical software, version 3.5.0 (www.r-project.org). We calculated the Pearson coefficient *R* for the relationship between the Terra and Aqua retrievals of MAIAC AOD data and collocated PM concentrations (in the same MAIAC AOD grid cell), for all three averaging intervals during the 2012–2017 period. In our calculation of the mean diurnal and daily PM concentrations, we included only days on which at least two thirds of the measurements were valid (CETESB, 2017).

In order to eliminate atypical AOD values, which were identified in the analysis of the dispersion graphs between AODs and PM, we excluded all AOD with values above the 99th percentile from our analyses. The atypical AOD values occurred in the spring, when the MASP is impacted by long-range transport of smoke from biomass burning. This takes place in the countryside of São Paulo state, in the northern/central-west regions of Brazil, and in neighboring countries, including Bolívia, Paraguay, and Argentina, thus contributing to AOD enhancement in the MASP (Mariano et al., 2010; de Miranda et al., 2017; Yamasoe et al., 2017; Vara-Vela et al., 2018).

3. Results and discussion

3.1. Correlations between PM concentrations and MAIAC AODs

The correlations between PM concentrations and MAIAC AODs, for the satellites overpass, in the period as a whole and in each season, based on data from all of the CETESB stations, are presented in Table 1. Table S2 in the Supplementary Information (SI) presents the same information but for diurnal and daily averaging intervals. Seasons are defined as summer (December–February), fall (March–May), winter (June–August), and spring (September–November). For both PMs, the correlations were relatively weak. The strongest correlations were between the PM concentrations during Aqua overpasses and the MAIAC Aqua AODs. The MAIAC Terra AODs and the PM concentrations presented mostly negative correlations. Comparing the seasons in each averaging interval, the strongest correlations were observed in the spring.

The differences between satellites in terms of the strength of the correlations can be explained by the aerosol mixing state in the atmospheric column. The Terra satellite passes over the MASP during the mid-morning hours, when the PBL begins to evolve and, as a

Table 1
Linear correlations, by period/season, between PM concentrations and MAIAC AOD values, for the satellites overpass.

	Period/season	PM_{10}		$PM_{2.5}$	
Satellite		n	R	n	R
Aqua	2012–2017	3563	0.201	1345	0.229
	Summer	230	0.236	50	0.284
	Fall	503	0.243	174	0.144
	Winter	1949	0.179	759	0.241
	Spring	881	0.249	362	0.245
Terra	2012–2017	5681	−0.031	1942	0.021
	Summer	1019	−0.119	268	−0.030
	Fall	1159	−0.030	352	0.003
	Winter	2086	−0.089	768	−0.056
	Spring	1417	0.111	554	0.120

Summer, December–February; Fall, March–May; Winter, June–August; Spring, September–November.

consequence, aerosols are not well mixed in the atmospheric column. In contrast, the Aqua satellite passes over the MASP in the early afternoon, when the PBL has evolved and aerosols are well mixed in the atmospheric column.

In addition, some studies demonstrated that the quality of the MODIS Terra retrievals worsened in MODIS Collection 5, due to the degradation of the blue band sensor. (Levy et al., 2010; Wang et al., 2012; DeVisser and Messina, 2013; Zhang et al., 2017). Even after the improvements in MODIS Collection 6, in 2014, the degradation of the MODIS Terra sensor affected MAIAC Terra AODs "via striping and a positive bias on the left-hand side of the MODIS scan, mostly over bright surfaces" (Lyapustin et al., 2018).

In earlier studies, mainly conducted in the United States, weak correlations between ground-level $PM_{2.5}$ concentrations, and AODs were reported. They found that correlations were stronger in the eastern portion of the United States ($R = 0.4$–0.8) than in the western portion ($R = 0.2$–0.4) (Toth et al., 2014; Engel-Cox et al., 2004; Paciorek et al., 2008; Zhang et al., 2009b). Loría-Salazar et al. (2016) suggested that the correlations are stronger in the eastern portion of the United States because of the well-behaved boundary layer, the more uniform vegetation coverage (which leads to lower surface reflectance), and the lower elevations, as well as the homogeneous distribution of sulphates and organic carbon.

Koelemeijer et al. (2006) reported that PM (hourly and daily) concentrations correlated weakly with MODIS (collection 4)-derived AOD in Europe. Using a decadal dataset, Hersey et al. (2015) studied the AOD–PM relationship across five major metropolitan areas in South Africa and also found weak, negative correlations between mean monthly PM concentrations and AOD.

To our knowledge, only Natali (2008) has investigated the relationship that ground-level PM concentrations have with MODIS AODs in the MASP. The author used the methodology proposed by de Almeida Castanho et al. (2008) to retrieve AODs at high resolution (1×1 km^2) over the MASP between 2002 and 2005, reporting correlations that were weak, negative, or both. Because there were few stations monitoring $PM_{2.5}$ in the MASP during the 2002–2005 period, that author analyzed $PM_{2.5}$ data from only two ground-based stations. Therefore, in the present study, we summarize the PM_{10} results from that study. We found that the Aqua AODs correlated positively (although weakly) with the PM_{10} concentrations during satellite overpasses ($R = 0.03$) and during the diurnal averaging intervals ($R = 0.02$), whereas the Terra AODs correlated negatively with both ($R = -0.09$ and $R = -0.1$, respectively). The correlations between the AODs and the mean PM_{10} concentrations during the daily averaging intervals were negative for the Aqua and Terra satellites ($R = -0.06$ and $R = -0.20$, respectively). Natali (2008) identified patterns comparable to those observed in the present study.

3.2. *Effect of PBL height and RH on the AOD–PM relationship*

The correlations between PM concentrations and MAIAC AODs (corrected on the basis of equations (1), (2) and (4)) in the study period as a whole and for each season, for satellites overpass, based on data from all of the CETESB stations, are presented in Table 2. SI Table S3 presents the same information but for the diurnal and daily averaging intervals. Because of missing data related to PBL height estimates, the correlations in Table 1 and SI Table S2 were recalculated and are presented in Table 2 and SI Table S3, respectively, for comparison.

The fact that some PBL height (balloon sounding) data were missing was due to technical problems during balloon launches, problems during data acquisition, or problems related to the estimation method, such as when it was unable to detect inversions in the profiles (mainly during stable conditions). The mean reduction in the sample size associated with the missing PBL height estimates was 38% and 53% for the morning and afternoon periods, respectively. The frequency of missing data was higher in the afternoon period because the afternoon PBL heights were estimated indirectly on the basis of the RML height, which is not recorded every day. For the morning and afternoon periods, we attempted to use PBL height estimates from the Climate Forecast System Reanalysis, version 2, developed by the National Center for Environmental Prediction (https://rda.ucar.edu). However, those estimates did not present good agreement with the data obtained from soundings and were therefore not employed as a proxy for the missing data.

After recalculation of the correlations in Table 1 and SI Table S2, the pattern of the results did not change. Slight improvements were observed after the correction by PBL height (equation (1)), mainly for the MAIAC AOD Terra data, suggesting that the assumption of a well-mixed PBL is not suitable for the MASP (Tsai et al., 2011; Chu et al., 2013). Therefore, information on the vertical profiles of aerosols, provided by Lidar instruments, at ground level or aboard satellites (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation), appears crucial to the further investigation of the AOD–PM relationship. There is a need for investigations of this specific topic in the MASP.

The correlations were also improved by the f(RH) correction (equation (2)), for both satellites and all three averaging intervals. For the Aqua data, the improvements after f(RH) correction were more pronounced than were those achieved by PBL height correction, although the inverse was seen for the Terra data. In general, the improvements achieved after both corrections (equation (4)) were more pronounced than those achieved by applying only one. Despite the improvements, the correlations remained weak, for both satellites and all three intervals. In addition, the results related to the Aqua data should be interpreted with caution, because more than 50% of those data (mainly PBL height data) were missing after the corrections. For both

Table 2
Linear correlations, by period/season, between PM concentrations and MAIAC AOD values, corrected as in equation 1 (ASH), as in equation (2) (AODdry), and as in equation (4) (ASHdry), for the satellites overpass.

	Period/season	Aqua[a]					Terra[b]				
PM		n	AOD	ASH	AODdry	ASHdry	n	AOD	ASH	AODdry	ASHdry
PM_{10}	2012–2017	1649	0.176	0.195	0.265	0.280	3480	−0.076	0.153	−0.007	−0.020
	Summer	103	0.319	0.317	0.346	0.277	623	−0.184	−0.142	−0.033	−0.021
	Fall	231	0.320	0.298	0.338	0.314	669	−0.032	0.188	0.001	0.234
	Winter	850	0.110	0.122	0.146	0.168	1087	−0.074	0.160	−0.025	0.171
	Spring	465	0.214	0.248	0.328	0.353	1101	0.014	0.176	0.033	0.099
$PM_{2.5}$	2012–2017	549	0.241	0.290	0.264	0.318	1014	0.034	0.064	0.236	0.025
	Summer	17	0.272	0.142	0.193	0.017	124	−0.093	0.161	−0.100	0.163
	Fall	43	0.267	0.275	0.292	0.295	117	0.073	0.159	0.318	0.399
	Winter	269	0.176	0.208	0.181	0.223	333	−0.005	0.027	0.225	0.196
	Spring	220	0.321	0.355	0.335	0.385	440	0.084	−0.019	0.230	−0.011

ASH, AOD corrected by PBL height; AOD_{dry}, AOD corrected by f(RH); ASH_{dry}, AOD corrected by PBL height and f(RH).
Summer, December–February; Fall, March–May; Winter, June–August; Spring, September–November.
[a] AOD corrected by the RML height estimated by the air temperature gradient method from 00:00 UTC balloon sounding data.
[b] AOD corrected by the PBL height estimated by the parcel method from 12:00 UTC balloon sounding data.

satellites and PMs, the seasonal correlations were typically stronger than were the annual correlations, although they were still weak, even after the corrections by PBL height and f(RH).

In an attempt to understand why the correlations were weak or negative in the MASP, Natali (2008) focused the investigation on diurnal PM_{10} data and found that the AODs, from the Terra and Aqua satellites, correlated more strongly (although still weak) with PM_{10} concentrations during the winter, when the sensor scattering angle was less than 140°, and on cloud-free days. In addition, for some stations, the author evaluated the AOD–PM_{10} relationship considering the quantity of water vapor content in the atmospheric column and concluded that high AOD values were associated with high water vapor content in the atmosphere. However, the author did not explore the effects of PBL height and f(RH), as was done in the present study.

In the United States, the poor correlations between $PM_{2.5}$ and AODs reported were justified as: the vertical distribution of the aerosols in the atmospheric column/aerosol transport at elevated levels (Toth et al., 2014; Paciorek et al., 2008), the variability in the aerosol type (Engel-Cox et al., 2006), surface reflectance (Engel-Cox et al. 2004, 2006; Paciorek et al., 2008; Zhang et al., 2009b) and the type of aerosol model used by MODIS in the AOD algorithm retrieval (Engel-Cox et al. 2004, 2006).

3.3. Evaluation of MODIS–MAIAC data over the MASP

The expected error (EE) envelope, defined as $\pm(0.05 + 0.05 \times \text{AOD})$, is used in order to evaluate the accuracy of MAIAC AOD retrievals, and it is expected that two thirds of the retrievals (approximately 66% or one standard deviation) fall within that envelope (Martins et al., 2017). The validation study of MAIAC data for South America showed that in urban area (São Paulo and Buenos Aires), 54.7% and 57.7% of the MAIAC Terra and Aqua AOD retrievals, respectively, fell within the EE envelope. Considering the MASP data, despite the relatively strong correlations between MAIAC AODs and AERONET AODs, we found that those percentages were 38% and 45%, for the Terra and Aqua retrievals, respectively (Fig. 2). Because MODIS AOD retrievals are based mainly on the aerosol model specification and surface reflectance estimation (Levy et al., 2007; de Almeida Castanho et al., 2008; Lyapustin et al., 2011b, 2018), these results indicate that one or both are inappropriate in the MAIAC AOD retrieval algorithm.

The intercept and slope of the linear regression between AOD retrieval and AERONET AOD can be used in order to evaluate the uncertainties related to the surface reflectance estimation and aerosol model specification in the AOD retrieval algorithm, respectively. An intercept close to 0 and a slope close to 1 indicate good assumptions about the surface reflectance and the aerosol model algorithm, respectively (Sayer et al., 2013). The evaluation of those quantities for MAIAC AOD retrievals over the MASP suggested the following: the surface reflectance is better estimated during Aqua overpasses than during Terra overpasses (intercepts of −0.0003 and 0.0190, respectively); and the aerosol model specified by the MAIAC AOD retrieval algorithm for Aqua and Terra overpasses might not be suited for use in the MASP (slopes of 0.67 and 0.52, respectively).

The MAIAC algorithm has improved the estimation of surface spectral ratios, a central component of aerosol retrievals. Unlike the MODIS DT algorithm, which is based on prescribed spectral reflectance ratios, the MAIAC algorithm dynamically derives surface spectral ratios using the time series approach (Lyapustin et al., 2018), thus avoiding empirical assumptions about surface properties (Martins et al., 2017). Therefore, MAIAC typically increases AOD accuracy over brighter surfaces, as demonstrated in studies conducted by Lyapustin et al. (2011b) over the United States and by Zhang et al. (2019) over China.

The MAIAC algorithm has eight background aerosol models (Lyapustin et al., 2018). The background aerosol model specified by the MAIAC AOD retrieval algorithm for South America is the same as that specified for the eastern United States. That model is based on the regional climatology data obtained at the Goddard Space Flight Center (GSFC) AERONET site, in Greenbelt, Maryland, which include the aerosol characteristics of the east coast of United States (Lyapustin et al., 2018).

In Fig. 3, we present the data obtained at the GSFC and São Paulo AERONET sites in the 2000–2011 period, using the single scattering albedo (SSA) and fine-mode fraction (FMF) data to classify the aerosols by type, as proposed by Lee et al. (2010). The SSA is defined as the ratio of the scattering coefficient to the extinction coefficient of aerosols and indicates the relative capacity of the aerosols to absorb radiation at a given wavelength, a higher SSA translates to lower absorption of solar radiation (Lee et al., 2010). The FMF is defined as the ratio of fine-mode AOD (of particles with a diameter < 1 μm) to the total AOD and indicates the predominant size (fine or coarse) of the aerosols under study. According to the classification proposed by Lee et al. (2010), fine-mode aerosols are those with an FMF above 0.6 and coarse-mode aerosols are those with an FMF below 0.4. Aerosols with an FMF between 0.4 and 0.6 are designated aerosol mixtures.

As depicted in Fig. 3, the São Paulo and GSFC AERONET sites are both dominated by fine-mode aerosols. However, the aerosols evaluated at the GSFC site present a relatively homogeneous SSA, whereas those evaluated at the São Paulo site presents high variability in the degree of absorption of solar radiation. These opposing patterns were described by Levy et al. (2007), who showed that the eastern United States was dominated by non-absorbing aerosols, whereas South America (including the MASP) was dominated by aerosols that are more absorptive. Despite the differences between the two sites in terms of the aerosol properties, the aerosol model specified by the MAIAC AOD retrieval algorithm is the same for both regions. That indicates that there is a mismatch between the current MAIAC aerosol model and the local aerosol properties observed across the MASP.

The MODIS AOD products are also retrieved using the DT algorithm, at spatial resolutions of 10 × 10 km^2 and 3 × 3 km^2 (Levy et al., 2013), the Deep Blue algorithm (Hsu et al., 2013) and the algorithm that

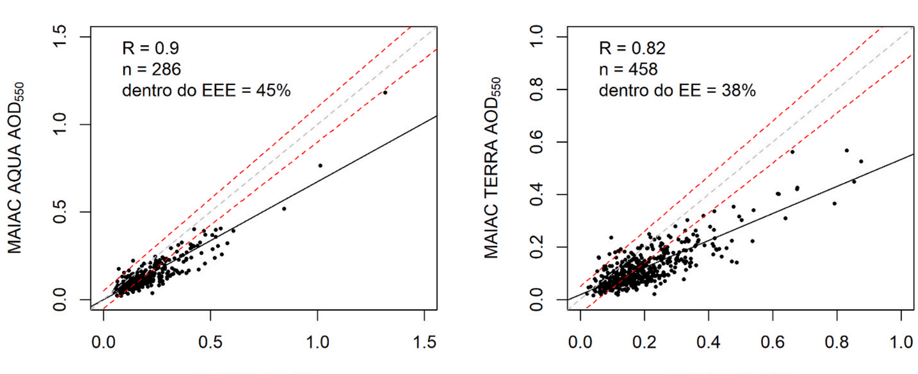

Fig. 2. Comparison between the AERONET data for AOD at 550 nm (AOD_{550}) and those derived from the MAIAC algorithm for the Aqua satellite (left) and Terra satellite (right) in the MASP. The red and grey dashed lines indicate the MAIAC EE envelope, defined as $\pm(0.05 + 0.05 \times \text{AOD})$ and 1:1 lines, respectively. The solid line displays the regression line. R is the linear correlation between MAIAC AODs and AERONET AODs, n is the number of data pairs (MAIAC AODs; AERONET AODs), and within EE is the fraction of MAIAC retrievals that fell within the EE envelope. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

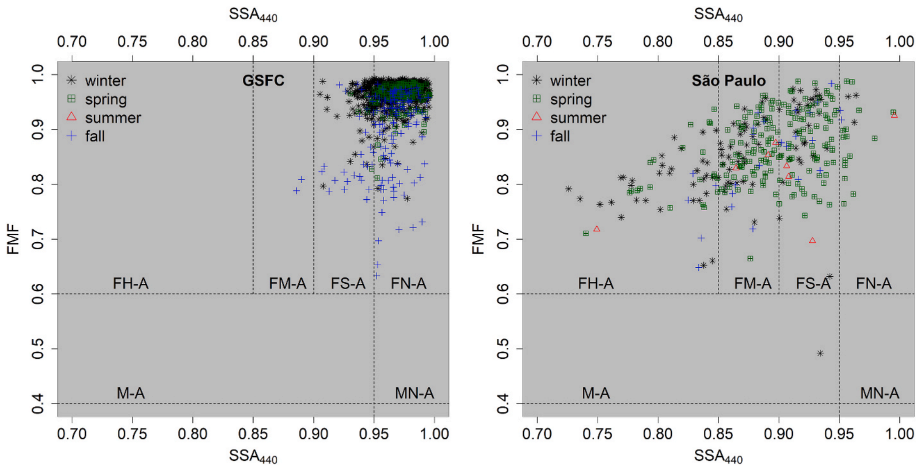

Fig. 3. The aerosol classification proposed by Lee et al. (2010), showing the dispersion of the SSA at 440 nm (SSA_{440}) and FMF, by period (season), for the AERONET sites at the GSFC (left panel) and in São Paulo (right panel), in the 2000–2011 period. FH-A, fine highly absorbing; FM-A: fine moderately absorbing; FS-A: fine slightly absorbing; FN-A: fine non-absorbing; M–A: mixed absorbing; MN-A: mixed non-absorbing; winter, June–July–August; spring, September–October–November; summer, December–January–February; fall, March–April–May.

combines the two (Levy et al., 2013), both at a spatial resolution of 10 × 10 km^2. The coarse spatial resolutions of the MODIS AOD products have been an important limitation for their application in air quality and epidemiological studies at urban areas such as the MASP and could explain the lack of studies applying AOD data in this region.

A comprehensive comparison of MODIS AOD products over the MASP was beyond the scope of the present study. However, to increase our confidence in the conclusions drawn, we compared the AOD retrievals from the DT algorithm at 3 × 3 km^2 (collection 6.1) with those from the AERONET site. We also calculated the validation measures—the *R* and the fraction of retrievals that fell within the EE envelope, as well as the slope and intercept of the regression line between DT at 3 × 3 km^2 AODs and AERONET AODs—and compared them with those obtained for the MAIAC. Although the DT at 3 × 3 km^2 AOD product is not suited for epidemiological applications in a relatively small region such as the MASP, we chose it for the comparison because it is the product for which the spatial resolution is closest to that of the MAIAC AOD product. In the comparison between the DT at 3 × 3 km^2 AODs and the AERONET AODs, we used data for the 2012–2017 period, applying the same spatial and temporal windows used in the validation study of MAIAC data for South America (25 × 25 km^2 and 1 h, respectively). There is a need for a comprehensive validation study of the MODIS DT at 3 × 3 km^2 AOD product over the MASP.

The correlations between AOD retrievals from the AERONET site and from the DT at 3 × 3 km^2 algorithm were strong for the Terra and Aqua retrievals (0.83 and 0.88, respectively) and were comparable to those obtained for the MAIAC retrievals. For the DT at 3 × 3 km^2 algorithm, the EE envelope was defined as $\pm$ (0.05 + 0.2 × AOD), as described by Levy et al. (2013); 86.6% and 87.0% of the Terra and Aqua retrievals, respectively, fell within that envelope. The intercept (slope) of the regression lines between AERONET AODs and DT at 3 × 3 km^2 AODs were 1.0049 (0.0174) and 1.048 (0.027) for the Terra and Aqua retrievals, respectively. As expected, the DT at 3 × 3 km^2 algorithm overestimated the surface reflectance at the MASP, because it is more suitable for use over dark surfaces. The results suggest that, for surface reflectance estimation, the MAIAC algorithm performs better than does the DT at 3 × 3 km^2 algorithm for the Aqua retrievals and that the two algorithms have a similar performance for the Terra retrievals. For aerosol model specification, the DT at 3 × 3 km^2 algorithm appears to perform better than does the MAIAC algorithm for retrievals from both satellites, because the former assumes a more absorbing aerosol optical model, which is more suitable for use over the MASP (Levy et al., 2013). Nevertheless, there are opportunities for improvements in the aerosol models for both algorithms over this region.

The atmosphere over the MASP contains a wide variety of aerosol types. Local aerosols are composed mainly of vehicle and industrial emissions. During the fire season (August to September), the MASP is also affected by the long-range transport of smoke from biomass burning in the countryside of the state of São Paulo, in the northern/central-west regions of Brazil, and in neighboring countries, including Bolívia, Paraguay, and Argentina (Mariano et al., 2010; de Miranda et al., 2017; Yamasoe et al., 2017; Vara-Vela et al., 2018). The studies conducted by Martins et al. (2009) and Yamasoe et al. (2017) showed that the local aerosols are more absorptive than are those transported from areas of biomass burning. These results explain the high AOD variability, mainly in the spring (Fig. 3). Furthermore, the MASP is affected by sea-breeze circulation (Freitas et al., 2007), an additional aerosol source that also contributes to the high aerosol variability in the MASP. All of those aerosol sources increase the variability in aerosol properties, making AOD retrieval more complex (Engel-Cox et al., 2006; Lee et al., 2016).

The RH effect on the AOD–PM relationship can also be related to the aerosol model specification in the MAIAC AOD retrieval algorithm, because RH can change the chemical and extinction characteristics of aerosols (personal communication with Alexei Lyapustin, 2019). According to the fraction of hygroscopic components present in the aerosol, the increase in RH can enhance the aerosol extinction, due to the increase in the scattering efficiency (Liu et al., 2013). Since sulphates and nitrates, which are highly hygroscopic, are the main species in the aerosol in São Paulo (Pereira et al., 2017) and in the Eastern United States (Bell et al., 2007), it is possible that an increase in the RH change the SSA of aerosols in the MASP, and make it comparable to that of aerosols in the eastern United States. That would explain why MAIAC uses the same aerosol model for both locations, despite the differences in the SSA properties shown in Fig. 3.

To elucidate that, Fig. 4 presents the mean SSA for each bin of FMF and Total Precipitable Water (TPW) at the GSFC and São Paulo AERONET sites. Changes in TPW are caused in part by RH and in part by seasonal variation, because TPW is a function of temperature and RH (Lee et al., 2010). The increase caused by a change in SSA for TPW was lower in São Paulo than at GSFC. Therefore, even if the RH effect is taken into account, the aerosols in São Paulo appear to be more absorptive than those in the eastern United States, highlighting the mismatch between the current MAIAC aerosol model and the aerosol properties observed across the MASP.

Although in both locations sulphates and nitrates are the dominant species, which are highly reflective of solar radiation (Wallace and Hobs, 2006), and explain the high SSA in both locations, the relative amount of elemental carbon (EC)/black carbon (BC) in the $PM_{2.5}$ mass of aerosol in São Paulo is at least four times higher than that in the Eastern US. The study conducted by Pereira et al. (2017) in São Paulo, reported an EC contribution of about 16% to the $PM_{2.5}$ mass, while the contribution of the EC to the $PM_{2.5}$ mass in the Eastern US reported by Bell et al. (2007) was about 4%. The studies conducted by de Almeida Castanho and Artaxo (2001) and Andrade et al. (2012b) in São Paulo reported a BC

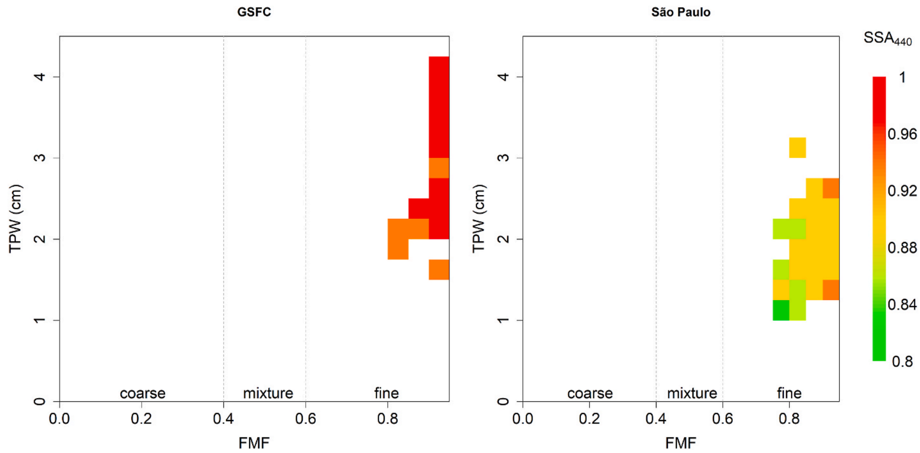

Fig. 4. Mean SSA at 440 nm (SSA_{440}) for each bin of FMF and TPW for the AERONET sites at the GSFC (left panel) and in São Paulo (right panel). Only bins with more than five data points are shown.

contribution of about 25% and 36% to the $PM_{2.5}$ mass, respectively. On the other hand, the study by de Almeida Castanho et al. (2005) reported a BC contribution of 3% of the $PM_{2.5}$ mass on the East Coast of the United States. Since EC/BC are, in general, strong absorbing, SSA in São Paulo is lower than the aerosol in the Eastern US.

In previous versions of the MODIS AOD retrieval algorithms, aerosol models were updated to improve the representativeness of the physical and optical properties of local aerosols. In MODIS collection 5, a seasonal (winter and spring) absorbing aerosol model with an SSA of approximately 0.85 was assigned to the region comprising the south-central and southeastern regions of Brazil, whereas a moderately absorbing aerosol model (SSA of approximately 0.90) was assigned to the remaining regions and for the summer and fall (Levy et al., 2007, Fig. 3). The MAIAC AOD is a relatively new product for which updates are underway. In the upcoming version of the MAIAC algorithm, retrievals will take into account the effects of regional aerosol seasonality, such as those caused by variations in humidity or by periods of biomass burning (Lyapustin et al., 2018). Until then, the MAIAC AOD data should not be used as a predictor of ground-level PM concentrations in the MASP.

The results of the present study indicate that MAIAC AOD retrievals over this region are affected mainly by the mismatch between the current MAIAC aerosol model for the MASP and the local aerosol properties. Compared with the DT algorithm at 3×3 km^2 (collection 6.1), the MAIAC surface reflectance estimation is more accurate for Aqua retrievals than for Terra retrievals.

4. Conclusions

This study examined the high-resolution AODs derived from the MAIAC algorithm as a predictor of ground-level PM concentrations at the MASP. We calculated the correlations between ground-level PM concentrations (at the hour of the satellites overpass, diurnal averaging, and daily averaging intervals) and the collocated AODs. We found those correlations to be weak. Our findings corroborate those of the study conducted by Natali (2008). Although the correlations were slightly improved after corrections by PBL height and f(RH), they remained weak. Our results indicate that the mismatch between the current aerosol model specified by the MAIAC algorithm for the MASP and the physical and optical properties of the local aerosols is the main issue affecting MAIAC AOD retrievals. Therefore, high-resolution AODs from the current MAIAC version will not serve well to predict ground-level PM concentrations in the MASP.

The MAIAC algorithm would greatly benefit from an improved aerosol model applied to MASP. Our results suggest that sources of aerosol particles in the MASP are quite diverse and that there is no single optical model suitable for use with satellite-derived AODs in the region. In this context, de Almeida Castanho et al. (2008) presented a tool that, using critical reflectance, dynamically assigns an SSA value to the aerosol model that reflects the optical properties of the aerosol layer in the MASP. For the spectral reflectance ratio, those authors used the prescribed ratio of 0.6. Therefore, we recommend that future studies employ an algorithm combining the dynamic specification of surface spectral ratios from the MAIAC algorithm and the dynamic specification of SSA in the aerosol model proposed by de Almeida Castanho et al., (2008). We believe that such a combination would improve AOD retrievals over the MASP, because it will combine the improvements of the MAIAC algorithm over bright surfaces and the correct specification of the aerosol model, taking advantage of its higher spatial resolution.

As seen in applications in other regions, high resolution MAIAC AOD retrievals could be assimilated into statistical (e.g., Chudnovsky et al., 2014b), geophysical (e.g., Jin et al., 2019) or a combination of both approaches (e.g., Meng et al., 2019), in order to retrospectively predict PM concentrations, resulting in PM databases for future use in health studies in the MASP.

CRediT authorship contribution statement

Aline Santos Damascena: Conceptualization, Software, Formal analysis, Investigation, Writing - original draft. **Márcia Akemi Yamasoe:** Conceptualization, Supervision, Writing - review & editing. **Vitor Souza Martins:** Investigation, Software, Validation. **Jorge Rosas:** Investigation, Software. **Noelia Rojas Benavente:** Investigation, Software. **Maciel Piñero Sánchez:** Investigation, Software. **Nelson Ithiro Tanaka:** Conceptualization, Supervision. **Paulo Hilário Nascimento Saldiva:** Conceptualization, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2020.117949.

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