



## Sulfur dioxide: Behaviour and trends at the industrial city of Araucaria/Brazil

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### ABSTRACT:

**Introduction:** It is known that the levels of sulfur dioxide have a close relationship to the industrial emissions and they are also related to the climate, but it's dispersion its mainly caused by the wind dynamics. The goal of this paper is to analyze the behaviour of this compound in two stations at Araucária city, in the state of Paraná, Southern Brazil, the city which is located in the industrial zone of Curitiba, the capital of the state.

**Materials and methods:** Measurements of SO<sub>2</sub> were done in two monitoring stations in different regions of the city; one was located in the industrial zone and the other one was a residential zone. A basic statistical treatment was performed the R, which includes the construction of time series plots, histograms and analysis of SO<sub>2</sub> levels and wind direction. The Theil-Sen trend estimator was applied on both time series in order to understand the past and present behaviour of the pollutant.

**Results:** The analysis of wind and SO<sub>2</sub> levels revealed a straight relationship between the location of the industrial zone and air pollution. The trend calculation revealed for both stations, similar situations, in which the sulfur dioxide levels are decreasing.

**Conclusion:** During this research, it was possible to notice that there is an environmental problem on the suburbs of the city with a direct result from urban growth and the lack of planning for the suburban areas. This study reinforces the importance of urban planning policies as well as monitoring and fiscalization.

### Introduction

According to the World Health Organization (WHO), air pollution is responsible for about 3 million deaths annually. The same report pointed out that 90% of the world's population breathes air that does not comply with the WHO Air Quality Standards [1]. The sulfur dioxide levels are responsible for such statistics. It's variability was significantly correlated with ischemic stroke,

bronchial asthma and chronic bronchitis [2-5].

High levels of sulfur dioxide were found in the city of Araucária, in the state of Paraná, Southern region of Brazil (Fig. 1), with an amount of 137.452 habitants which is considered as a part of industrial zone of Curitiba, the capital of the state since the 70's. During the military government of Brazil, a state program to industrialize Curitiba's region was implemented by the government. By

that time, the federal government offered lots of fiscal benefits to industries to settle in Araucária, making this city become one of the richest but also most polluted in the south of Brazil [6].

In order to monitor and fiscalizates the industries, the government keeps at this moment three air quality monitoring stations in Araucária. Two of them were located in the industrial zone, and the other in a residential location close to the industries. The establishment of a monitoring network is pointed out as the most efficient way to manage environmentally a territory in terms of air quality issue [7].

The Brazilian standards for air quality were established in 1900 by the National Environmental Council Resolution 003/90 which were never updated again. According to this resolution, the minimum standard for sulfur dioxide is  $365 \mu\text{g}/\text{m}^3$  for a 24 h average.

It is known that the levels of sulfur dioxide have a close relationship to the industrial emissions and they are also related to climate variability, but it's dispersion is mainly caused by wind dynamics [8]. The goal of this paper is to analyze the time behaviour of this compound in two stations at Araucária; one was located in the industrial zone and the other was in a residential zone. The distance between both zones is around 2.5 km. Ten years of hourly data (2005-2014) were analyzed from the CSN (industrial) and the Assis (residential) stations. The concentrations were analyzed using different time scales (annually, monthly, weekly and hourly), and their correlation with wind direction was also investigated. After all, a long-term trend calculation was elaborated in order to observe the general behaviour of this pollutant in the study area.

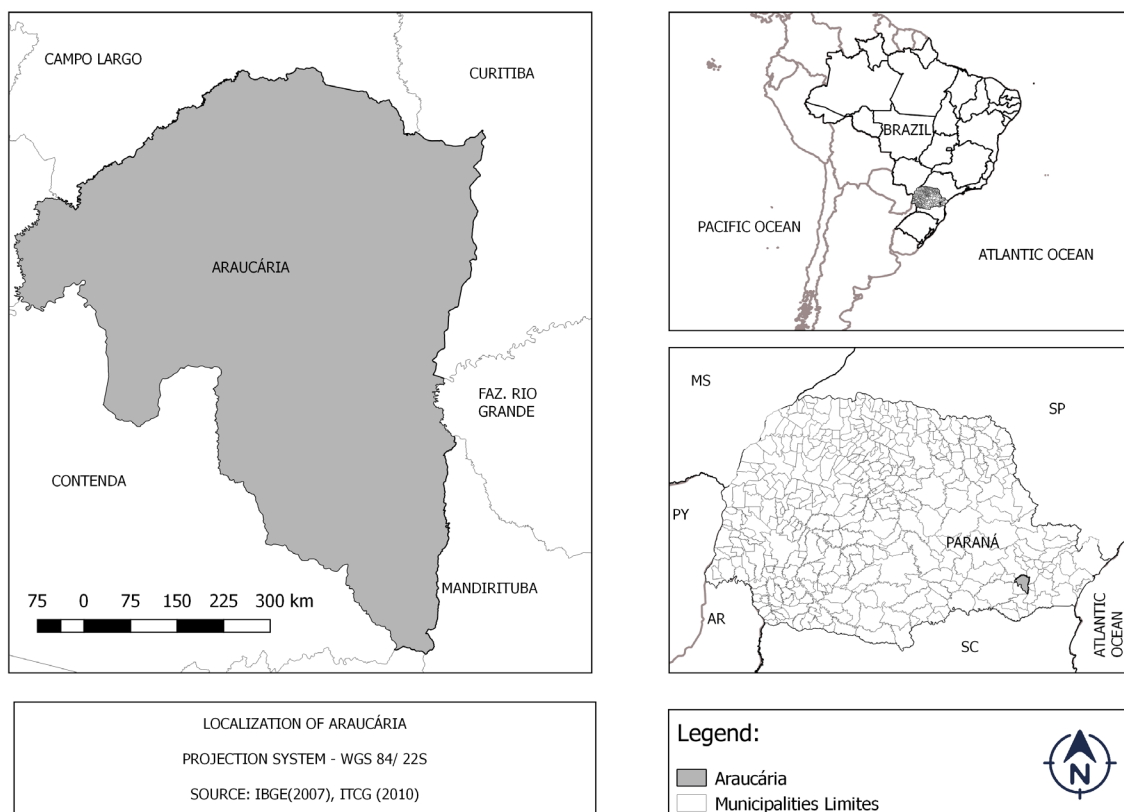


Fig. 1. Location of Araucária.

## Materials and methods

### Data collection and study area description

Measurements of SO<sub>2</sub> were done in two monitoring stations in different zones of the city of Araucária. The “Assis” station represents a typical low dense residential site with low vehicle traffic but still close to the big industrial area of the city (Fig. 2). On the other hand, the “CSN” station is located inside the industrial zone, close to a highway, that connects Curitiba to the southern states of Brazil, what results in a more intense traffic (Fig. 2).

The data was collected using an hourly scale. The time series analyzed in this paper starts on January of 2005 up to December of 2014, resulting in a ten-years time series. This period was chosen because that is the period in which the stations were installed.

### Statistical analysis

After data collection, a basic statistical treatment was made using the R, which includes the construction of time series plots, histograms and analysis of SO<sub>2</sub> levels and wind direction.

The Theil-Sen trend estimator was applied on both time series in order to understand the past and current behaviour of the pollutant. Taking into account that pollutant data are non-normal and have non-constant error variance, this method is considered to be suitable for using in this case [9]. Thus it is commonly used to describe trends and behaviours of pollutants [10-13]. Furthermore, graphics with trend lines by season and wind direction were created in order to demonstrate a more detailed behaviour of sulfur dioxide in the present location.

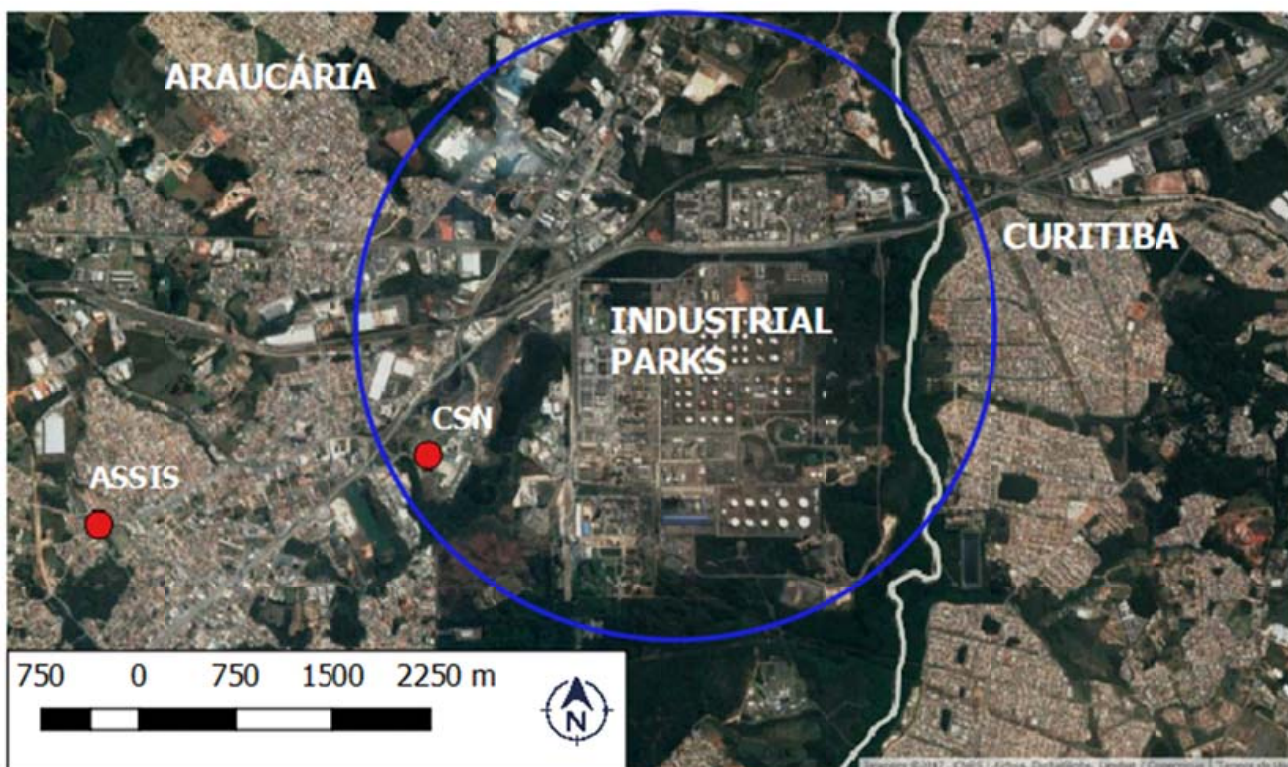


Fig. 2. Location of the monitoring stations used in this study

**Results and discussion**

**Time series analysis**

Fig. 3 reveals the time variation for both stations of this study. Both stations present different SO<sub>2</sub> concentration levels. While at Assis the maximum concentration is 231.3 ppb in 2012, at CSN the maximum value reaches 893.4 ppb in 2005. Both time series present few days in 2010 without data, in which, according to IAP (Environmental Institute of Paraná – responsible for the maintenance of the monitoring stations) they presented malfunctions.

It was noticed that the oscillation at a yearly scale are similar, in both situations the high values occurred during the summer season (December, January and February). Meanwhile during the winter, they reached their lowest values.

On the other hand, hourly time oscillations are different. It was noticed that, at Assis there are two peaks during the day time, related to the traffic rush hours in the morning and in the afternoon. Meanwhile, at CSN, there is only one pronounced peak, at night.

The difference between both places is also described by Fig. 4. The histograms reveal a higher frequency of episodes in which the pollutant levels exceeded the national standards at CSN station. At Assis station, the frequency of records higher than 10 ppb were not significant, meanwhile at CSN, it was relatively more common. The red line in the histograms indicates the national standard values for SO<sub>2</sub>

**Wind direction and sulfur dioxide levels**

According to one of the main responsibilities for dispersion and concentration of air pollution, the analysis of wind direction and SO<sub>2</sub> levels revealed a straight relationship between the location of the industrial sites and air pollution.

On both places, the west and north wind directions are those with the lowest frequency of high values. Meanwhile, east and southeast directions, the regions in which the industrial emissions are more intense, are those with the highest frequencies of high pollution levels (Fig. 5).

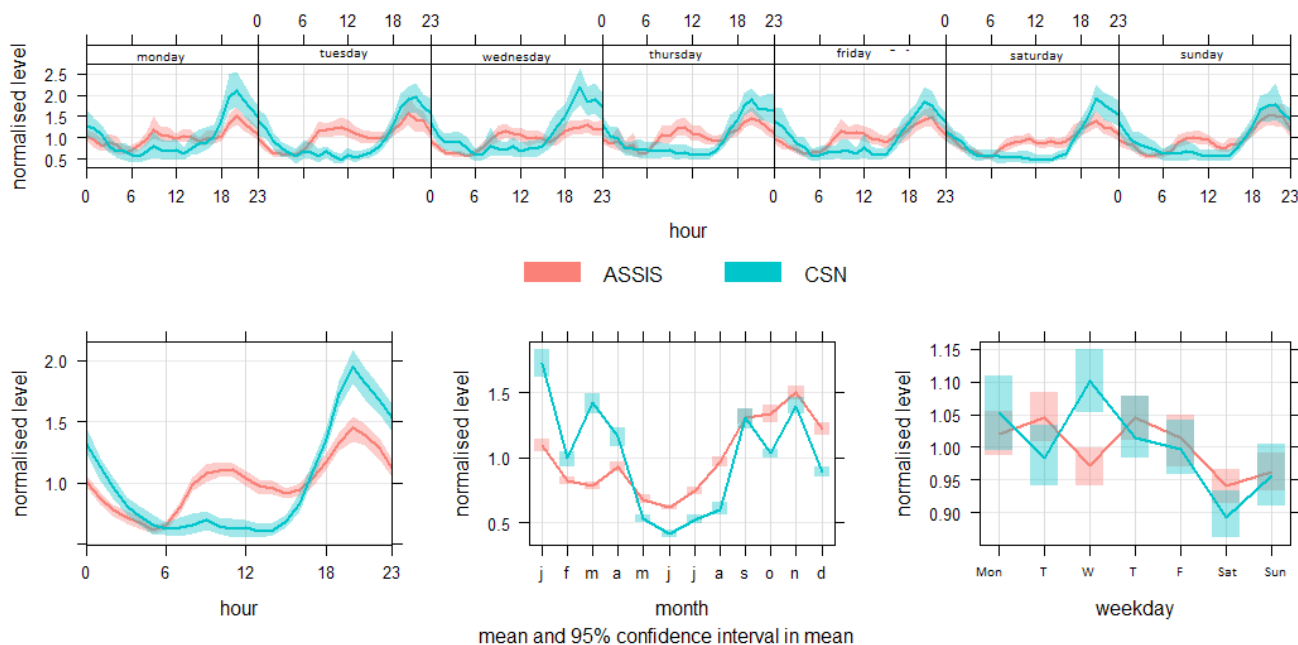


Fig. 3. SO<sub>2</sub> Time variation



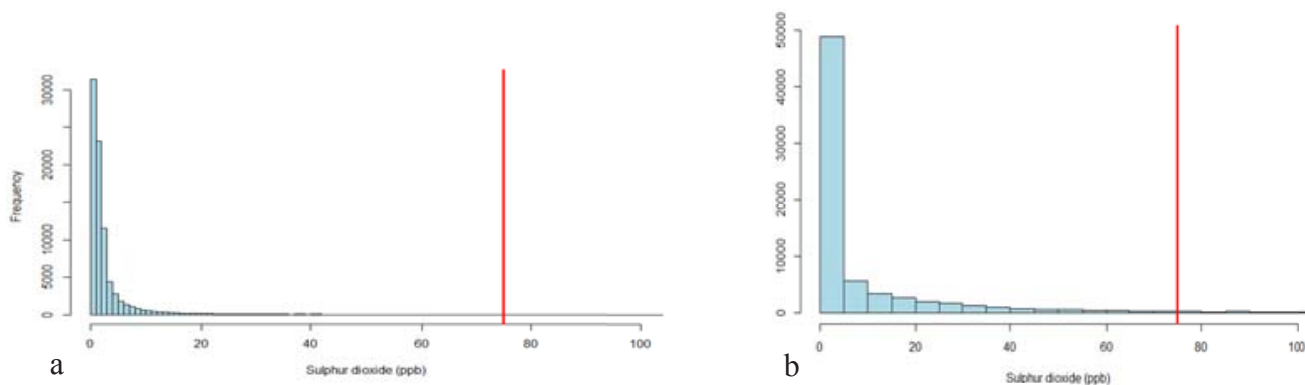


Fig. 4. SO<sub>2</sub> Histograms. A – Assis; B – CSN

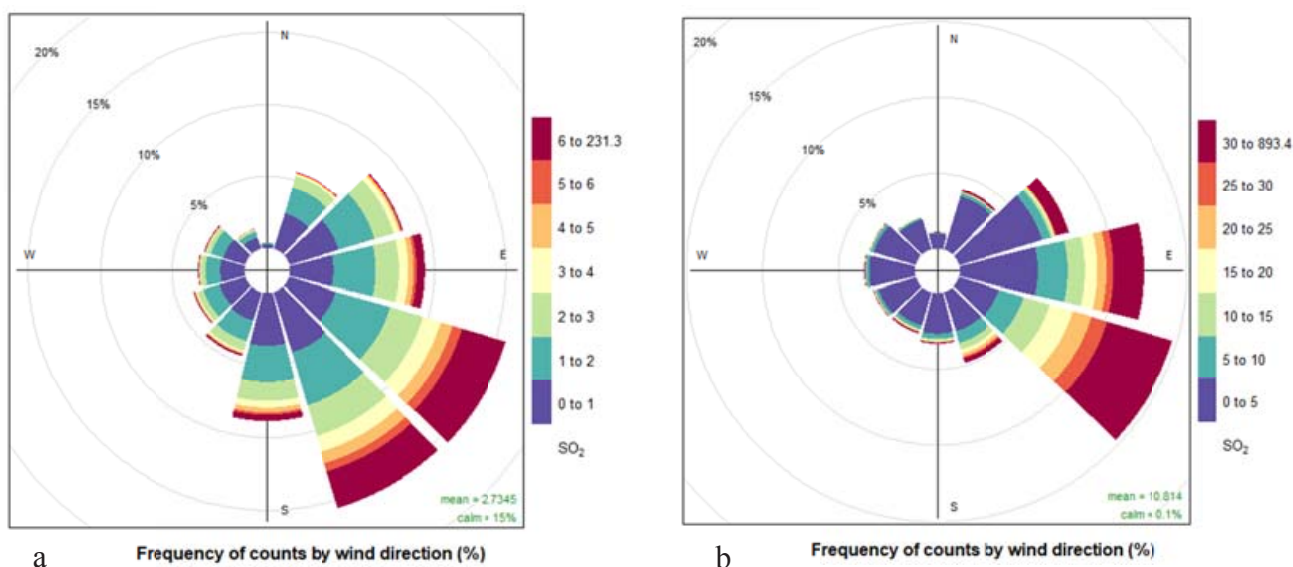


Fig. 5. SO<sub>2</sub> Pollution Rose ( a : Assis; b :CSN)

A recent study analyzing wind direction on a 10-year data time series revealed that east and south-east are the most common direction of winds in this region [14].

In general, this dynamics occurred during the entire time series analyzed in this study. The concentration of high values from southeast direction

at CSN is more expressive when it is compared to Assis. In a second moment, due to the distance to the industrial zone, the pollution frequency by wind direction is slightly more diffused, still, the highest values came from the southeast (Fig. 6) but generally, it is noted that no significant changes occurred during the time series.

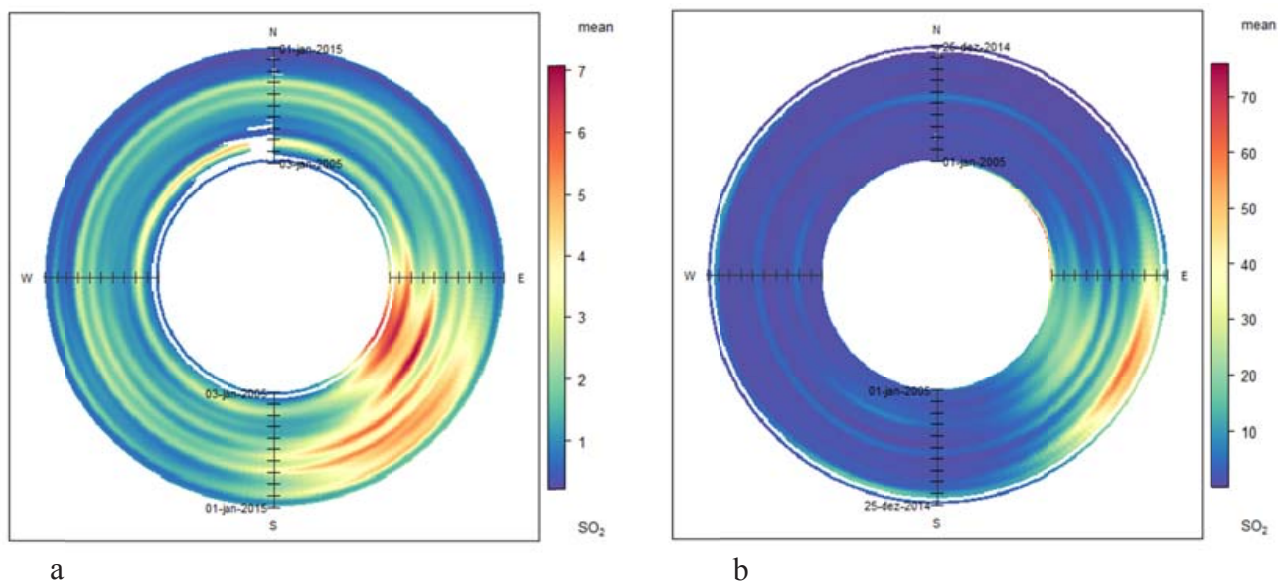


Fig. 6. SO<sub>2</sub> Time Series Pollution Frequency by Wind Direction (a: Assis; b: CSN)

Fig. 7 reveals the same dynamics relating wind direction and SO<sub>2</sub> average values by month. At Assis, it was noticed that there are no significant changes throughout the year. High concentrations were always found on southeast winds, in July and August. Meanwhile at CSN, it was possible to verify a slightly change on pollution high val-

ues frequency. In January, the east and northeast are the directions in which the highest values were registered, during the year, this situation changes. After August it was possible to verify that the southeast is again the situation in which the highest values are registered.

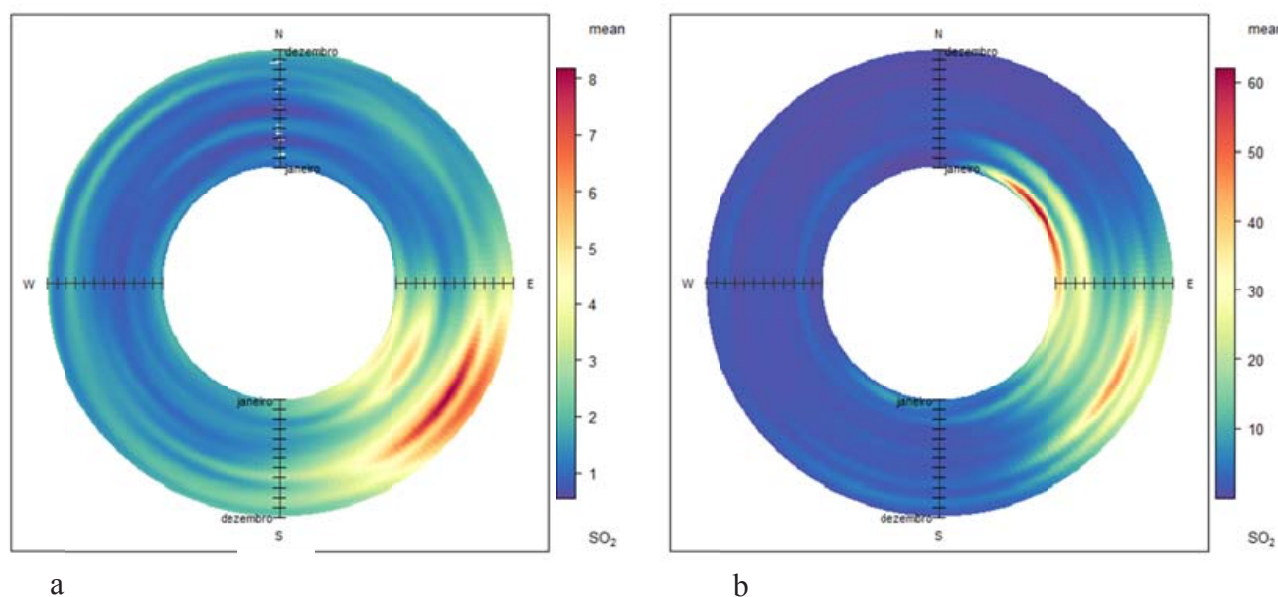


Fig. 7. SO<sub>2</sub> monthly pollution frequency by wind direction. (a: Assis; b: CSN )

The frequency of sulfur dioxide by wind direction during a day complements the current analysis. Fig. 8 reveals high values at CSN after 6 pm concentrated from east/northeast/southeast directions. Meanwhile, the highest values at Assis started after noon and always the highest values were registered during southeast winds, since, at this station the effects of traffic emissions are more significant.

### Long-term trends

The Theil-Sen trend estimator revealed for both stations, similar situations. On Fig. 9a, the Assis oscillation shows a more expressive decline in  $\text{SO}_2$  after 2007. Fig. 9b reveals the trend oscillation at CSN. Due to its higher values, the trends on CSN oscillate more than at Assis, revealing a fast decrease from 2005 to 2006, followed by a slightly increase until 2013, from where a new

decrease starts. In general, it is noticed that in both situations the sulfur dioxide is decreasing and the intra annual oscillation at Assis is more regular than at CSN.

### Seasonal trends

Observing the same estimator by season, the general situation is similar. At Assis (Fig. 10), it was possible to notice a regular decrease trend during spring and summer seasons. However, the situation during fall and winter changes a little when it is possible to observe a more irregular trend. During fall, it is possible to verify an increase in  $\text{SO}_2$  levels in 2010, followed by a decrease until 2013, when it was found a minor increase in this pollutant levels. During winter, there was a major peak on  $\text{SO}_2$  levels on 2012, followed by a high decrease.

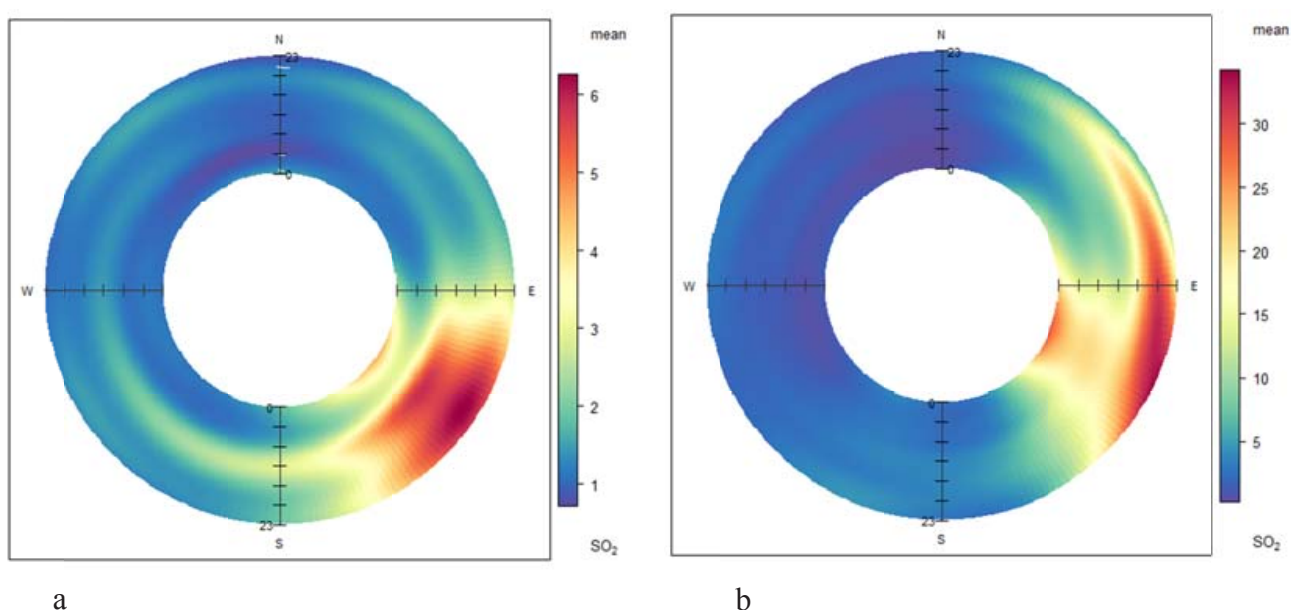


Fig. 8.  $\text{SO}_2$  hourly pollution frequency by wind direction (a: Assis; b: CSN)

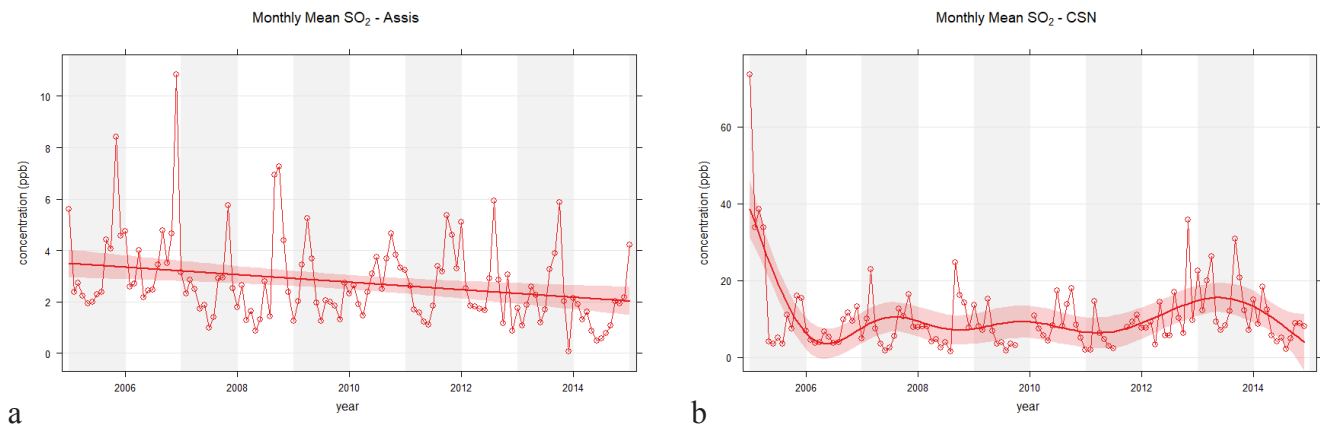


Fig. 9. SO<sub>2</sub> Theil-Sen trends 2005-2014 (a: Assis; b: CSN)

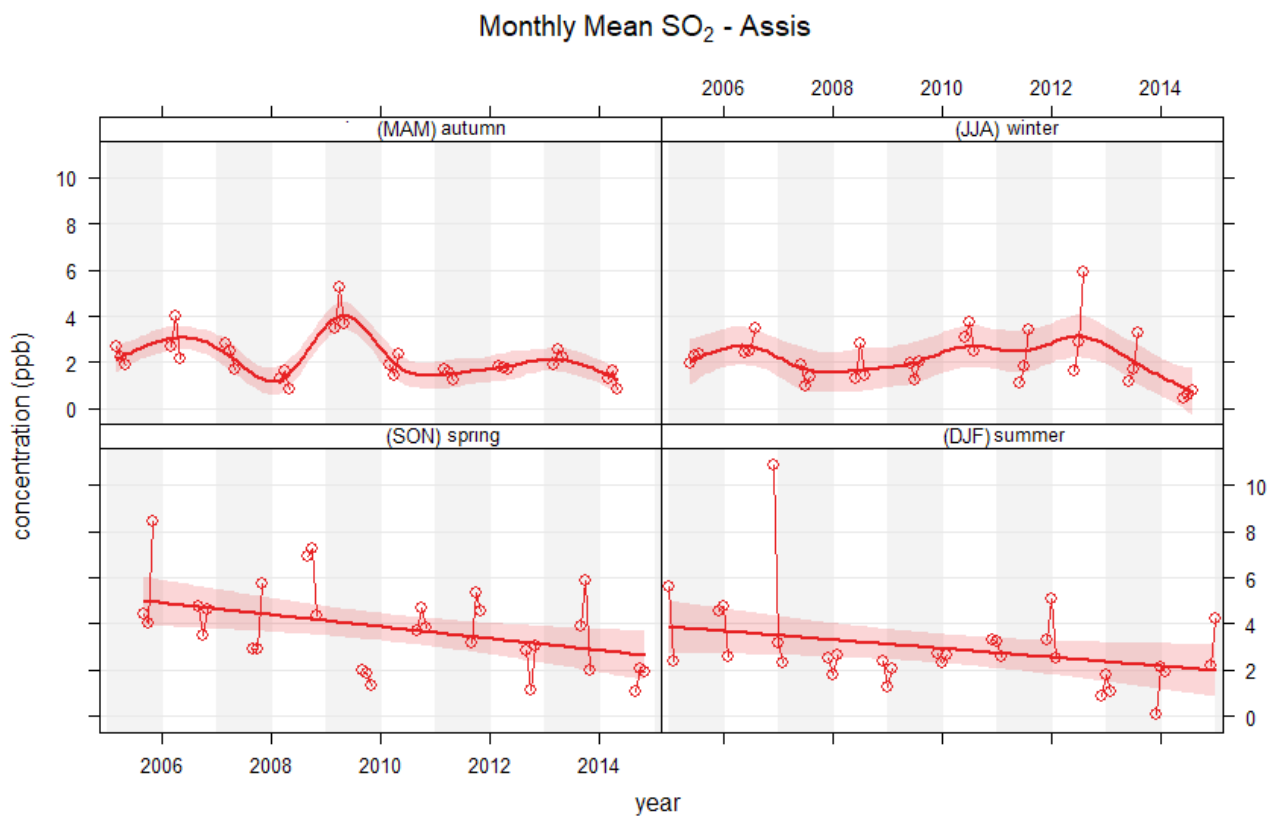


Fig.10. SO<sub>2</sub> Theil-Sen trends 2005-2014 by season at Assis

At CSN (Fig. 11), the trends by season are more regular. In both, fall and summer, it revealed a major decrease on the pollutant levels after 2005, that remains low until 2013 when it got a little bit

higher. During spring and winter, the trend oscillation is more irregular, there are peaks in 2009 (spring), 2011 (summer) and 2013 (spring and summer) followed by a regular decrease.



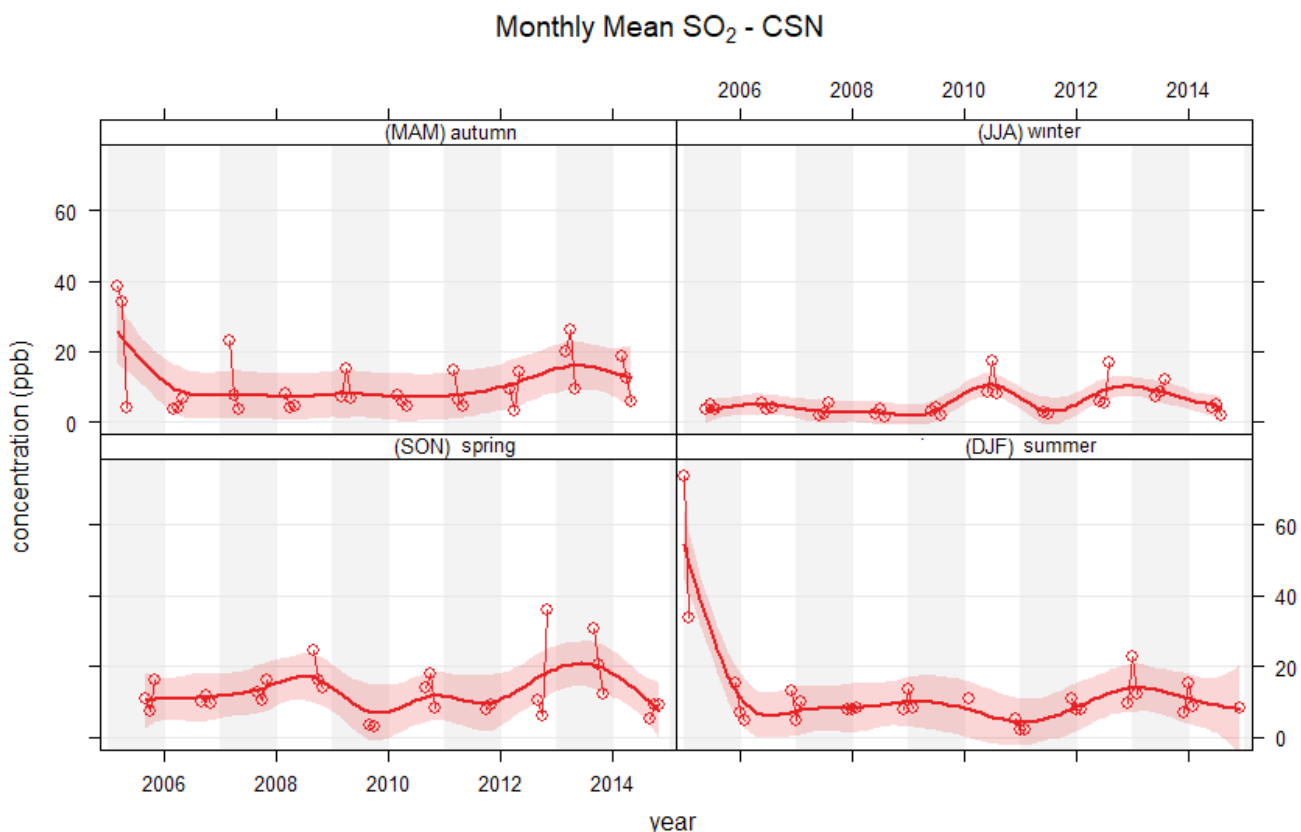


Fig. 11. SO<sub>2</sub> Theil-Sen trends 2005-2014 by season at CSN

**Wind direction trends**

Figs. 12 and 13 reveal the pollution trends on both sites by wind direction in order to detect any changes on the relationship between wind direction and SO<sub>2</sub> levels.

According to Fig. 12, it was detected a decrease in pollution levels only on northeast, east and southeast wind directions at Assis. For the rest of the wind direction quadrants, no trends were detected.

Fig. 13 reveals the same analysis at CSN station. The results highlights negative trends at east and southeast directions, both after an increase in 2013, quite similar to what was observed on Fig. 9b. For the other wind directions, no trend was detected.

**Conclusion**

The current study revealed the behaviour and trends of SO<sub>2</sub> concentration levels in Araucária, one of the most important industrial cities of Brazil.

Comparing the results on both stations studied, the spatial factor is understandable since both stations are close to the industrial park. However, since CSN is closer to the industrial parks than Assis, the pollution cycle on this station responds more to the effect of the industrial emissions.

At Assis, besides the industrial emissions, traffic intensity also influences on pollution oscillation as we can notice by the double peak during a day. This factor is also verified by the wind direction analysis. It was noticed that at CSN the relation-

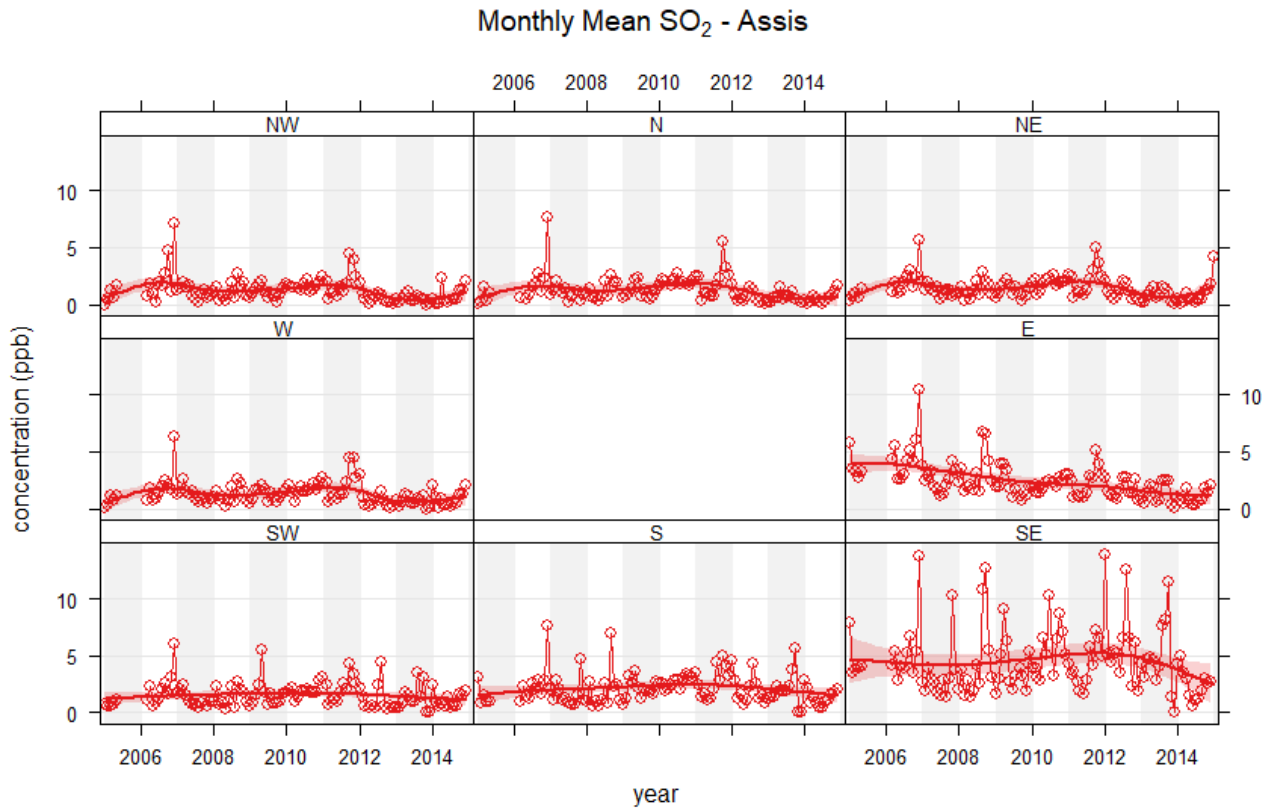


Fig. 12. SO<sub>2</sub> Theil-Sen trends 2005-2014 by wind wirection at Assis.

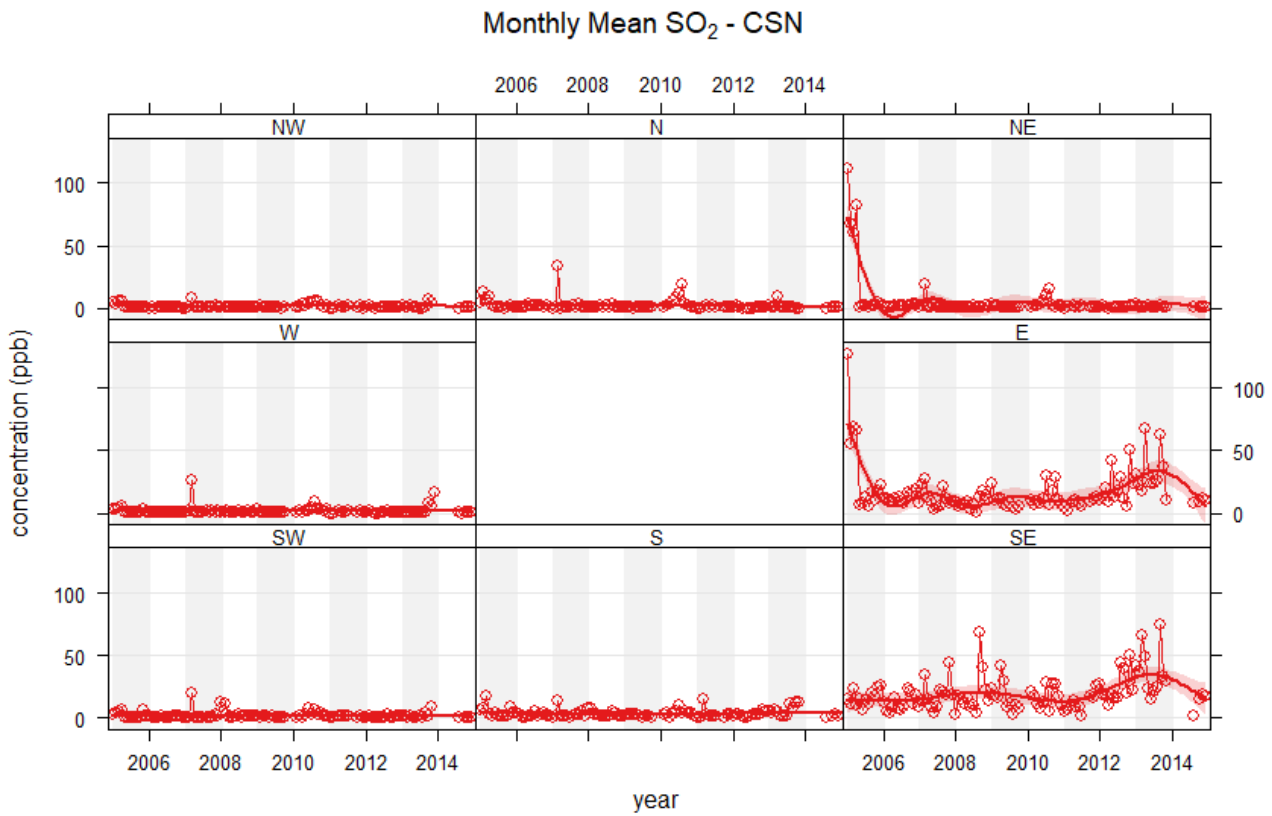


Fig. 13. SO<sub>2</sub> Theil-Sen trends 2005-2014 by wind direction at CSN

ship between wind direction and SO<sub>2</sub> concentrations is more direct, what was also shown on the trend levels by wind direction.

At Assis, it was noticed a relationship between wind direction and SO<sub>2</sub> concentrations but in a more diffuse way, well represented by Fig. 6.

In both stations, the east quadrant was the one with the highest pollutant values, since it is on this direction, at both stations, that the industrial parks are located.

Also, according to wind direction tests, during the entire time series, no strong changes on the relationship between wind and pollution were noticed, what reveals that the emission source on this location remains the same on both sites. That counts also for the hourly scale. At monthly scale, a low change was observed at CSN, revealed by Fig. 7b, since it was observed that during January a higher frequency of high pollution values originated during northeast winds, and after the winter, the station with the highest average levels, it changed to east and southeast winds.

This change during the year did not occur on the Assis station but it can be explained by the atmospheric circulation and the location of the CSN station. Industrial parks are found at northeast, east and southeast of CSN, and during the winter season, due to the entrance of polar masses, the prevalent wind direction is the southeast.

On both stations a decrease in pollutant levels was revealed. Even though, there are not new policies on industrial emissions published in the last ten years, and the industrial park increased on area, a huge difference was verified during the analyzed period.

Fig. 14 makes a quick comparison between using a satellite image from 2005 and one from 2015.

To support that, the Paraná's Institute of Economic and Social Development [15] revealed an increase of 63.2% in the number of industries

with pollutant emissions in 2012 in comparison to 2003.

Also, it was noticed that not only the industrial park grew but also the urban area surrounding it, the car/habitant index also increased. According to the National Traffic Agency [16], the number of cars by habitant in Curitiba's urban perimeter was 0.48 car/hab. in 2003, in 2013 this index reached 0.77 car/hab.

These data do not support the decrease of SO<sub>2</sub> on the last years, what suggest the need of a further analysis involving the record track of emission inventories and the specific origin of the compounds.

Curitiba's urban core, where Araucária is located, is internationally famous for its urban planning. However, during this research it was verified that there is an environmental problem on the suburbs of the city that is a direct consequence of urban growth and the lack of planning for the areas distant from the city center.

In the 70's, when the first industrial parks were constructed in this city, there was already a large neighborhood at west of this planned area. Knowing the wind dynamics of the region, in which the east winds are predominant, it should be expected high values of pollution concentrations on the existing residential areas. That is why the industrial areas of Araucária should not be planned to be developed in this specific area.

Furthermore, Fig. 14 reinforces the urban growth surrounding the industrial area. Knowing the problems with air pollution in this situation, a stronger urban land use policy should be adopted to avoid urban sprawl towards those areas.

Even though the current data current and trends reveal substantial decreases on the SO<sub>2</sub> for the last ten years, new and more effective policies have to be adopted not only at Curitiba but also for the entire country.





Fig. 14. Araucária Industrial Parks 2005 and 2015. Source: Google earth



Studies such as this, reinforce the importance of urban planning policies but also monitoring and fiscalization as well. As mentioned, the Brazilian standards on air quality were defined in 1990, and since then they were never updated. Pollutants such as PM<sub>2.5</sub> have no standards in Brazil, and the other reveal higher concentration standards when compared to the US or European legislation.

Besides that, it is important to emphasize the lack of air quality monitoring stations in the Brazilian cities and the difficulty to find large time series data. In terms of time, the oldest monitoring data for Curitiba's urban core starts at 2003. A larger amount of data could make possible to improve the analysis in terms of trends and comparison with urban growth. Also, in further analysis, a better use of remote sensing data can reinforce the importance of the urban planning in this field, and other pollutants such as NO<sub>2</sub> TSP could also be analyzed, reinforcing the studies on such area.

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### Competing interests

The authors declare that there are no competing interests.

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### Ethical considerations

Ethical issues have been completely observed by the authors

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