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Original Research

Urban particulate matter reaching Atlantic Rainforest remnants near Brazilian metropolis: an alert for environmental protection authorities

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Abstract

The increasing air pollutant emission, mainly in big cities, attracts significant attention to environmental sciences. Brazil boasts an important tropical forest, the Atlantic Rainforest, and the most important remnants areas are located in the state of Rio de Janeiro. In that regard, this study aimed to assess the impact of particulate matter (PM) emitted by traffic in ecosystems belonging to environmental protection areas (EPAs) near important highways with heavy traffic in southeastern Rio de Janeiro, Brazil. To the best of our knowledge, this is the most comprehensive study on air pollution and its impacts on EPAs in Brazil. PM concentrations (TSP, PM_{10}, and $PM_{2.5}$) from 14 air quality monitoring stations and meteorological parameters were obtained between 2014 and 2016 near EPAs. It was verified that CONAMA annual standard (Brazilian legislation) was overtake in five monitoring stations and in eight of them CONAMA daily standard was exceed. Wind direction was mainly from urban centers to EPAs, indicating that urban pollutants reach forest remnants in most cases, which may represent ecological risk. In order to guarantee environmental preservation, new studies should be performed to evaluate deeply the effect of air pollutants on fauna and flora of preserved areas.

Keywords: Air quality, urban pollution, environmental sustainability, forest preservation.

INTRODUCTION

Atmospheric pollution is a highlighted theme due to increasing emissions involving both organic and inorganic substances and particulate matter (PM) (Rai 2016a; Al-Thani et al. 2018; Xie et al. 2018; Swislowski *et al.* 2020). PM comprises an important class of pollutants that may affect both human health and ecosystems, while also influencing local or even global climate (Agnan *et al*. 2017; Chaligava *et al*. 2020; Beringui *et al.* 2021). Major air pollutant sources are located in urban environments and include industrial and vehicle emissions (Galal and Shehata 2015; Beringui *et al*. 2021).

Total Suspended Particles (TSP) comprise particles with an aerodynamic diameter of less than 100 µm, classified according to their aerodynamic diameter as PM_{10} (smaller than 10 µm), or $PM_{2.5}$ (smaller than 2.5 µm) (Gioda *et al.* 2021). Although TSP is not a threat to human health, it can pose risks to the environment through incorporation of its components into the soil or deposited on flora surfaces (Datta *et al*. 2016; Rai 2016b; Mateus *et al*. 2020), leading to deleterious effects to plants (Jaconis *et al.* 2017). PM fine fraction, in addition to health risks, also poses environmental risks, mainly due to long distances transport (Li *et al.* 2020). In addition to size, the chemical composition of PM can vary from one

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area to another, as it depends on meteorology, topography, and characteristics of emission sources. (Gioda *et al*. 2011; Mkoma *et al*. 2014; Ventura *et al.* 2018; Beringui *et al.* 2021).

Trees are directly associated with air quality, as they act as particle filters and emitters (Terzaghi *et al*. 2013; Chen et al. 2016; Aponte-aponte *et al.* 2018). The ability of plants to filter particles make them useful tools concerning air pollution mitigation in urban environments (Dzierzanowski *et al*. 2011; Agnan *et al*. 2017; Qiu *et al*. 2018; He *et al.* 2020; Swislowski *et al*. 2020). Although several studies have sought to assess PM impacts on human health, few reports ecosystem effects. In most cases, green areas or forests are considered pollutant reducers, and vegetation receives attention, mainly, due to the provision of ecosystem services (Terzaghi *et al.* 2013; Chen *et al*. 2016; Gómez-Moreno *et al.* 2019). However, considering a sustainability scenario, the impact of atmospheric pollution on vegetation must be considered, since damage caused to plant and tree species can compromise ecosystem biodiversity, since the particles deposited on the plants can cause a nutritional imbalance, affecting epiphytic species, and also altering the decomposition process by fungi. In addition, the physiological processes affected by the particles can reduce the development of the species, causing an ecological imbalance. (Rai 2016b; Koch *et al.* 2018; Molnár *et al.* 2020). Futhermore, pollutants incorporated into the soil can also alter biogeochemical cycles of metals, since atmospheric emissions contribute significant amounts of metals, metals can accumulate in the soil or in plants. (Prajapati 2012; Rai 2016b; Shahid *et al.* 2017; Kosior *et al.* 2020).

The Atlantic Forest, one of the most important tropical forests in the world, is an integral part of the climate regulation and water cycle of part of the Brazilian territory (Silva *et al.* 2010). The development of cities and exploitation of natural resources has devastated this biome since the colonization of Brazil (Silva *et al.* 2010; Morellato *et al.* 2014). Aiming to preserve the Atlantic Forest remnants, which is home to more than 20,000 plant species and more than 2,000 animal species, Brazilian legislation established the National System of Nature Conservation Units (in Portuguese, *Sistema Nacional de Unidades de Conservação da Natureza* – SNUC), with rules for the creation and management of conservation units (Law 9985 18/06/2000). Environmental Protection Areas (EPAs) currently suffer from several conflicting activities, such as the proximity of highways, subway lines and urban centers, as well as the presence of pipelines and the growing demand for water supply (IEF-RJ 2006; MMA/IBAMA 2006; MMA/ICMBio 2008a; MMA/ICMBio 2008b; INEA 2015).

In this context, this study aims to carry out a broad assessment of PM concentrations near ten Brazilian Environmental Protection Areas located in the state of Rio de Janeiro, southeastern Brazil, to verify the potential influence of urban pollution on vegetation. Different sites in the coastal region, urban-industrial region, and mountain range were selected to evaluate PM levels and the influence of meteorological parameters on pollutants transport. Such studies are necessary to signal to the authorities the need for changes in the environmental legislation or intensification of the inspection of the emission sources.

MATERIAL AND METHODS

Study area

The state of Rio de Janeiro boasts of 36 Environmental Protection Areas (EPAs), established with the purpose of preserving biodiversity and native ecosystems (INEA, 2020). These conservation units are managed by the Environmental Institute of Rio de Janeiro State (in Portuguese, *Instituto Estadual do Ambiente – INEA*) and by the Federal Government through Chico Mendes Institute (in Portuguese, *Instituto Chico Mendes – ICMBio*).

The ten investigated EPAs comprised the following: six units under federal government administration: 1) Itatiaia National Park (PARNA Itatiaia), located in Itatiaia; 2) Serra dos Orgãos National Park (PARNA Serra dos Orgãos), in the cities of Teresópolis, Petrópolis, Magé and Guapimirim; 3) Jurubatiba Restinga National Park (PARNA Jurubatiba), in Macaé, 4) Tijuca National Park (PARNA Tijuca), in the city of Rio de Janeiro; 5) Tinguá Biological Reserve (Tinguá Rebio), in the cities of Duque de Caxias, Nova Iguaçu and Belford Roxo, and 6) Guanabara Ecological Station (ESECN Ecológica da Guanabara), in the cities of Guapimirim, Itaboraí and São Gonçalo; three units managed by the state government: 7) Chacrinha State Park, a fully protected area in Copacabana, a coastal Rio de Janeiro city neighborhood; 8) Environmental Protection Area of Mangaratiba(APA Mangaratiba) and 9) Chunhambebe State Park, both in Mangaratiba, a southern coastal city in the Rio de Janeiro state; and one unit managed by city hall of Rio de Janeiro city: 10) Darke de Mattos Municipal Natural Park, in the island Paquetá. The sampling sites descriptions are presented in Table 1 and displayed in Figure 1. The list of the EPAs close to each sampling site is presented in Table 2.

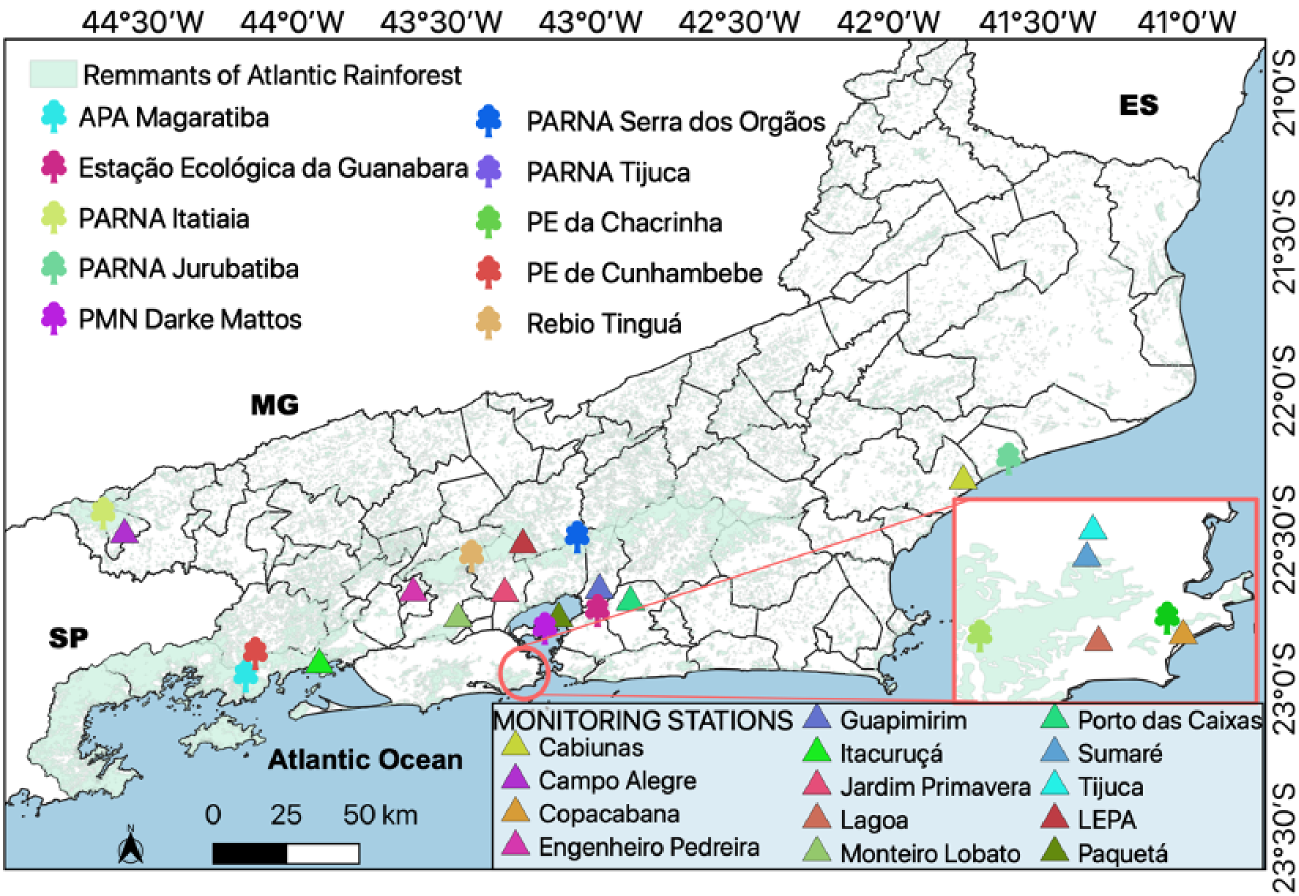

Figure 1 Map indicating the location of sampling sites and Environmental Protection Areas (EPAs) located in the state of Rio de Janeiro, Brazil

Table 1 Particulate matter sampling sites characteristics

Station	**Surrounding characteristics**	**Samples**	**Altitude**
Campo Alegre - CA	Located on the highway that connects the city of Itatiaia to the entrance of PARNA Itatiaia	TSP and PM_{10}	425 m
Cabiunas - Cb	A district of Macaé, in the north of Rio de Janeiro, where a terminal for the transportation of petroleum products and an industrial complex are installed	TSP and PM_{10}	11 m
Lagoa - Lg	A residential neighborhood in the southern zone of the city of Rio de Janeiro, with heavy vehicular traffic in the early morning and late afternoon.	$PM_{2.5}$	5 m
Sumaré - Sum	A 700-meter-high hill located in the northern part of the city of Rio de Janeiro. Radio and television broadcast antennas are installed on it, and access is allowed only to authorized people.	TSP and PM_{10}	500 m
Tijuca - Tij	Residential and commercial neighborhood located in the northern part of the city of Rio de Janeiro. Vehicular traffic is intense throughout the day. The proximity to the Tijuca massif hinders the dispersion of pollutants.	TSP	15 m

Paquetá - Pqt	An island located in Guanabara Bay, which can be reached by boat. It is a neighborhood of Rio de Janeiro with population little over 3,000 inhabitants.	PM_{10}	2 m
Monteiro Lobato - ML	A residential neighborhood in Nova Iguaçu, a municipality in the metropolitan region of Rio de Janeiro. It is located near important highways that connect to neighboring municipalities and the train line that connects to downtown Rio de Janeiro.	PM_{10}	22 m
Jardim Primavera - JP	Located on federal highway BR-040 in the region belonging to the municipality of Duque de Caxias. It is located in the area where BR-040 meets the Arco Metropolitano, a highway that connects the five main highways in the state of Rio de Janeiro	PM_{10} and $PM_{2.5}$	10 m
Engenheiro Pedreira - EP	The main district of Japeri, a municipality in the metropolitan region of Rio de Janeiro located near the Serra do Mar. It is a residential and commercial region where the train line connecting to downtown Rio de Janeiro passes.	PM_{10}	30 m
Copacabana - Copa	Residential coastal neighborhood of Rio de Janeiro. It is characterized by the presence of commercial enterprises and intense vehicular traffic throughout the day.	$PM_{2.5}$	10 m
Guapimirim - Guapi	Environmental protection area located on the shores of Guanabara Bay. It is in the area of influence of a petrochemical complex located in the neighboring city of Itaboraí.	PM_{10}	16 m
Porto das Caixas - PC	Residential neighborhood of Itaboraí, situated between Guanabara Bay and a petrochemical complex	TSP and PM_{10}	19 m
Itacuruçá - Itc	Coastal neighborhood of Mangaratiba, in the south of the state. It is located in the area of influence of the port of Itaguaí. It is characterized by the traffic of boats that make connections with several islands in the region.	TSP and PM_{10}	1 m
Serra de Petrópolis	Federal highway that connects Rio de Janeiro to Minas Gerais. This highway is used to transport products in and out of the state. Sampling sites in the region of Petrópolis and Duque de Caxias.	TSP	25 – 800 m

Table 2 Proximity of air quality monitoring stations to environmental protection areas

EPA	Sampling site	Distance between station and EPA
TSP		
PARNA Itatiaia	Campo Alegre - CA	2 km
PARNA Restinga Jurubatiba	Cabiunas - Cb	2 km
PARNA Tijuca	Sumaré - Sum	1 km
PARNA Tijuca	Tijuca - Tij	2 km
ESECN Ecológica da Guanabara	Porto das Caixas - PC	9 km
APA Mangaratiba	Itacuruçá - Itc	8 km
Parque Estadual de Cunhambebe		8 km
PARNA Serra dos Orgãos	Serra de Petrópolis (Serra)	5 – 13 km
PM_{10}		
PARNA Itatiaia	Campo Alegre - CA	2 km
PARNA Restinga Jurubatiba	Cabiunas - Cb	2 km
PARNA Tijuca	Sumaré - Sum	1 km
ESECN Ecológica da Guanabara	Porto das Caixas - PC	9 km
APA Mangaratiba	Itacuruçá - Itc	8 km
Parque Estadual de Cunhambebe		8 km

Parque Natural Municipal Durke de Matos	Paquetá - Pqt	0 km
Tinguá Rebio	Monteiro Lobato - ML	10 km
Tinguá Rebio	Jardim Primavera - JP	9 km
Tinguá Rebio	Engenheiro Pedreira - EP	2 km
ESECN Ecológica da Guanabara	Guapimirim - Guapi	3.5 km
$PM_{2,5}$		
PARNA Tijuca	Lagoa - Lg	3 km
Tinguá Rebio	Jardim Primavera - JP	9 km
Parque Estadual do Chacrinha	Copacabana - Copa	0.5 km

PM sampling

Particulate matter was collected according to Brazilian standard methods (ABNT – NBR 9547/86 for TSP and 13412/95 for PM_{10} and $PM_{2.5}$), in agreement with the United States Environmental Protection Agency (US EPA) methods (Method IO – 2.1), using Hi-Vol samplers and fiberglass filters every six-day. Automatic and semi-automatic monitoring stations were used for PM sampling. Most of them make up the monitoring network of INEA, however, Atmospheric Pollution Studies Laboratory – LEPA (in Portuguese, *Laboratório de Estudos da Poluição Atmosférica* – LEPA) performed sampling in two locations: Serra and Paquetá.

The majority of monitoring stations belongs to INEA monitoring system. A total of 282 samples were obtained in 2014 (126 $PM_{2.5}$, 60 PM_{10}, and 96 TSP), 356 in 2015 (107 $PM_{2.5}$, 108 PM_{10}, and 141 TSP), and 329 in 2016 (125 $PM_{2.5}$, 92 PM_{10}, and 112 TSP) at semiautomatic monitoring stations. Hourly data obtained through automatic stations from private companies provided by INEA through environmental licensing activities were also used. A total of 58,663 measurements were obtained in 2014, 71,784 in 2015, and 71,197 in 2016.

The sampling in Serra de Petrópolis was conducted by LEPA. Hi-Vol samplers were placed at the top of local houses and TSP sampling was conducted for 24 h (on a weekly basis) on fiberglass filters at an average flow rate of 1.15 m^3 min^{-1} every six days. Twelve Hi-Vol samplers were installed over 20 km of highway that borders the EPA. A total of 2,117 TSP samples were collected between 2014 and 2016. They will be considered in this study as one monitoring station (Serra de Petrópolis) which represents the average concentration of the twelve. For PM sampling in Paquetá, LEPA used an automated monitoring station, which totaled 10.042 measurements between 2014 and 2016.

The semi-automatic monitoring stations operate following the PM sampling protocols (ABNT-NBR 9547 and ABNT-NBR 13412). Filters were weighed three times prior to and three times after the sampling efforts to determine PM mass. During weighing, relative humidities were from 20 to 30 % and temperatures from 20 to 25 ºC. PM concentrations were determined by dividing the PM mass by the sampled air volume.

Meteorological data

Some air quality monitoring stations from INEA network (Jardim Primavera, Monteiro Lobato, Porto das Caixas, Engenheiro Pedreira and Cabiunas) also provide meteorological data. In order to assess PM transport, the wind speed (WS) and wind direction (WD) data obtained at these stations were used to verify the origin of the air masses to predict the possible sources of PM and whether the particles could reach the EPAs. Rainfall data were obtained only for the stations Porto das Caixas, Paquetá, Guapimirim and Cabiunas. These data are available online at INEA's website (INEA)

The R Software (RCoreTeam 2020) was used to construct wind roses with those data, employing Openair package. For this, WS < 0.5 was considered as calm (WS = 0). The timePlot function from the openair (Carslaw, 2012) package was used to plot the graphs with the daily precipitation sums obtained in each year evaluated.

RESULTS

Particulate matter concentration

The annual average PM concentrations for the three years and the standard deviations are presented in Figure 2. During the monitoring period the Brazilian legislation established two air quality standards. The primary standard indicates concentration thresholds above which human health damage may occur. The secondary standard corresponds to concentrations below which the population, fauna, flora, material, and the general environment are expected to suffer minimal damage.

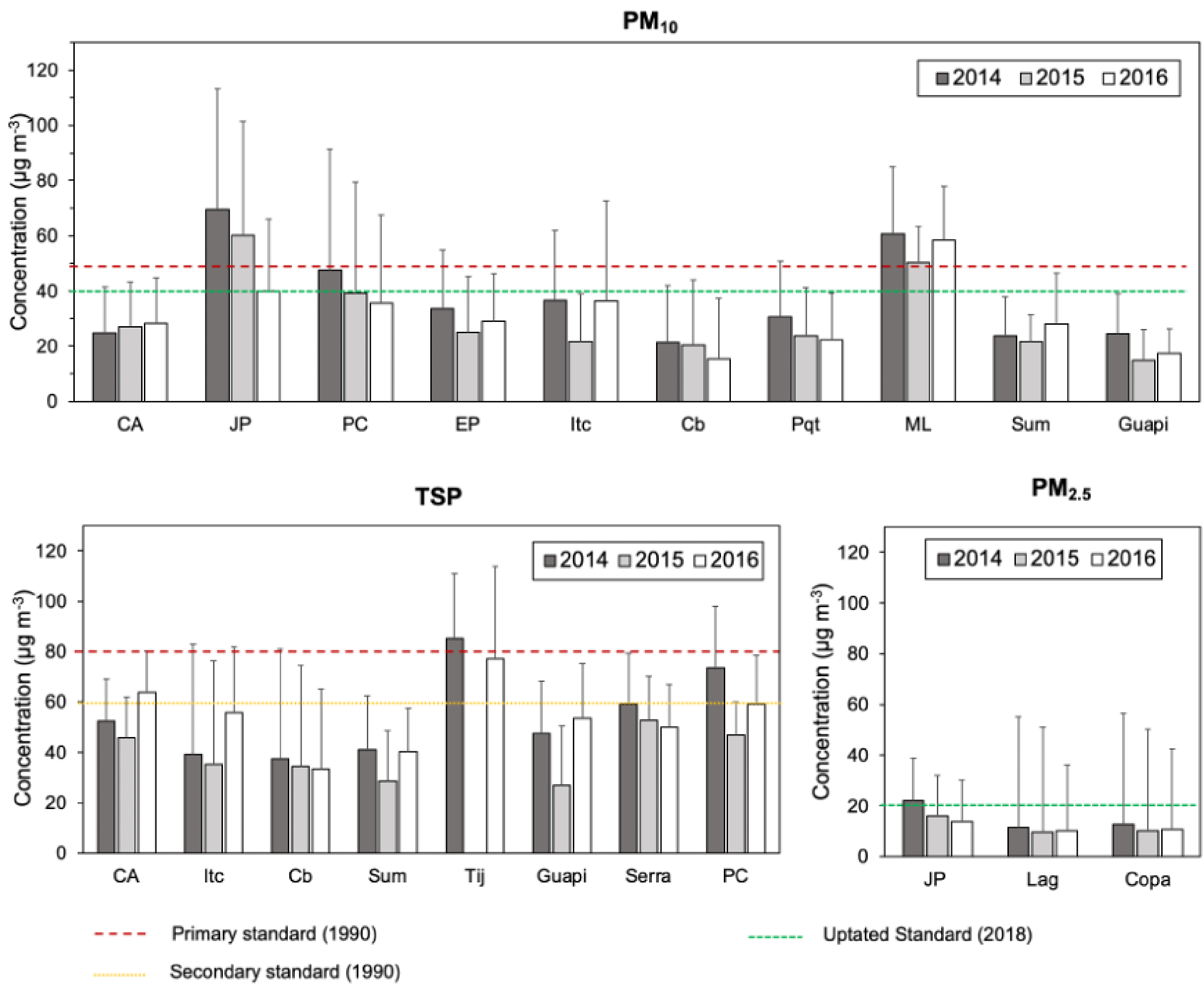

Figure 2 Annual average particulate matter concentrations in the sampling sites located near environmental protection areas. Dashed lines indicate CONAMA limits during the sampling period: 80 $\mu g\ m^{-3}$ (primary standard) and 60 µg m^{-3} (secondary standard) for TSP, and 60 µg m^{-3} for PM_{10}, and the CONAMA limits established in 2018: 40 $\mu g\ m^{-3}$ for PM_{10} and 20 $\mu g\ m^{-3}$ for $PM_{2.5}$.

According to CONAMA limits, the primary standard concerning the annual geometric average of TSP (80 µg m^{-3}) was exceeded only at Tijuca (Tij) in 2014, while the secondary was exceeded at Tijuca in 2014 and 2016 and at Porto das Caixas (PC) in 2014. During the sampling period (2014-2016), the CONAMA limit for PM_{10} was 50 µg m^{-3} and no limit was set for $PM_{2.5}$. The PM_{10} annual average limit was exceeded at Monteiro Lobato (2014, 2015 and 2016), Jardim Primavera (2014 and 2015) and Porto das Caixas (2014). In 2018, Brazil's air quality standards were revised, new PM_{10} limits were established, and air quality standards for $PM_{2.5}$ were implemented. Considering these new values for annual average standards (40 µg m^{-3} for PM_{10} and 20 µg m^{-3} for $PM_{2.5}$), the mean PM_{10} concentrations detected at Jardim Primavera and Monteiro Lobato exceeded the annual limit during the three study years, as well as Porto das Caixas in 2014. Regarding $PM_{2.5}$, Jardim Primavera exceeded the CONAMA limit only in 2014.

Figure 3 presents daily PM concentration and Table 3 presents descriptive statistics and the number of violations of CONAMA daily limits. All monitoring stations presented a huge variation on daily concentration for three particle sizes, thus, the data range was large. However, the number of CONAMA limit violations was low. Violations of PM_{10} limits were recorded at Jardim Primavera, Porto das Caixas and Engenheiro Pedereira, however, the overtaken occurred less than five times per year. Although no air quality standard for $PM_{2.5}$ was in effect during the sampling period, the standard established in 2018 (20 µg m^{-3}) was overtook 12 times in Copacabana in 2015. The highest number of violations was verified at Itacuruçá, where the TSP limit was overtaken 16 times in 2016. This limit was also exceeded in Campo Alegre in 2015.

Table 3 Descriptive statistics and number of violations of daily CONAMA limits for all monitoring stations near EPAs.

Site	2014			2015			2016		
	Range	**Mean ± sd**	**Viol.**	**Range**	**Mean ± sd**	**Viol.**	**Range**	**Mean ± sd**	**Viol.**
TSP									
CA	14 - 176	53 ± 22	-	11 - 134	47 ± 16	1	22 - 134	63 ± 18	2
PC*	63 - 82	74 ± 10	-	4 - 179	48 ± 33	-	11 - 138	59 ± 36	-
Itc	4 - 116	37 ± 18	-	4 - 97	36 ± 16	-	7 - 215	55 ± 30	16
Cb	7 - 116	37 ± 18	-	4 - 130	35 ± 18	-	0.92 - 99	33 ± 19	1
Sum*	10 - 115	41 ± 25	-	12 - 65	31 ± 12	-	17 - 140	40 ± 25	-
Tij*	34 - 168	85 ± 32	-	-	-	-	32 - 143	77 ± 24	-
Guapi*	22 - 64	41 ± 14	-	8 - 69	28 ± 15	-	11 - 138	53 ± 57	-
Serra*	22 - 158	67 ± 25	-	30 - 131	61 ± 18	-	32 - 106	56 ± 16	-
PM_{10}									
CA	5 - 92	25 ± 12	-	5 - 74	27 ± 11	-	6 - 78	28 ± 12	-
JP	14 - 153	69 ± 27	2	11 - 158	61 ± 27	-	7 - 113	40 ± 18	-
PC	7 - 155	49 ± 28	1	6 - 148	40 ± 25	-	6 - 124	36 ± 19	1
EP	9 - 99	34 ± 14	-	4 - 83	26 ± 13	3	5 - 56	29 ± 12	-
Itc	5 - 115	35 ± 17	-	3 - 64	22 ± 11	-	4 - 107	36 ± 18	-
Cb	3 - 74	21 ± 12	-	0.04 - 116	21 ± 14	-	0.04 - 75	15 ± 13	-
Pqt	11 - 62	30 ± 11	-	5 - 90	25 ± 15	-	3 - 96	23 ± 11	-
ML*	17 - 113	60 ± 25	-	23 - 79	51 ± 13	-	8 - 113	50 ± 19	-
Sum*	1 - 65	24 ± 14	-	11 - 55	23 ± 11	-	11 - 99	28 ± 18	-
Guapi*	15 - 36	23 ± 7	-	0.46 -62	16 ± 12	-	3 - 42	17 ± 9	-
$PM_{2.5}$									
JP*	5 - 50	22 ± 10	-	2 - 35	16 ± 8	-	3 - 35	14 ± 8	-
Lg*	3 - 28	12 ± 7	-	1 - 32	10 ± 6	-	1 - 28	10 ± 5	-
Copa*	3 - 28	13 ± 7	-	1 - 24	12 ± 7	12	3 - 29	11 ± 8	-

*Semiautomatic stations, which sampling occurs during 24 h every six-day.

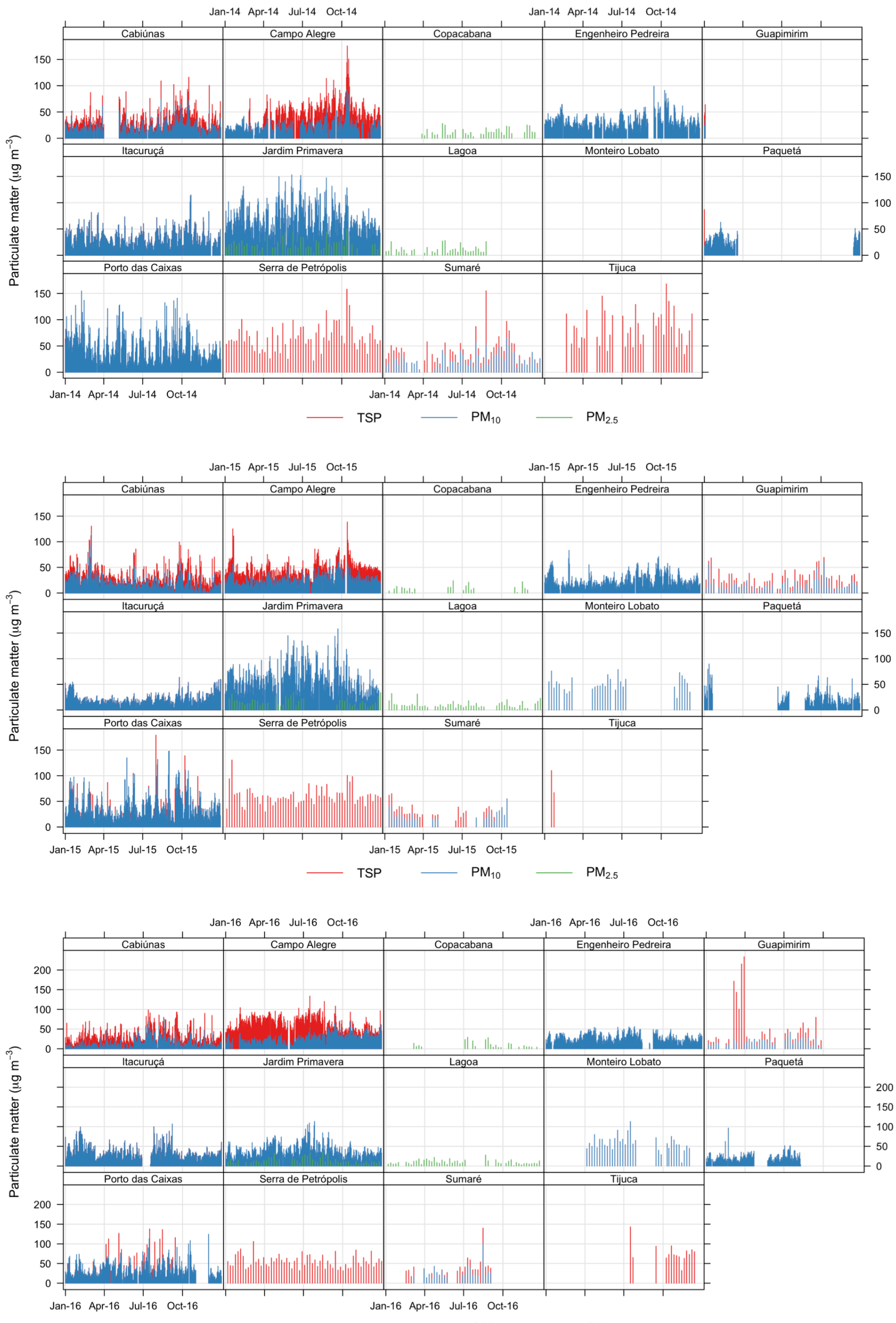

Figure 3 Particulate matter daily concentration recorded in 2014, 2015 and 2016 at all sampling sites.

Cluster analysis

The cluster analysis was performed in order to evaluate the similarities between sites. The dendrograms with sites grouping for each particle size are presented in Figure 4. For PM_{10} three groups can be verified and for TSP and $PM_{2.5}$ only two groups.

Wind direction and speed

In order to evaluate the PM transport near EPAs, wind roses were constructed using meteorological data provided by INEA (Figures 5 and 6). In general, the wind came from the same direction during the three years evaluated. However, in 2015 higher wind speed was recorded, which favor faster pollutants dispersion.

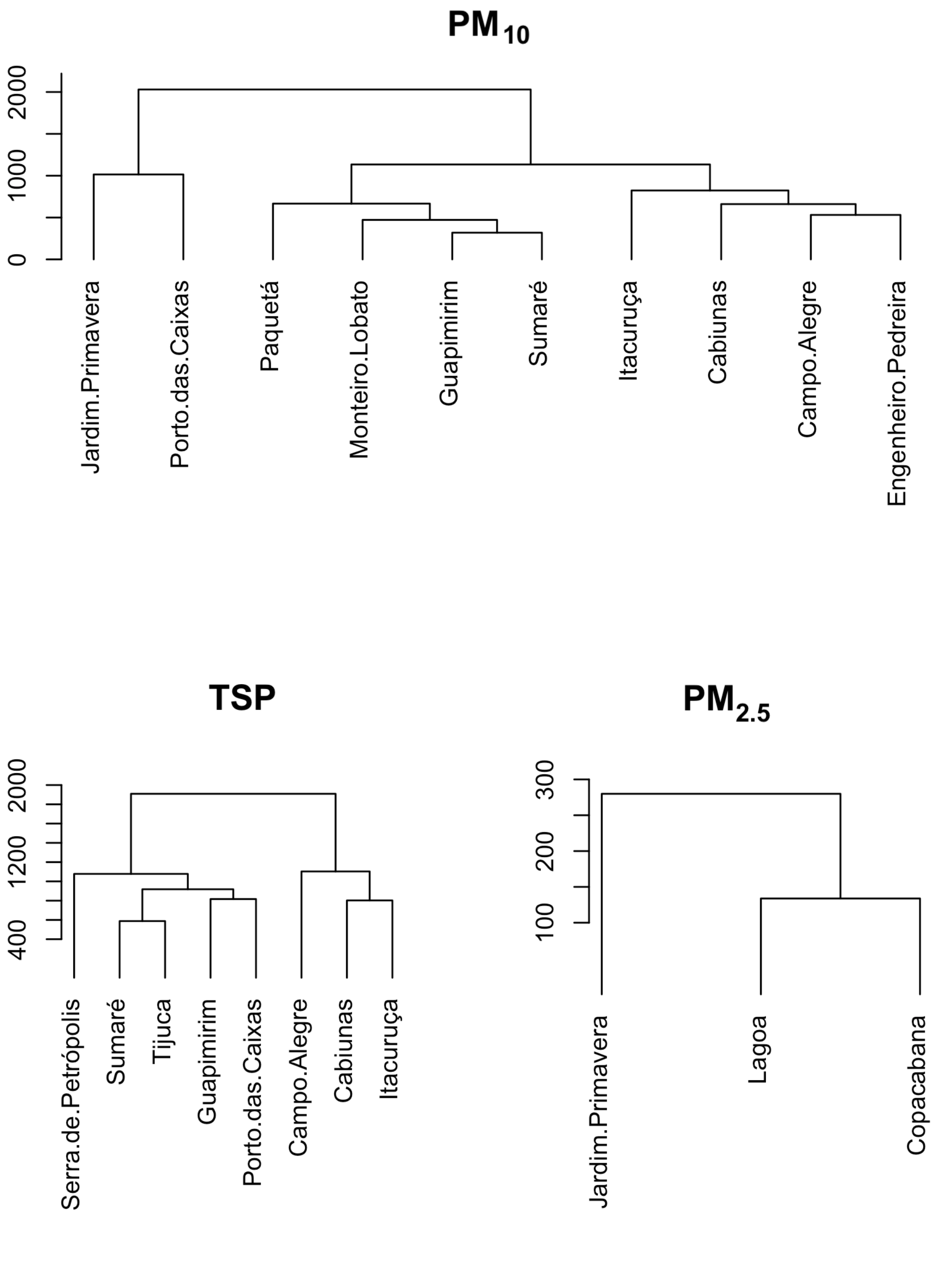

Figure 4 Hierarchical cluster analysis (dendrogram) of TSP, PM_{10} and $PM_{2.5}$ sampling sites considering concentration PM concentration measured between 2014 and 2016.

In Jardim Primavera and Monteiro Lobato were recorded winds from all directions, once they are located in the lowland. In Porto das Caixas winds from east region directing to Guanabara Bay. In Engenheiro Pedreira and Cabiunas winds come from the north region where slightly elevated areas can be found (60 – 100 m).

The wind speed was predominantly below 5 m s^{-1}, which is classified by Beauford scale as a gentle breeze, which can move trees leaves. Cabiunas and Engenheiro Pedreira recorded wind speed higher than 6 m s^{-1}, which may be classified as a strong breeze to storm. For all the monitoring stations, higher wind speeds were recorded in 2015.

Rainfall

Rainfall data can be related to PM concentrations because it contributes to the removal of particles from the atmosphere. Unfortunately, few stations monitor rainfall. Among sampling sites, Paquetá, Guapimirim and Porto das Caixas, located within a 15 km radius, and Cabiunas in the north of the state presented rainfall records that are presented in Figure 7.

The precipitation profiles were similar, registering lower volumes between the months of July and September. However, it is possible to highlight the high volume of precipitation in Paquetá in 2014, which differed greatly from the following years. The records of Guapimirim and Porto das Caixas were very similar due to the proximity and similarity of the two stations.

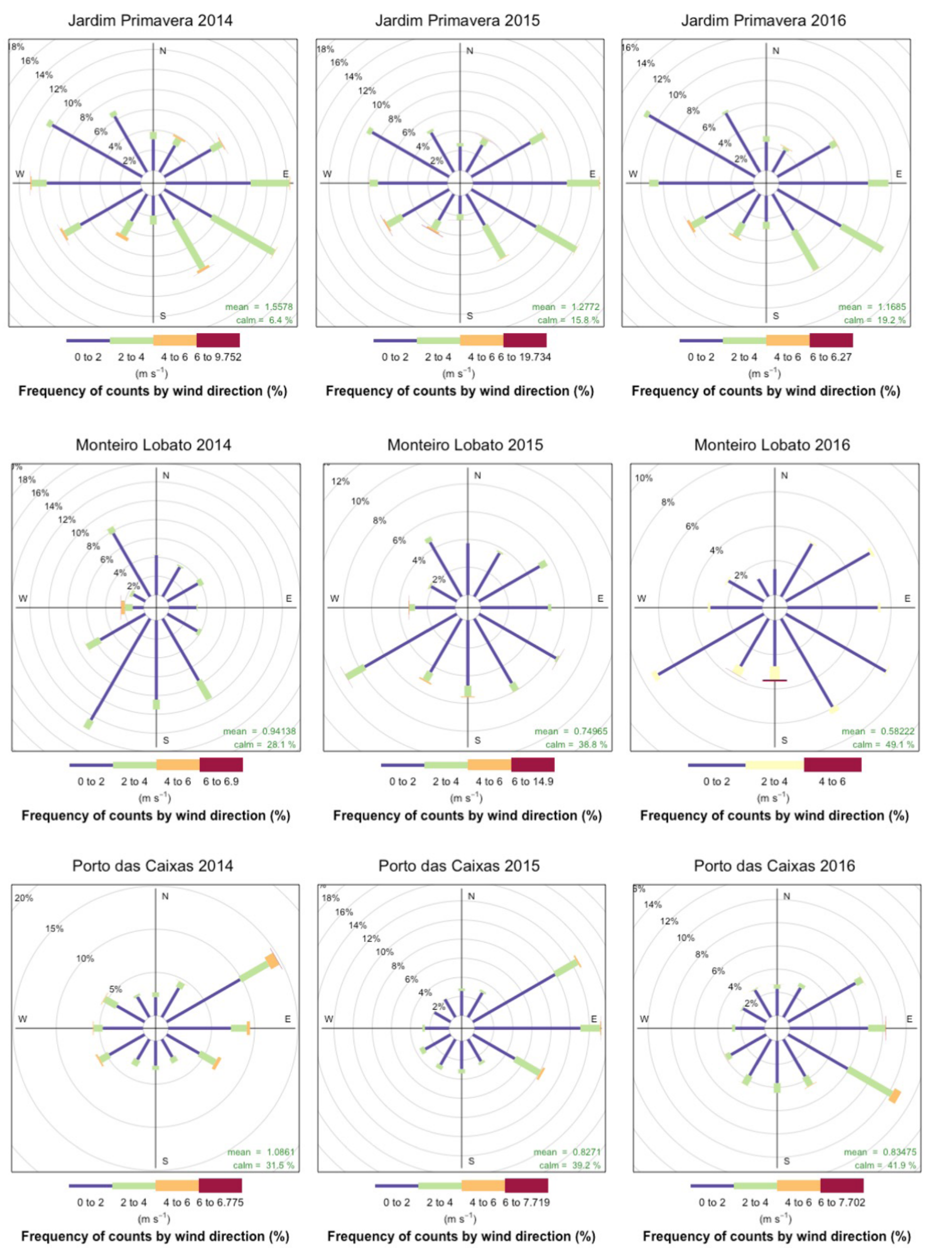

Figure 5 Wind Roses for the Jardim Primavera. Monteiro Lobato and Porto das Caixas areas for the three investigated years.

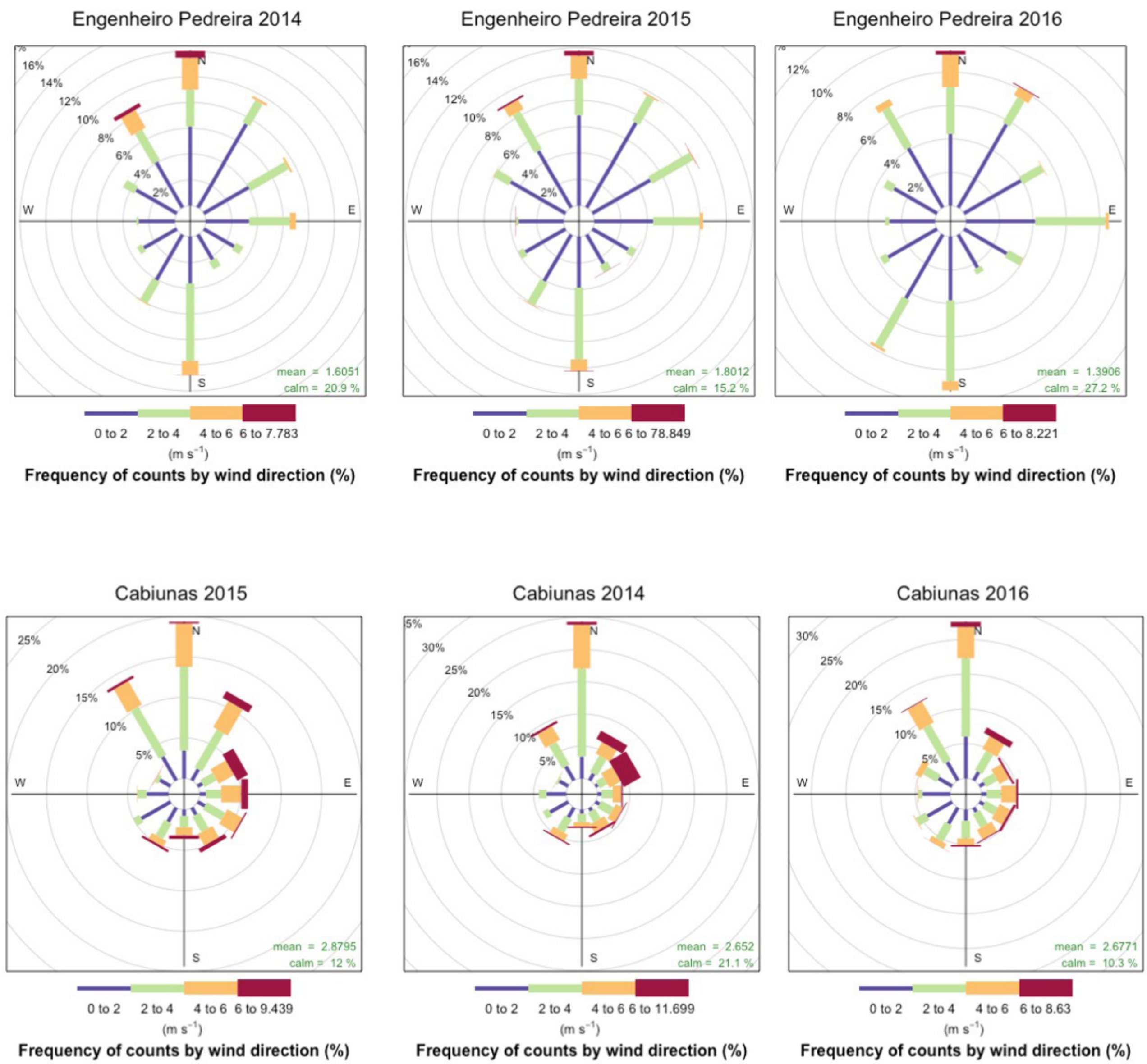

Figure 6 Wind Roses constructed for the Engenheiro Pedreira and Cabiúnas for areas for the three investigated years.

DISCUSSION

Particulate matter concentration did not vary much over the three years (2014 to 2016), which indicates that routine activities are accountable for air pollutants emission. Stations located in urban centers characterized by intense traffic and great density of people exceeded CONAMA annual standard for TSP and PM_{10}. These same stations also exceed CONAMA daily standard a few times per year. Once PM emission is similar in different years, meteorological variables determine if this pollutant would reach EPAs.

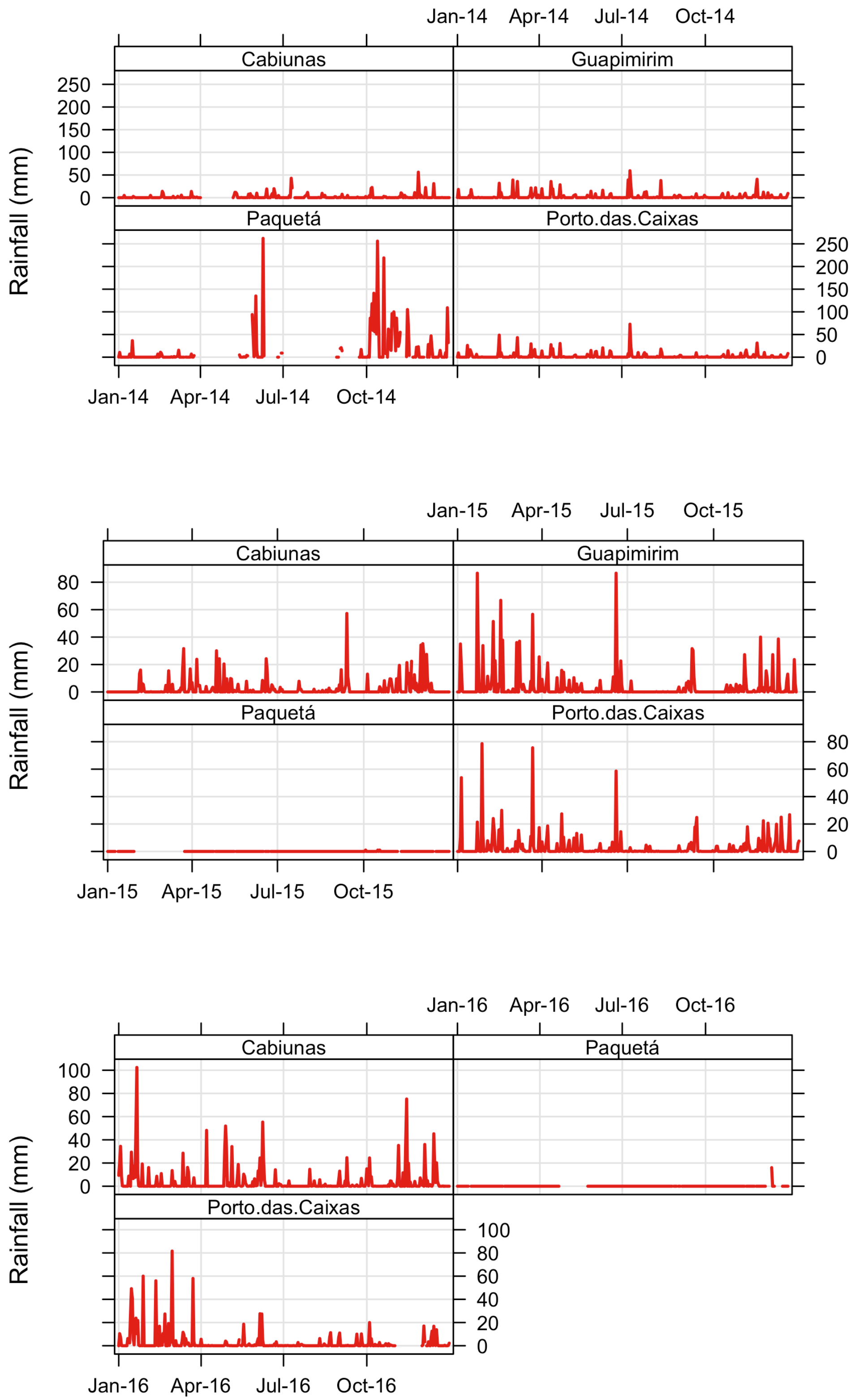

Figure 7 Rainfall records for Cabiúnas, Guapimirim, Porto das Caixas and Paquetá in 2014, 2015 and 2016.

Jardim Primavera, located in Duque de Caxias city, exceeded the PM_{10} annual limit in 2014 and 2015. This site is located near an important highway and an industrial complex, which may contribute to the high PM concentrations detected herein. The higher PM_{10} and $PM_{2.5}$ concentrations detected in 2014 may be associated with construction works on a new federal highway (Highway Raphael de Almeida Magalhães – BR-493 and BR-116) carried out near this sampling site. The works were finalized at the end of 2014 when the PM_{10} and $PM_{2.5}$ concentrations began exhibiting a decreasing trend.

At Jardim Primavera station, winds originated predominantly from the southeast, where the Duque de Caxias Oil Refinery (in Portuguese, *Refinaria de Duque de Caxias* – REDUC) is located. Wind from the northwest was also observed, which may have carried particulates emitted from the federal highway construction, and from the west, where an urban and industrial area is located. In 2015, winds were predominantly from the east, where an urban area is located, and from the southeast, where REDUC is located. In 2016, winds presented the same behavior as in 2014. The Tinguá Rebio is located northwest of the station, where part of emitted PM may reach. Western winds were also noted at this station, which may have hampered particulates from reaching the protection area.

Monteiro Lobato is an urban area located in the city of Nova Iguaçu. According to INEA, the high PM_{10} detected at this site can be attributed to local vehicle fleet growth and intensification of civil works. The lower means observed in 2015 may be due to the absence of sampling efforts between July and November of that year, a period when higher concentration was recorded in 2014 and 2016, which coincides with lower rainfall. A study of the climatological characteristics of Rio de Janeiro made by Neiva et al. 2017 reported that the lowest rainfall volumes are recorded between July and August. In the 50-year temporal evaluation of PM concentration, Gioda et al. 2016 observed that the highest PM concentrations are recorded between June and September, when rainfall is lowest. Predominantly south-north winds were noted at Monteiro Lobato. The train line is located south of this station, which connects the Baixada Fluminense (a group of 10 cities that belong to the metropolitan region of the city of Rio de Janeiro) to the city of Rio de Janeiro. The Tinguá Rebio is located north of this station. Wind direction contributes to the particulate matter emitted in urban areas reaching the investigated preservation area.

Jardim Primavera and Monteiro Lobato are located in the vicinity of the Tinguá Rebio. This reserve was created in the 19th century to protect the Atlantic Forest and the water sources that supply the Rio de Janeiro metropolitan area. The surrounding urbanization, however, conflicts with the aims of this preservation area and can contribute to pollutant emission, representing a risk for the EPA biodiversity.

Porto das Caixas is an urban sampling site located 4 km far from the Rio de Janeiro Petrochemical Complex (Comperj). The construction of this complex began in 2008 and was halted in 2015. Comperj activities and works contributed to the high TSP values. PM_{10} concentrations decreased from 2015, coinciding with the end of the works, while the mean TSP concentration increased in 2016, which may be associated with the absence of sampling efforts between October and December when lower concentrations are usually found due to the higher rainfall rates.

At Porto das Caixas, winds flow from the northeast to southwest, originating from the Comperj region and reaching the Guanabara Ecological Reserve EPA. Wind origin may explain the highest PM_{10} and TSP concentrations in 2014 compared to the other studied years. In 2015, winds originated predominantly from the east, where a rural area is located. In 2016, winds originated from the east, northeast, and southeast, from both rural and urban regions. Rainfall records for this season show low volumes for 2014, which, along with emissions from the Comperj construction sites, contribute to high PM concentrations. In fact, in the year 2014 Rio de Janeiro recorded the lowest rainfall volumes in 85 years (INEA 2014).

The Guapimirim station is located inside the ESECN Ecológica da Guanabara, which is located in the northeastern part of Guanabara Bay. It is a total preservation area dedicated to wildlife protection and was created to protect remaining mangroves and associated fauna and flora. The cities of Rio de Janeiro and Duque de Caxias are situated at the west of the bay, and Niterói and São Gonçalo are located to the east. All these cities are urbanized and densely populated, and several economic developments are installed around the bay, resulting in increased emission sources. Once winds come from different directions at this monitoring site, pollutants emitted in nearby urban centers (10 – 25 km away) may reach the EPA.

The sampling site with the highest TSP concentration was Tijuca, an urban neighborhood with intense traffic. The entrance of the PARNA Tijuca is located within this neighborhood. The Tijuca Mountain Forest results in decreased air circulation, preventing the sea breeze from dispersing pollutants. PARNA Tijuca is one of the first EPA in the world, created in the 19th century to protect local forest and promote the reforestation of areas used for coffee cultivation. It is currently one of the largest urban forests in the world, comprising almost 40 km^2. Construction activities and the transport sector are the main pollutant emission sources that pose a risk to the conservation of the PARNA Tijuca.

Paquetá is an island located in the central-northeast portion middle of Guanabara Bay, which implies, that it is under influence of pollutants emitted in cities around the bay. In addition, oil and gas utilities and vessels that travel and are anchored in the bay also represent emission sources. The sea breeze contributes to pollutants transport from continent and the bay to the island. This monitoring site present similar behavior for three years evaluated, a decreasing concentration since 2014, which may be explained by decrease on infrastructural works in Rio de Janeiro city. The island

Paquetá, insert in archipelago of the same name, is considered a national heritage, and beside of that, host a Municipal Park that corresponds a remnant of Atlantic Forest that ocupies the entire island territory. The study conducted by Gioda et al. 2018 showed that although there are few anthropogenic sources on the island of Paquetá, the concentrations of air pollutants are similar to nearby neighborhoods on the mainland, which have a greater number of sources. This similarity is a result of the transport of pollutants that are emitted on the continent.

Campo Alegre, Engenheiro Pedreira, Guapimirim, and Cabiunas are located in smaller, wooded cities with fewer people and vehicles. Lagoa, Copacabana, and Itacuruçá are located in the coastal region, which contributes to pollutant dispersion through ocean-continent exchanges. The Sumaré station is located on a 700 m high hill in the coastal region of the city of Rio de Janeiro. These characteristics may have contributed to the lower concentrations detected herein, below CONAMA limits, at these stations.

The Guapimirim concentration profile was similar to that observed for Porto das Caixas during the three study years. Both are influenced by emissions generated in Itaboraí, which explains the 2015 decreases, when Comperj works were halted, and the 2016 increases, when they began again.

The Serra de Petrópolis station is located over the federal highway between Duque de Caxias and Petrópolis, on a mountain range. The clear decreased concentration from 2014 to 2016 reflects alterations in constructions works that were conducted at this place. As the intensity of the works diminished, lower concentrations were registered. However, TSP levels were high, even when no works were in progress. Although TSP concentrations did not exceed CONAMA standard, they almost reached this limit and were comparable with values recorded in urban centers. The main PM source at this place is vehicle traffic, once sparse constructions may be found along the highway because it crosses an EPA.

Air mass origins and directions revealed that particles migrated towards nearby protection areas where the highest PM concentrations were observed (e.g., Jardim Primavera and Monteiro Lobato). When PM reaches EPAs, particles can be deposited either on plant surfaces or on the forest floor. Most remains on leaf surfaces, allowing for atmospheric resuspension, followed by transference to the ground by rain, becoming available for soil incorporation and plant absorption, or causing direct damage to plant systems (Shahid et al. 2017; Nowak et al. 2018), leading to morphological (stomatic blocking), physiological (pH alterations), biochemical (photosynthesis decreases) and molecular (intoxication) damages (Datta et al. 2016; Rai 2016b; Shahid et al. 2017).

The cluster analysis of the PM_{10} monitoring stations showed a clear clustering of Jardim Primavera and Porto das Caixas, two stations under the influence of petrochemical complexes. The second group formed by Paquetá, Monteiro Lobato, Guapimirim and Sumaré show influence of urban emissions, and the third group formed by Itacuruça, Cabiunas, Campo Alegre and Engenheiro Pedreira show mixed contribution of soil resuspension and urban emissions. For TSP similar groupings were observed. The first group with a predominance of urban sources, especially vehicular emissions, and the second group with natural and anthropogenic sources. On the other hand, the grouping of stations that monitored $PM_{2.5}$ grouped the urban stations separately from the station under industrial influence.

The evaluation of PM emissions near environmental protections areas indicates that urban activities contribute to higher particulate matter concentration averages. Considering the Rio de Janeiro scenario during the evaluated years, construction activities were one of the most important contributors to PM emissions, in addition to usual urban anthropogenic sources, such as fuel vehicle combustion and industrial activities. Wind direction indicated that, in some areas, particulate matter emitted by urban activities may reach environmental protection areas. When wind direction originated from PM sources, the sampling site presented higher PM concentrations, as expected.

This study sought to verify if urban pollutants affect the EPAs distributed throughout the state of Rio de Janeiro, however many limitations prevented a more in-depth evaluation of the impact of urban pollution on the EPAs. Although INEA has one of the largest air quality monitoring networks in the country, the pollutants monitored vary from one station to another, as do the meteorological variables. A more complete assessment should include more pollutants and consider the climatological effects that influence the dispersion of pollutants. However, it was possible to signal that urban pollution reaches the EPAs and can cause ecological imbalance.

Sustainability and environmental preservation are important issues today. Once the state of Rio de Janeiro boasts some of the most important Atlantic Forest remnants, the impact of pollutants emitted in urban centers should be monitored to ensure the preservation of those areas. Although the state monitors and inspects the air pollutants emission, more multidisciplinary studies are necessary to evaluate the effect of pollutants on fauna and flora to be preserved, once this study shows that pollutants emitted in urban centers reach EPAs.

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AUTHORSHIP CONTRIBUTIONS

Karmel Beringui: Conceptualization, Methodology, Data curation, Original draft preparation; Tatiana D. Saint'pierre: Reviewing and editing; Luiz Francisco P. G. Maia: Visualization and Investigation and Adriana Gioda: Reviewing and Editing.

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