

What Can We Learn about Improvements in Air Quality During the COVID-19 Pandemic? A Case Study in Four Cities Located in the Same State but with Different Emission Profiles

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This study aimed to assess how a partial lockdown due to the coronavirus disease 2019 (COVID-19) pandemic affected air quality in four cities with dissimilar characteristics. In three cities, Araraquara (ARQ), Presidente Prudente (PPE), and Santos (STS), reductions in NO₂ concentrations were observed due to social distancing. Conversely, in Santa Gertrudes (SGD), NO₂ concentrations increased, indicating that a brief pause in ceramics industry activity was not sufficient to reduce NO₂ emissions. A variable behavior was observed in O₃ concentrations; in some cases, it followed the trends observed in previous years, but in others, an increase or decrease in concentrations was observed due to variations in concentrations of NO₂ and volatile organic compounds and/or climatic conditions. Particulate matter (PM) concentrations decreased in SGD and STS due to social distancing, meteorological conditions, such as wind speed, and reductions in industrial and port activities. Nevertheless, in the cities of ARQ and PPE, particulate matter with aerodynamic diameter $\leq 10 \mu\text{m}$ (PM₁₀) concentrations were elevated during the pandemic period, due to numerous biomass burning events in 2020. Thus, although vehicular and industrial emission control/reduction policies are effective in improving air quality, they may not be sufficient to achieve air quality standards if they are not combined with more restrictive measures to manage biomass burning.



Keywords: air quality, nitrogen dioxide, ozone, particulate matter, coronavirus

Introduction

At the end of 2019, a new type of coronavirus, severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2), was discovered in Wuhan, China. In March 2020, the World Health Organization (WHO) declared that coronavirus disease 2019 (COVID-19), the disease caused by this new virus, would be classified as a pandemic.¹ After that, governments worldwide implemented different actions to reduce the number of people infected by SARS-CoV-2, such as encouraging social distancing, restricting travel, and closing schools and non-essential services.²

With the restrictions imposed by governments as a way to reduce contamination by SARS-CoV-2, reductions in pollutant emissions from industrial activities and the transport sector have been reported in some cities worldwide. In urban and rural areas of the Netherlands, reductions in the average concentrations of nitrogen oxides (NO_x) and particulate matter (PM) were reported during the lockdown period (March to May 2020), while ozone (O₃) concentration increased as a direct result of the decrease in NO_x emissions.³ Similar results have been reported in studies carried out in other countries such as Germany, China, India and Austria.⁴⁻⁹

In Brazil, improvements in air quality due to partial lockdown were reported in different cities of the São Paulo state and in the city of Rio de Janeiro, during the first months of 2020 compared to the same period in previous years.^{2,10-15} Similarly to the studies conducted in other countries mentioned above, the Brazilian studies reported that the levels of the main air pollutants: NO₂, NO, particulate matter

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with aerodynamic diameter $\leq 10 \mu\text{m}$ (PM_{10}), particulate matter with aerodynamic diameter $\leq 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$), carbon monoxide (CO) and sulfur dioxide (SO_2) decreased, while concentrations of O_3 increased.

Although most studies have reported improvements in air quality during lockdown periods, no significant enhancement in air quality remained during the entire period of pandemic. In New York City (the United States), concentrations of NO_2 (51%) and $\text{PM}_{2.5}$ (36%) decreased shortly after the introduction of the lockdown measures. However, the decreases were not significant when compared to the 5-year historical data.¹⁶ In Nigeria, the reductions in $\text{PM}_{2.5}$ concentrations found in 2020 compared to historical data (2002-2020) were not attributed to lockdown, but rather to favorable meteorological conditions.¹⁷

A more recent and global study¹⁴ found a positive correlation between the reductions in NO_2 and NO_x concentrations and peoples' mobility in most cities across the world. However, a more complex and heterogeneous behavior was observed for PM and ozone, indicating that other sources besides vehicular emissions also contributed to change the air quality.

Therefore, to evaluate the effects of lockdowns on air quality, it is important to consider both long-term changes in air quality and short-term fluctuations in pollutant concentrations. These changes may be due to local or regional environmental regulations, changes in weather conditions, seasonal variation, or other factors, such as events of fire.¹⁶ Research concerning these issues is important since it can assist individuals and governments in understanding how to reduce pollution in the long term.^{11,18}

In the present work, an extensive set of *in situ* air quality observations (PM_{10} , $\text{PM}_{2.5}$, NO_2 , and O_3 concentrations) from four cities in São Paulo State was analyzed, covering the years from 2016 to 2021, including not only the first weeks, but the entire period of the pandemic in which vehicular and economic activities were partially paralyzed. Despite being situated in the same state, the four cities chosen have dissimilar local factors that influence air pollutants emissions, such as the number of inhabitants, vehicular fleet, type of economic activity, land use and meteorological conditions. Therefore, the aim was to evaluate how important is the role of mobility and industry restrictions in the air quality compared to the other factors that influence air pollution in urban centers, such as meteorological variables and fire frequency. This detailed analysis can be used to improve public policies in regions with similar characteristics to the region of this study.

Experimental

Study area

São Paulo is the most populous Brazilian state, with approximately 46 million inhabitants, and has the largest vehicle fleet, with ca. 30 million vehicles, including light and heavy-duty.¹⁹ In this study, four cities were selected, namely Araraquara (ARQ), Presidente Prudente (PPE), Santa Gertrudes (SGD), and Santos (STS), located in different regions of the state and with different air quality indexes in 2019 (Figure 1).²⁰ Further details of each city are provided in Table S1 (Supplementary Information (SI) section).

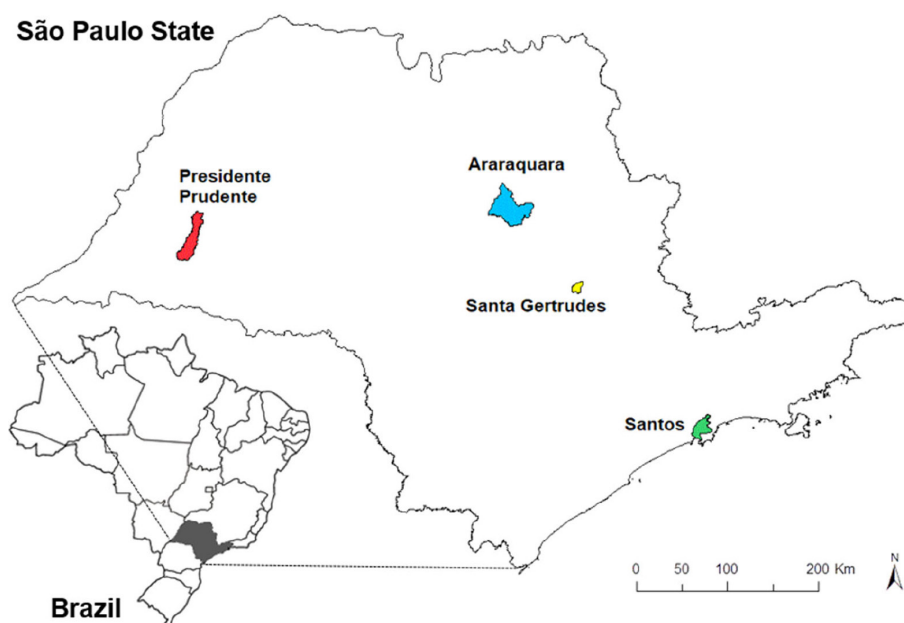


Figure 1. Map showing the location of the four cities used in this study.

Air pollutants, meteorological and social distancing data

The period considered in this work was from April 1, 2016, to March 31, 2021. Wind direction (degree), air temperature ($^{\circ}\text{C}$), relative humidity (%), atmospheric pressure (hPa), wind speed (m s^{-1}), and the hourly concentrations ($\mu\text{g m}^{-3}$) of NO_2 , PM_{10} , $\text{PM}_{2.5}$, and O_3 were obtained from the website of the Environmental Company of São Paulo State (CETESB), using the QUALAR system.²¹ PM_{10} and $\text{PM}_{2.5}$ concentrations were measured by the beta radiation method, NO_2 concentrations were quantified by chemiluminescence, and O_3 concentrations were determined by ultraviolet photometry.²¹ Pollutant quantifications were carried out continuously and the data are available online as hourly means.²¹ Precipitation values (mm) were obtained from the agrometeorological monitoring system, the Department of Water and Electric Energy website, the National Center for Monitoring and Warning of Natural Disasters, and the Santos city hall website.²²⁻²⁵ Daily mean concentrations used in the statistical analyses were calculated from the hourly concentrations obtained from CETESB website.

The social distancing index values were obtained from the São Paulo Smart Monitoring System, which uses cell phone antennas to detect the location of the population.^{26,27} The first mobility restricting measure in São Paulo State started on March 24, 2020, with only the functioning of essential sectors being allowed.²⁸ Subsequently, the state government proposed a plan called “São Paulo Plan”, which entered into force on July 1, 2020, providing restrictive measures according to health data.²⁹ These measures persisted until March 31, 2021, which was consequently the last day considered in the present study.

Statistical analysis

The statistical analyses were performed with Minitab® 19.2020.1 software.³⁰ In all tests, data from the same season, either dry (April to September) or wet (October to March) were compared.

The Mann-Whitney test was used to determine whether the pollutant concentrations median during the pandemic period was significantly different from the concentration median determined in the previous years. A unilateral approach was used and, because of this, in some cases two tests were performed. In the first, the null hypothesis tested was that there was no difference between the central positions of the two analyzed populations and the alternative hypothesis that the period after the beginning of the restrictive measures had a lower central position. If the alternative hypothesis was rejected ($p > 0.05$), in the second test the alternative hypothesis was that the period after the

beginning of restrictions had a higher central position. The Mann-Whitney test can be used as a non-parametric equivalent of the t -test. It is used to determine whether or not two unpaired groups belong to the same population.³¹ The Mann-Whitney test tests equality of medians, unlike the t -test which tests equality of means.

The Mann-Kendall test was used to determine whether the concentrations of the pollutants and the degree of social distancing showed any significant increasing or decreasing trend. In this test, data were divided into two groups for both seasons, in which the first group contained the concentrations determined before the pandemic, while the second group consisted of all the concentrations, including the pandemic period. The trends were calculated for both groups, so we could evaluate if the trend of previous years continued or not during the pandemic period. Data were arranged chronologically, testing the null hypothesis that there are no trends throughout the series. The two alternative hypotheses were that there is an upward or downward trend. The magnitude of the trends was determined by calculating the Sen's slope.

The Mann-Kendall test is a method used to identify whether there are significant trends in time series data.^{32,33} Because it is a non-parametric method, it does not require a normal distribution of the data.³²⁻³⁴ It has the advantage of being little influenced by sudden changes in data or non-homogeneous series.³⁵ The test is based on whether or not to reject the null hypothesis (H_0) that there is no trend in the data series. These trend analyzes are calculated to a certain significance level (α). Sen's Slope is used as a complement to the Mann-Kendall test, using the latter to identify a significant trend in the data. It is also a non-parametric method and is used to determine the rate of change of parameters in time series.³⁶ In this statistical test, the magnitude of the slope of the line is determined by calculating the median of the slopes of all lines between each pair of points in a time series.³⁶ By using the median, this test is less susceptible to the influence of outliers when compared to methods based on the mean, such as the least squares method.

The influence of meteorological parameters was evaluated using the Kruskal-Wallis test, as weather conditions can affect the accumulation or dispersion of pollutants.⁵ In this test, each season's median of monthly means of wind direction, air temperature, relative humidity, atmospheric pressure, wind speed and accumulated precipitation, were compared over the years. Although the Kruskal-Wallis test does not provide the stochastic dominance of a group, it indicates whether a year median was different from the others or not, and if different, a descriptive analysis was performed to evaluate if a certain meteorological parameter was higher or lower during the pandemic period compared to the previous years.

In addition, pollution roses were generated to evaluate the behaviors of the pollutants under different conditions of wind speed and direction, using the PolarPlot function of the OpenAir R package.³⁷⁻³⁹

The Spearman correlation was used to measure relation between two variables, among the meteorological parameters, the pollutant concentrations, and the social distancing index values, obtained from 2020 and 2021. This data analysis measures the monotonic relationship between two variables, that is, when the variables tend to change together without necessarily being at a constant rate. There is a positive correlation between two variables when one tends to increase as the other also increases. When there is a negative correlation, it means that high values of one of the variables correspond to low values of another.

Time-lagged and seasonally-lagged linear regression models were tested, but the results were similar to those obtained in the absence of these models, so they were not further considered.

Results and Discussion

Meteorological parameters

Most of the meteorological parameters barely varied in the cities studied (Table S2, SI section). In ARQ and PPE, none of the parameters varied significantly over

the years, both in the dry and the wet season. In the dry period, wind speed in SGD and STS, wind direction in SGD, and relative humidity in STS were different at least in one of the evaluated years. In the wet season, variations in wind direction and wind speed in SGD and STS, and relative humidity in STS were observed. More details will be discussed in the following sections. For temperature, atmospheric pressure (data not available in SGD), and precipitation, all medians were equal.

Temporal change in atmospheric NO₂ concentrations during the COVID-19 pandemic

In ARQ and PPE, the NO₂ concentrations were lower in the wet season than in the dry season (Figure 2). Although the NO₂ concentrations in ARQ were slightly higher than in PPE, similar downward trends were observed for both cities over the years, in both seasons in the pre-pandemic period (Table 1).

The downward trends were expected mainly due to improvements of vehicular emissions, with no significant variations of fuel consumption in ARQ and PPE between 2016 and 2019, and prohibition of burning sugarcane leaves before harvesting. In Brazil, vehicular NO_x emissions have decreased since the introduction of mandatory use of catalysts for vehicles produced from 2009 onwards, and in São Paulo State, NO_x emissions decreased by

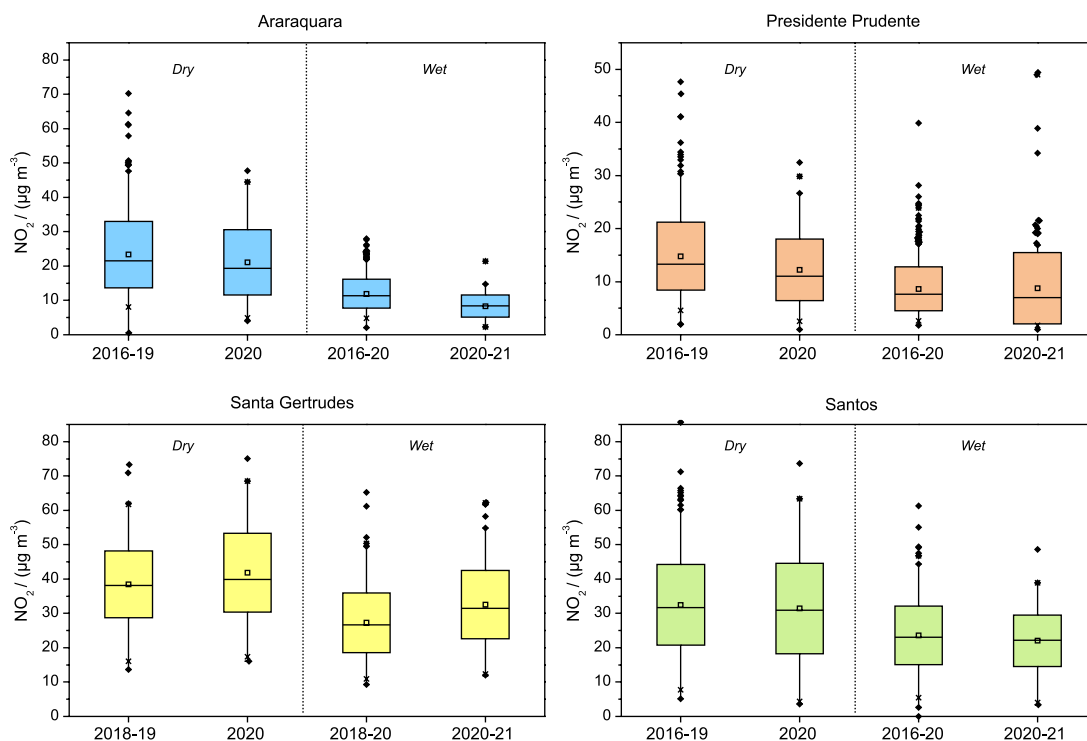


Figure 2. Concentrations of NO₂ (µg m⁻³) in the four cities studied, separated into pre-pandemic and pandemic periods, during the dry and wet seasons. The whiskers plots show the concentrations for the mean (□), median (–), outliers (•), 25th and 75th percentiles, minimum, and maximum.

Table 1. Mann-Kendall trends in NO₂ concentrations, and the corresponding Sen's slopes (Qmed), for the cities studied, separated into dry and wet seasons, and comparison between the concentrations during the pandemic and pre-pandemic periods, using the Mann-Whitney test

Pollutant	City	Season	time interval	n ^a	Trend	Qmed	Mann-Whitney hypothesis	
NO ₂	ARQ	dry	2016-19	715	downward ^b	-0.0044	2020 < 2016-19 ^b	
			2016-20	845	downward ^b	-0.0049		
		wet	2016-20	706	downward ^b	-0.0021		2020-21 < 2016-20 ^b
			2016-21	784	downward ^b	-0.0020		
	PPE	dry	2016-19	678	downward ^b	-0.0023	2020 < 2016-19 ^b	
			2016-20	861	downward ^b	-0.0040		
		wet	2016-20	698	downward ^b	-0.0038		2020-21 < 2016-20 ^b
			2016-21	872	downward ^b	-0.0026		
	SGD	dry	2018-19	363	no trend ^d	N/A	2020 > 2018-19 ^b	
			2018-20	544	upward ^b	0.0286		
		wet	2018-20	386	no trend ^d	N/A		2020-21 > 2018-19 ^b
			2018-21	568	upward ^b	0.0081		
	STS	dry	2016-19	661	upward ^c	0.0067	2020 = 2016-19 ^d	
			2016-20	835	no trend ^d	N/A		
		wet	2016-20	698	no trend ^d	N/A		2020-21 < 2016-19 ^c
			2016-21	880	no trend ^d	N/A		

^an = number of mean daily concentrations; ^bp-value < 0.01; ^cp-value < 0.05; ^dnot applicable. ARQ: Araraquara; PPE: Presidente Prudente; SGD: Santa Gertrudes; STS: Santos; N/A: not applicable.

approximately 8% between 2016 and 2019.⁴⁰ Regarding biomass burning, in the past, fire was widely applied to facilitate manual harvesting of sugarcane, but from 2007 onwards, this practice was gradually eliminated in São Paulo State.⁴¹ Currently, nearly 100% of sugarcane harvesting is mechanized.⁴² However, biomass burning is still an environmental problem in Brazil, especially since the highest annual number of fires in Brazil in the past ten years was recorded in 2020, according to estimates by the National Space Research Institute (INPE).⁴³

During the pandemic period, the NO₂ concentrations in ARQ and PPE were lower than in previous years, during the wet and dry seasons (Table 1). In the dry season, the concentrations were lower than expected from the downward trend of the previous years, which was confirmed by the more negative slopes obtained when the pandemic data were added (Table 1). In addition, in these cities, a significant negative correlation between the social distancing index values and the concentrations of NO₂ was determined ($p < 0.01$; Table S3, SI section). Moreover, no significant changes in weather conditions that could cause this decrease were observed. Thus, the most likely cause of these reductions in NO₂ emissions was social distancing, mainly due to the decrease in vehicular emissions (Table S3).

Similarly, to ARQ and PPE, the concentrations of NO₂ in the dry season in SGD were higher than in the wet season (Figure 2). However, no trends in the

concentrations in the pre-pandemic period were found in either season (Table 1). Among the four cities studied, SGD had the smallest vehicular fleet, but the highest NO₂ concentrations. This is because, although the small vehicular fleet of the city remained virtually constant in the years studied, fuel consumption has increased, mainly due to greater use of diesel.^{44,45} Other important sources of NO₂ are the ceramics industries. The region is the main producer of ceramics in Brazil (60% of total production in the country), and this activity remaining approximately constant during the study period.⁴⁶ The maintenance of ceramics production and a possible balance between the improvement of vehicular emissions and increased fuel consumption may also have been responsible for the lack of any trend in the concentrations. It is important to note that the SP-310 highway, one of the busiest in the state, is located 150 meters from the SGD sampling site, and that the ceramics industry is a major consumer of diesel.⁴⁷

During the pandemic period, NO₂ concentrations in SGD were higher than in the previous years, for both dry and wet seasons (Table 1). Moreover, for both seasons, the NO₂ concentrations showed increasing trends when the pandemic period was included (Table 1). These trends and observed increases in NO₂ emissions during the pandemic period can be explained by the activities of the ceramic industry. In 2020, although ceramics production was slightly lower than in previous years, the wet ceramics production process was mostly used, consequently resulting

in higher thermal NO₂ emissions.⁴⁸ Furthermore, the highest concentrations of NO₂ in 2020 were found in periods when low-velocity winds mainly came from directions corresponding to the locations of most of the ceramics industries and the SP-310 highway (Figures S1a and S1b, SI section). In general, the wind directions in SGD in 2020 were different from those observed in previous years. Besides, wind speeds were higher than in previous years, and favored a greater dispersion of pollutants, which would otherwise have led to even higher NO₂ concentrations.

The NO₂ concentrations in STS in the dry season during the pandemic period were not different from those determined in previous years, but in the wet season NO₂ concentrations were lower during the pandemic period. (Table 1).

STS city has the 14th largest vehicle fleet in São Paulo State, and from 2016 to 2019 no major variations in either fuel consumption or in the types of fuel consumed were observed.⁴⁵ STS attracts many tourists and, consequently, a substantial flow of vehicles towards the city.^{49,50} In addition, the port in STS is the largest in Brazil and the second largest in Latin America, with an average 5% growth in cargo volume *per* year between 2011 and 2020.⁵¹

The magnitude of NO₂ concentrations in STS was the second-highest among the cities studied, being similar to the levels found in SGD, probably because the STS monitoring station was located approximately 2 km distant from the port, and it has been shown that port activities make an important contribution to NO_x concentrations.⁵² Therefore, the port emissions, together with vehicular emissions, were the main sources of NO₂ emissions to the STS atmosphere.

In the city of STS, the concentrations of NO₂ were higher in the dry season than in the wet period (Figure 2 and Table 1). However, while an upward trend in NO₂ concentrations occurred from 2016 to 2019 in the dry season, when the period was expanded to include 2020, no significant trend was observed (Table 1). In this season, a negative correlation was observed between the values of the social distancing index and NO₂ concentrations (Table S3). During this time, most meteorological parameters did not show significant differences from those recorded in previous years, except for wind speeds, which were lower (Table S2). These results demonstrated that although port activities increased throughout the pandemic period, emissions of NO₂ were lower than expected. The reason for this reduction was the decrease in vehicular emissions. This hypothesis is in line with the negative correlation between the values of the social distancing index and NO₂ concentrations. In addition, in the same year, the lowest flows of passenger and commercial vehicles towards STS

were recorded (Table S4, SI section), and the lowest hotel occupancy has been recorded since 2015, when recording began.^{49,50} It should be noted that wind speeds in the dry period in 2020 were lower than in previous years, so even higher concentrations would have been obtained if the pandemic had not occurred (Table S2).

In the wet season, NO₂ concentrations were lower during the pandemic period than in previous years, and no significant trend was observed from 2016 to 2019 or when the period was expanded to include 2020 (Table 1). In this season, a negative correlation between the social distancing index and NO₂ data was also observed (Table S3). Social distancing during the pandemic also led to a decrease in NO₂ emissions in STS during the wet season, although no trend was observed in the concentrations of this pollutant. As in the dry period, decreases in NO₂ concentrations were associated with lower vehicular emissions, as confirmed by the negative correlation between the social distancing index and NO₂ data (Table S3).

Furthermore, the highest concentrations of NO₂ in STS during the dry and wet seasons of 2020 were determined when low velocity winds came from directions where the port facilities are located, indicating the important contribution of this source to NO₂ emissions and of meteorological parameters (Figures S1c and S1d). The influence of the port was lower in the wet season, when the monthly cargo volume was lower (Mann-Whitney test, $p < 0.05$).⁵³ The absence of a trend in the wet season could have been a result of this, together with the increased flow of vehicles in recent years being compensated by lower individual vehicle emissions. In contrast, vehicle emissions may be lower in the dry season, which is the coldest season of the year, when the influx of tourists is smaller. The observed increase in the concentrations of NO₂ in the dry season may have been due to the annual and seasonal increase in port activity.

Temporal change in atmospheric O₃ concentrations during the COVID-19 pandemic

The behavior of ozone concentrations from 2016 to 2020 (before the pandemic) varied greatly in both seasons in the cities where this pollutant was monitored: ARQ, PPE and STS (Figure 3 and Table 2).

In the pandemic period, in the city of ARQ, the concentrations of O₃ in the dry season were lower than before the pandemic, but in the wet season they were higher (Table 2 and Figure 3). It was noted that the concentrations of O₃ were higher in the wet season than in the dry season (Mann-Whitney test, $p = 0$), while the opposite was true for the concentrations of NO₂ (Mann-Whitney test, $p = 0$).

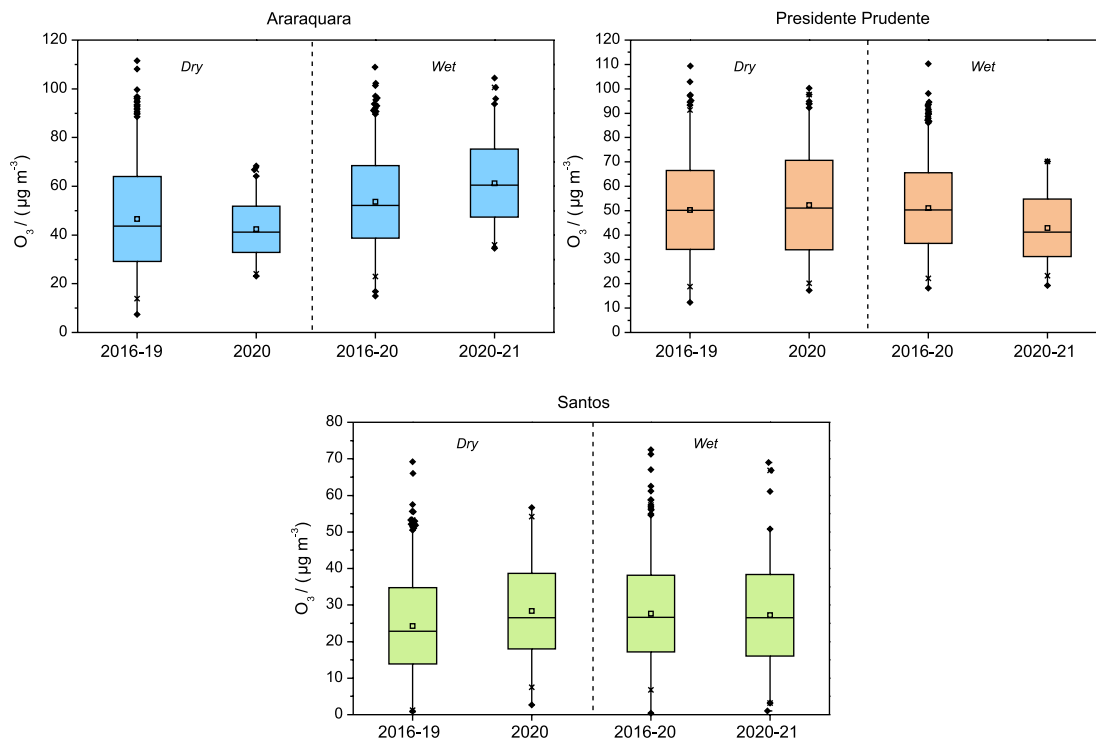


Figure 3. Concentrations of O₃ (µg m⁻³) in the three cities studied, during the dry and wet seasons, separated into pre-pandemic and pandemic periods. The whiskers plots show the concentrations for the mean (□), median (—), outliers (•), 25th and 75th percentiles, minimum, and maximum.

Table 2. Mann-Kendall trends in O₃ concentrations, and the corresponding Sen’s slopes (Qmed), for the cities studied, separated into dry and wet seasons, and comparison between the concentrations during the pandemic and pre-pandemic periods, using the Mann-Whitney test

Pollutant	City	Season	time interval	n ^a	Trend	Qmed	Mann-Whitney hypothesis
O ₃	ARQ	dry	2016-19	663	upward ^b	0.0079	2020 < 2016-19 ^c
			2016-20	790	upward ^b	0.0128	
		wet	2016-20	702	no trend ^d	N/A	2020-21 > 2016-19 ^b
			2016-21	838	upward ^b	0.0075	
	PPE	dry	2016-19	715	no trend ^d	N/A	2020 = 2016-19 ^d
			2016-20	898	no trend ^d	N/A	
		wet	2016-20	721	downward ^b	-0.0177	2020-21 < 2016-19 ^b
			2016-21	844	downward ^b	-0.0170	
	STS	dry	2016-19	661	no trend ^d	N/A	2020 > 2016-19 ^b
			2016-20	835	upward ^b	0.0023	
		wet	2016-20	706	no trend ^d	N/A	2020-21 = 2016-19 ^d
			2016-21	871	no trend ^d	N/A	

^an = number of mean daily concentrations; ^bp-value < 0.01; ^cp-value < 0.05; ^dnot applicable. ARQ: Araraquara; PPE: Presidente Prudente; STS: Santos; N/A: not applicable.

In the dry season during the pandemic period, O₃ concentrations were positively correlated with the social distancing index and negatively correlated with NO₂ (Table S3). A synchronous inverse correlation between the two pollutants has already been observed in the same region.⁵⁴ In the dry season, for both 2016-2019 and 2016-2020, upward trends in concentrations were observed, with a greater slope when including the pandemic period

concentrations (2016-2020, Table 2). These results indicate a reduction in the consumption of O₃ in reactions with nitrogen oxides, due to reductions in emissions of the latter as a consequence of social distancing. However, it is important to note that other variables also influence the formation and consumption of this pollutant.

In the wet season, no trend in O₃ concentrations was observed before the pandemic (2016-2020), while an

upward trend was observed when data from the pandemic period were added (Table 2). In this case, there was no significant correlation between the concentrations of O_3 and NO_2 (Table S3). The correlation between the social distancing index and the concentration of O_3 was negative, while the correlation between this pollutant and the concentration of PM_{10} was positive (Table S3). Considering these correlations, it could be hypothesized that the concentrations of O_3 were predominantly influenced by nucleation of new particles, rather than by the effect of the pandemic, produced from their reactions with volatile organic compounds (VOCs) and/or semi-volatile compounds. It should be noted here that, in the wet season in the ARQ region, the nucleation of new particles is favored by lower particle concentrations and by higher concentrations of biogenic VOCs, compared to the dry season.⁵⁴ This happens because the nucleation of new particles can occur by means of reactions involving VOCs or semi-volatile organic compounds and O_3 or nitrate radicals.⁵⁴

In the city of PPE, the O_3 concentrations were similar in the dry and wet seasons, and they were close to the concentrations observed in ARQ (Figure 3). For both seasons, the concentrations of O_3 were not influenced by social distancing (Table S3) and were as expected based on trends observed in previous years.

In the dry season, compared to the pandemic period, the concentrations of O_3 before the pandemic were not significantly different (Table 2 and Figure 3), and no trends in O_3 concentrations were observed from 2016 to 2020 or from 2016 to 2021 (Table 2). In addition, there was a positive correlation between O_3 and PM_{10} , a negative correlation between the social distancing index and O_3 , and a non-significant correlation between O_3 and NO_2 (Table S3).

In the wet season, decreasing trends in O_3 concentrations were observed both before the pandemic and with the addition of the data obtained during the pandemic period (Table 2). In the latter case, the downward trend was slightly less accentuated, and the O_3 concentrations in 2020-2021 were lower than in previous years (Table 2 and Figure 3). These same trends were also observed for NO_2 , but the correlation between O_3 and NO_2 was not significant (Table S3). The correlation between O_3 and PM_{10} was positive (Table S3). It is worth noting that in the wet season, O_3 concentrations were lower than those recorded in previous years. This behavior is in line with expectations, considering that decreasing trends were observed in O_3 concentrations before the pandemic and with the addition of pandemic data, the downward trend was only slightly less accentuated. Furthermore, in both seasons the correlation between O_3 , NO_2 , and PM_{10} were similar to those observed

for ARQ in the wet season (Table S3). This indicates that also in this city the O_3 concentrations were limited by the availability of VOCs and/or semi-volatile compounds.

For the city of STS, the dry season concentrations of O_3 before the pandemic were lower than during the pandemic period, while no significant difference was observed for the wet season (Figure 3 and Table 2). It was also noted that for the pre-pandemic period, O_3 concentrations were higher in the wet season than in the dry season (Mann-Whitney test, $p < 0.05$), while no significant difference (Mann-Whitney test, $p > 0.05$) was observed for the pandemic period.

The O_3 concentrations in STS were about half of the concentrations of this pollutant in ARQ and PPE, probably due to the higher concentrations of NO_2 , an O_3 sequesterant, in STS. In this city, no trends in the O_3 concentrations and no significant correlations between the social distancing index values and O_3 concentrations were observed in the dry and wet seasons (Table S3). However, in the dry season during the pandemic period, the O_3 concentrations showed an upward trend (Table 2), with no significant correlations with the social distancing index, and negative correlations with other pollutants (Table S3). These results indicated that social distancing did not directly influence the O_3 concentrations, with the processes of formation and/or consumption being most important. Since the port activities continued to show an upward trend, while NO_2 concentrations did not increase as in previous years, the O_3 concentrations showed an upward trend and were higher than in previous years. The increase in the concentrations may also have been due to the lower wind speeds observed during the pandemic period (Table S2). In the wet months of the year, O_3 concentrations were not influenced by social distancing or by the other pollutants analyzed. No significant correlations were observed between the concentrations of O_3 and other pollutants (Table S3), and that no significant trends were observed in O_3 concentrations (Table 2), although there was less dispersion of pollutants in 2020, due to lower wind speeds.

Temporal change in atmospheric PM_{10} and $PM_{2.5}$ concentrations during the COVID-19 pandemic

In the city of ARQ, the concentrations of PM_{10} in the pre-pandemic dry season were higher than in the pre-pandemic wet season (Figure 4). Before the pandemic, downward trends in PM_{10} concentrations were found in both seasons (Table 3). In this city, before the pandemic, the downward trends in PM_{10} concentrations were expected, because PM emissions in ARQ were mainly due to biomass burning and vehicular emissions, which had been reduced over the years by restrictive legislation.

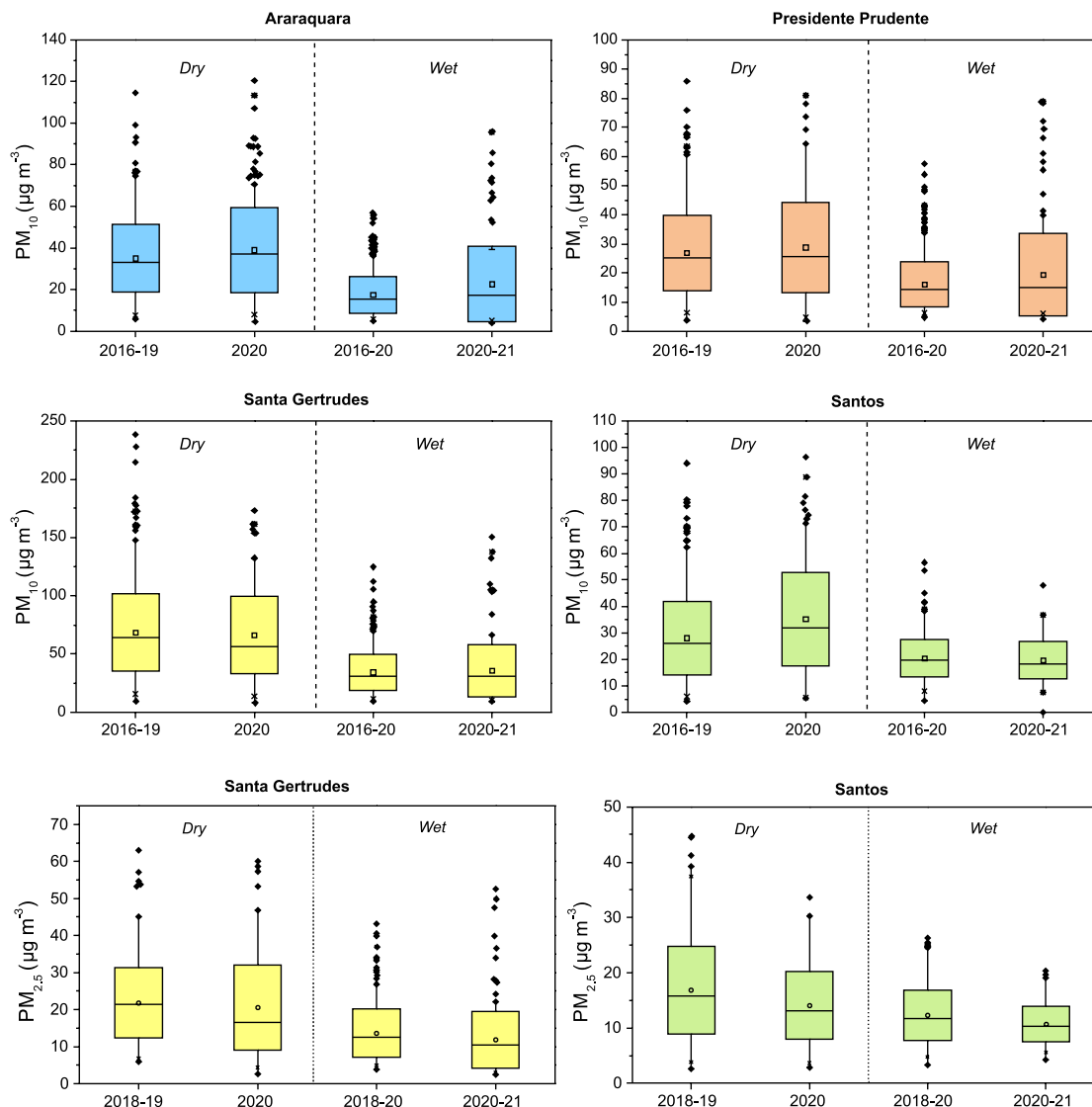


Figure 4. Concentrations of PM₁₀ and PM_{2.5} ($\mu\text{g m}^{-3}$) during the dry and wet seasons in the four cities studied, separated into pre-pandemic and pandemic periods. The whiskers plots show the concentrations for the mean (\square), median ($-$), outliers (\bullet), 25th and 75th percentiles, minimum, and maximum.

During the pandemic period, the concentrations of PM₁₀ in ARQ in the dry and wet seasons were higher than in previous years (Table 3), due to a greater number of fire spots in the years 2020 and 2021 (Table S5, SI section). The relevance of biomass burning as a source of PM was supported by the trends of increase of the PM₁₀ concentrations in the dry season and decrease in the wet season, which coincided with the number of fire spots. Although before the pandemic, downward trends in PM₁₀ concentrations were found in both seasons, when the data from the pandemic period were included, no trend was observed in the dry season, while a trend with a smaller decrease was observed in the wet season (Table 1), due to the higher concentrations of PM₁₀ during the pandemic period. Even though PM₁₀ emissions in both seasons were higher than in previous years, they showed negative correlations

with the social distancing index values (Table S3), which may have reflected lower vehicular emissions of particles. In PPE, the concentrations of PM₁₀, in the pre-pandemic and pandemic periods, were higher in the dry seasons compared to the wet seasons. In the dry season, combining the data from before and during the pandemic, a trend of increasing PM₁₀ concentrations was observed for PPE, with a greater increase before the pandemic (Table 1). In the wet season, before and during the pandemic, no trends were observed in the PM₁₀ concentrations (Table 1).

The PM₁₀ concentrations in the city of PPE were similar to those in ARQ. In the dry season, the concentrations for the pandemic period did not differ from previous years, while in the wet season they were higher (Figure 4 and Table 3). This behavior could be explained by the increase in the number of fire spots in 2020, at the end of the dry season and

Table 3. Mann-Kendall trends in PM₁₀, and PM_{2.5} concentrations, and the corresponding Sen's slopes (Qmed), for the cities studied, separated into dry and wet seasons, and comparison between the concentrations during the pandemic and pre-pandemic periods, using the Mann-Whitney test

Pollutant	City	Season	time interval	n ^a	Trend	Qmed	Mann-Whitney hypothesis
PM ₁₀	ARQ	dry	2016-19	718	downward ^c	-0.0052	2020 > 2016-19 ^c
			2016-20	900	no trend ^d	N/A	
		wet	2016-20	720	downward ^b	-0.0057	2020-21 > 2016-19 ^b
			2016-21	853	downward ^c	-0.0020	
	PPE	dry	2016-19	713	upward ^b	0.0082	2020 = 2016-19 ^d
			2016-20	896	upward ^b	0.0053	
		wet	2016-20	719	no trend ^d	N/A	2020-21 > 2016-19 ^c
			2016-21	884	no trend ^d	N/A	
	SGD	dry	2016-19	727	upward ^b	0.0204	2020 = 2016-19 ^d
			2016-20	908	upward ^b	0.0086	
		wet	2016-20	670	no trend ^d	N/A	2020-21 = 2016-19 ^d
			2016-21	852	no trend ^d	N/A	
STS	dry	2016-19	727	downward ^b	-0.0137	2020 > 2016-19 ^b	
		2016-20	910	no trend ^d	N/A		
	wet	2016-20	725	no trend ^d	N/A	2020-21 = 2016-19 ^b	
		2016-21	905	no trend ^d	N/A		
PM _{2.5}	SGD	dry	2018-19	235	upward ^b	0.0279	2020 < 2018-19 ^c
			2018-20	371	no trend ^d	N/A	
		wet	2018-20	339	downward ^b	-0.0061	2020-21 < 2018-20 ^b
	2018-21	507	downward ^b	-0.0032			
	STS	dry	2016-19	720	no trend ^d	N/A	2020 < 2016-19 ^b
			2016-20	903	downward ^b	-0.0022	
		wet	2016-20	721	downward ^c	-0.0015	2020-21 < 2016-20 ^b
	2016-21	905	downward ^b	-0.0023			

^an = number of mean daily concentrations; ^bp-value < 0.01; ^cp-value < 0.05; ^dnot applicable. ARQ: Araraquara; PPE: Presidente Prudente; SGD: Santa Gertrudes; STS: Santos; N/A: not applicable; PM₁₀: particulate matter with aerodynamic diameter ≤ 10 µm; PM_{2.5}: particulate matter with aerodynamic diameter ≤ 2.5 µm.

the beginning of the wet season, compared to the previous years (Table S5). Bearing in mind that no significant changes were observed in precipitation or wind speed and direction (Table S2). For both dry and wet seasons, a negative correlation was obtained between PM₁₀ and the social distancing index (Table S3), indicating a reduction in emissions of this pollutant as a result of social distancing. Another indication of this is that with the inclusion of data during the pandemic, there was a reduction in the slope of the trend of increase in PM₁₀ concentrations observed in the dry season before the pandemic. In the dry season, before the pandemic, the PM₁₀ concentration for 2020 did not differ significantly from the values for previous years, while the PM_{2.5} concentration was lower than previously and upward trends in PM₁₀ and PM_{2.5} were observed for SGD (Table 3). When the data for the pandemic period were included, PM₁₀ still showed an upward trend, with a smaller increase than before the pandemic, while no trend was observed for PM_{2.5} (Table 3). Among the cities studied, SGD showed the highest concentrations of PM₁₀ and PM_{2.5}

(Figure 4). Due to the upward trends in PM₁₀ and PM_{2.5} observed in the dry season before the pandemic, in SGD, increases in the amounts of these pollutants would have been expected in 2020. However, due to the pandemic, with social distancing and a slight decrease of activity in the ceramics industries, the PM₁₀ concentration for 2020 did not differ significantly from the values for previous years, while the PM_{2.5} concentration was lower than previously (Table 3). For both pollutants, negative correlations were obtained with the social distancing index (Table S3), which indicates that there was a reduction in emissions of these pollutants due to the latter. A notable feature of this city is that ceramics production is a relevant source of PM to the atmosphere, additional to vehicle emissions.⁴⁷ Vehicle emissions contribute more to PM_{2.5} than PM₁₀, so the concentrations of the former showed a greater impact of the pandemic.⁵⁵ When the data for the pandemic period were included, PM₁₀ still showed an upward trend, with a smaller increase than before the pandemic, while no trend was observed for PM_{2.5} (Table 3). The changes in ceramics

production and vehicle emissions could also explain these behaviors. It should be noted that although total ceramics production decreased slightly in 2020, compared to previous years, the wet production process continued to grow linearly. This type of processing is a more important source of coarse particles, compared to the dry process.⁵⁶

In the wet season, before the pandemic and with the inclusion of data for the pandemic period, there was no trend for the PM_{10} concentrations in SGD, while a downward trend was observed for $PM_{2.5}$ (Table 3). For 2020-2021, no significant differences in PM_{10} were observed, while $PM_{2.5}$ decreased. The absence of a trend in PM_{10} concentrations and the downward trend observed for $PM_{2.5}$ concentrations could be explained as follows: in the wet months, the emissions of particles from ceramics production were low, since the process of drying the materials on patios did not occur, and vehicle emissions were likely to be more important for these pollutant species.⁵⁷ Therefore, during the pandemic no significant differences in PM_{10} were found, while $PM_{2.5}$ decreased. The result for $PM_{2.5}$ was corroborated by the absence of correlation with the social distancing index, while a negative correlation was observed for PM_{10} (Table S3). This could also have been due to the increase in the wet production process in 2020. In the city of STS, the concentrations of PM_{10} in the dry seasons were higher than in the wet seasons, before and during the pandemic (Figure 4).

For the dry season during the pandemic period, PM_{10} concentrations in STS were higher than in previous years, however, lower concentrations were expected, given that there was a downward trend in PM_{10} concentrations (Table 3). The observed increase was related to the growth in port activities and the lower wind speeds during the pandemic period (Table S2). In the same season, there was no significant correlation between the concentration of this pollutant and the social distancing index (Table S3), indicating that the pandemic did not influence the emissions of this pollutant. This result was corroborated by the absence of any trend when data for the pandemic period were included (Table 3).

In the wet season, there was no significant difference between PM_{10} concentrations recorded before and during the pandemic (Table 3). This result was expected since there was no trend in the PM_{10} concentration (Table 3). On the other hand, a negative correlation was observed between the concentrations of PM_{10} and the social distancing index (Table S3). The concentrations of $PM_{2.5}$ were also higher in the dry seasons than in the wet seasons, both before the pandemic and during the pandemic period (Figure 4). During the pandemic period, the $PM_{2.5}$ concentrations in STS were lower than in previous years, in both seasons

(Table 3). Moreover, with the inclusion of data for the pandemic period, downward trends were observed for both seasons (Table 3). It should be noted that in the dry season, the decrease was correlated with the social distancing index (Table S3), and that it would not have occurred in the absence of social distancing, considering the trend in the concentrations from 2016 to 2020. However, in the wet season, the concentrations followed the trend observed in previous years (Table 3), with no significant correlation with the social distancing index (Table S3). With the inclusion of data for the pandemic period, downward trends were observed for both seasons (Table 3), in agreement with the correlation observed between $PM_{2.5}$ and the social distancing index.

Comparison with results obtained in other Brazilian studies

Most of the previous studies carried out in Brazil compared the concentrations of pollutants recorded at the beginning of the pandemic (March to May) with those recorded in the same period of previous years.^{2,10,13,15,58-61} In these studies, the mean concentrations of air pollutants during the pandemic were compared with the mean concentrations observed in the same period of previous years, using statistical methods such as the Mann-Whitney test, Kruskal-Wallis test, and Pearson correlation.^{2,10,11,13,15,58-60,62,63}

As in the present study, other works carried out in Brazil reported reductions in NO_2 concentrations due to restrictions imposed during the pandemic period.^{2,10,11,13,15,58,60-62,64} Therefore, the reduction in NO_2 concentrations was a homogeneous behavior in the different cities studied. In general, for concentrations of O_3 , PM_{10} and $PM_{2.5}$, a more heterogeneous behavior than NO_2 was observed, varying according to the city.^{2,13,15,59,63} These differences are mainly due to changes observed in meteorological conditions, in removal processes and in emissions by industrial activity, as well as by fires.^{15,58-61}

Regarding O_3 , as previously discussed in this study, different behaviors were observed in the wet season and in the dry season. This type of evaluation had not been performed in previous studies. Moreover, the reported increases in O_3 concentrations were mainly due to lower removal of this pollutant due to lower NO_2 concentrations.^{15,58}

In the city of Rio de Janeiro, O_3 concentrations increased due to an increase in the ratio of non-methane hydrocarbons to NO_x , or reductions in NO_2 concentrations.^{2,60} Similar behavior was observed in the cities of PPE and ARQ. At the same city, Rio de Janeiro, PM_{10} concentrations were reduced only in the first week of lockdown, then there was

an increase in concentrations, which was attributed to the resumption of industrial activity.⁶⁰ This was also observed in the present study in the city of SGD, where the resumption of industrial activity led to an increase in PM₁₀ and PM_{2.5} concentrations.

Conclusions

The findings showed that social distancing led to decreases in NO₂ and PM_{2.5} concentrations, especially in cities where the main source of emissions was vehicular (PPE and ARQ). However, in SGD, there was an increase in NO₂ concentrations, which could be explained by activities in the ceramics industries, especially an increase of the wet production process. It is notable that in STS, where emissions were mainly from the port activities, the reduction in NO₂ concentrations was smaller than observed in PPE and ARQ. In SGD and STS, reductions of PM_{2.5} emissions were mainly due to social distancing, which reduced vehicular traffic.

The O₃ concentrations showed behaviors that differed according to city, as expected. In ARQ and STS, increases in O₃ concentrations were observed, indicating that a decrease in NO₂ concentrations led to higher O₃ concentrations, due to lower O₃ removal. In the case of STS, the slight increase observed in the dry period was due to the reduced dispersion associated with lower wind speeds. Reductions in O₃ concentrations were also observed due to O₃ production being limited by the concentration of VOCs, rather than by NO_x as in the previous case.

The PM₁₀ concentrations either followed the previous trends or increased. This last was due to an increase in the number of fire spots, and/or reduced dispersion by the wind. It should be noted that if the pandemic had not occurred, PM and NO₂ emissions could have been even higher, due to the increase in biomass burning, mainly in the regions of PPE and ARQ.

The results obtained here showed that there was a reduction in the concentrations of most of the pollutants analyzed during an entire year within the scope of the mobility reduction policy, however the emissions from the burning of biomass led to an increase in the concentrations of particulate matter. Therefore, it is of fundamental importance that new policies aimed at reducing emissions improve the regulation and control of biomass burning.

Supplementary Information

Supplementary information (Tables S1-S5 and Figure S1) is available free of charge at <http://jbcs.sbc.org.br> as PDF file.

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Author Contributions

Jonatas S. Carvalho was responsible for the conceptualization, formal analysis, investigation, visualization, writing-original draft, review and editing; Gabriel M. Ferraz for the conceptualization, formal analysis, investigation, visualization, writing-original draft, review and editing; Hugo L. I. Betim for the conceptualization, formal analysis, investigation, visualization, writing-original draft, review and editing; Rita de Kássia S. do Nascimento for the conceptualization, formal analysis, investigation, visualization, writing-original draft, review and editing; Caroline Scaramboni for the conceptualization, formal analysis, investigation, visualization, writing-original draft, review and editing; Roberta C. Urban for the conceptualization, formal analysis, investigation, visualization, funding acquisition, supervision, writing-original draft, review and editing.

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