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Noise and ozone continuous monitoring in an industrial urban area of northeastern Portugal

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Abstract. The major environmental pressures associated with urban centers are noise and air pollution, making its monitoring of utmost importance to evaluate and reduce the exposure of the population to these environmental risk factors. In this study, continuous monitoring of sound pressure levels, ozone, nitric oxides, carbon monoxide concentrations, and local meteorological variables were performed during the winter and spring months of 2019 at the Mirandela industrial park. Ozone and nitric oxide levels followed a characteristic daily cycle, consistent with the diurnal evolution of radiation and the intensity of the main air pollution sources prevailing in the local. Hourly ozone levels were highest in July, reaching magnitudes of approximately 80 ppb. Ozone concentrations in the industrial park had a strong local influence, mainly related to the local nitric oxides emissions. The results also showed high influence of meteorological parameters on ozone production, especially during daytime. Regarding noise, typical daily and weekly patterns were observed, and sound pressure levels were compatible with those defined for mixed zones according to the Portuguese General Noise Regulation.

Keywords: Air quality, Noise, Monitoring, Industrial Parks.

1 Introduction

The rapid urban expansion, associated with a high population growth rate over the last centuries, tends to influence and modify various environmental aspects, producing impacts on the air, water, soil, and biodiversity [1–4]. Under such circumstances, it is evident that the increase in urban traffic, expansion of industrial zones and suppression of vegetation are the main degradation factors of urban air quality and local meteorological changes [5]. In addition, a large part of the population is exposed to different levels of environmental noises, capable of producing diverse effects on human health and well-being [6]. Therefore, air pollution and environmental noise are the two major environmental pressures associated with decreased quality of life in cities [7].

Air pollution is defined as a condition where one or more substances are present in the atmosphere at concentrations above normal ambient levels and particularly during a sufficiently long period to produce adverse effects on the health of humans, animals, and plants, or to cause material damage [8, 9]. According to the European Directive, noise corresponds to any unwanted sound or set of sounds that cause annoyance or may have an impact on human health, emitted by human activities, such as road traffic, rail traffic, air traffic and industrial sites [10].

Several studies emphasize the adverse effects of these components on human health [7, 11, 12], including respiratory and heart diseases [13, 14], causing annoyance and decreasing the cognitive ability [15, 16]. Thus, one of the great challenges of modern cities managers is providing quality of life to their inhabitants by improving the urban environment. To achieve this purpose, the monitoring of noise levels and air quality are extremely important to assess environmental risks, as well as to maintain or improve the environmental quality in urban centers [15, 17].

The main aim of this study was to study ozone, carbon monoxide, nitrogen oxides, and noise levels in the industrial park of Mirandela - Portugal. Since ozone is a very unstable secondary pollutant, its presence in the troposphere is partially related to the transport from the stratosphere, but the main contribution is its photochemical production, which occurs through the oxidation of hydrocarbons and carbon monoxide in the presence of nitric oxides and solar radiation [18]. For this reason, nitric oxides, carbon monoxide and, meteorological parameters were monitored in this research.

For the study, a monitoring system composed by one weather station, three gas analyzers and one noise sensor were installed in the Mirandela industrial park. The data collected during the winter and spring months of 2019 was used to correlate the different variables monitored with ozone production, and the system allowed further monitoring of the daily noise levels and the identification of the week noise profile. In the next sections of this paper a brief characterization of the industrial park, the methodological details of the study and the main air quality and noise results are presented and discussed.

2 Methodology

2.1 Industrial Park of Mirandela - Brief Description

This study was carried out in the urban industrial park of Mirandela ($41^{\circ}29'N / 7^{\circ} 9'W$), located in the northeastern region of Portugal, in the region known as Trás-os-Montes. The Industrial Park has an area of 33 hectares with 97 lots distributed to different industrial and commercial sectors. It comprises approximately 65 companies of different typologies such as sausage and granite factories, oil mills, carpentry, locksmiths and car repair shops. The Industrial Park is mostly surrounded by rural areas with olive trees plantations and open grassy spaces and is bounded by roadways to the west and south. The Mirandela downtown is southwest of the industrial Park.

2.2 Monitoring and Analyses

The air quality monitoring started in December 2018, with the collection of hourly data for carbon monoxide (CO), nitrogen oxides (NO, NO₂, NO_x) and ozone (O₃). All these gaseous pollutants were monitored according to the reference methods described by the Directive 2008/50/EC, using three gas analyzers: one ozone analyzer HORIBA APOA-370 (non-dispersive ultra-violet-absorption), one nitrogen oxides analyzer HORIBA APNA-370 (chemiluminescence) and one carbon monoxide analyzer HORIBA APMA-370 (non-dispersive infrared absorption). Noise monitoring started in February 2019 using the CESVA TA120 noise sensor, with measurements taken every minute. The noise sensor has class 1 accuracy according to IEC 61672-1 and was deployed outdoors due to the weather protection cover. In addition, a meteorological station was used to characterize the prevailing local weather conditions, measuring the direction and wind speed, solar radiation, temperature, relative humidity, and precipitation. The gas analyzers, weather station and, noise sensor showed in Figure 1 have the capacity to send data remotely via GPRS system, enabling the data to be accessed through a remote server.



Fig. 1. Set of the monitoring equipment used in the study: (A) Weather station; (B) air intake; (C) smart noise sensor; (D) gas analyzers container.

Based on the solar radiation data, the days were divided into daytime (8:00-19:00) and nighttime (20:00-7:00) to determine the correlation coefficient between the variables and the local and regional contribution to the prevailing O_3 levels. These separate timeframes were chosen considering the ozone formation and depletion mechanisms that have a strong dependence on solar radiation. It should be mentioned that in this study it was adopted the Coordinated Universal Time (UTC).

Concerning noise, a daily and weekly profile were evaluated based on hourly average data observed during the spring months. Additionally, the noise assessment was based on the Portuguese General Noise Regulation (RGR) (Decree-Law No. 9/2007 of 17 January). The RGR does not set noise limits to industrial parks itself, but any activity located in an industrial park has to comply with criteria established in RGR for sensitive receivers (i.e., spaces where people live or stay) near industrial areas. The criteria set for those receivers depend on the classification established in the municipal master plan for the area where they are located. These areas can be classified into sensitive and mixed zones. The RGR defines sensitive zones as areas for residential use, schools, hospitals or similar, and recreational or leisure spaces, and may also contain small shops and services that do not operate at night, while mixed zones in addition to the uses for sensitive zones may contain shops, services, and industries working all day.

In the master plan of Mirandela the residential areas near the industrial park are classified as mixed zones. For these areas, the RGR specify the limit value of 55 dBA for the night noise indicator (Ln) and a 65 dBA for the day-evening-night noise indicator (Lden). Based on this, for research purpose, the monitoring point was evaluated as a mixed zone.

3 Results and Discussion

3.1 Daily Profile of Ozone Levels and its Precursors

Figure 2 shows the variation of solar radiation, ozone and its precursors throughout the day.

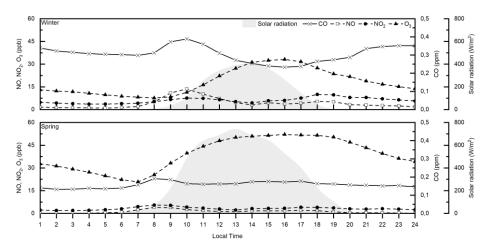


Fig. 2. Average hourly variation of NO₂, NO, CO, O₃ and solar radiation during winter (top) and spring (bottom).

The higher concentrations of the primary pollutants (CO and NOx) registered between 8:00 and 10:00 and between 17:00 and 18:00, coincides with the greatest intensity of traffic associated with the beginning and end of the daily activities inside and outside the industrial park. The higher levels of these primary pollutants in winter show clearly the influence of other combustion sources, such as domestic combustion for heating water and ambient air in dwellings.

In winter, ozone concentrations increase from 8:00 until they reach a peak of 33 ppb at 16:00. It is also noted that during rush hour (about 18:00), the increase in NO and NO₂ concentrations did not imply a significant increase in ozone concentration, partially explained by the prevailing low solar radiation. The ozone pattern in spring and winter was similar, however as the days are longer and the incidence of solar radiation is higher, there are some differences. At spring, ozone concentrations rise from 7:00 to reach its peak at 14:00, remaining stable until 19:00. Considering also that O_3 production depends on the concentration of NO, NO₂, CO, and volatile organic compounds, these gases are consumed as the O_3 concentration increases.

The reactions between O_3 , NO, and NO₂ (R1 - R3) are part of a well-established null-cycle [18]. Thus, during daylight hours (R1 - R2), NO, NO₂ and O₃ are typically balanced on a condition referred to photostationary state [19]. For this reason, it is possible to observe in winter an ozone concentration near 30 ppb between 14:00 and 17:00 and in spring a concentration near 50 ppb between 13:00 and 19:00.

$$NO + O_3 \rightarrow NO_2 + O_2 \tag{R1}$$

$$NO_2 + hv \rightarrow NO + O$$
 (R2)

$$O + O_2 + M \rightarrow O_3 + M \tag{R3}$$

An atmosphere only with nitrogen oxides do not favor the ozone production, as ozone can be recycled during the reactions. However, in the presence of other precursor gases such as carbon monoxide (R4 - R6) and volatile organic compounds (R7-R11), new reactions are triggered, resulting in excess of ozone production [20, 21]. These reactions lead to the formation of the hydroxyl (OH), hydroperoxyl (HO_2) and organo-peroxyl (RO_2) radicals, which results in the formation of nitrogen dioxide without ozone consumption and, consequently, leading to higher ozone production rates by reactions 2 and 3.

$$CO + OH \rightarrow H + CO_2$$
 (R4)

$$H + O_2 + M \rightarrow HO_2 + M \tag{R5}$$

$$HO_2 + NO \rightarrow OH + NO_2$$
 (R6)

$$CH_4 + OH \rightarrow CH_3 + H_2O$$
 (R7)

$$CH_4 + O_2 + M \rightarrow CH_3O_2 + M \tag{R8}$$

$$CH_3O_2 + NO \rightarrow CH_3O + NO_2 \tag{R9}$$

$$CH_{3}O + O_{2} \rightarrow HCHO + HO_{2}$$
 (R10)

$$HO_2 + NO \rightarrow OH + NO_2$$
 (R11)

3.2 Relation Between Air Quality and Meteorological Variables

Tables 1 and 2 show the daytime and nighttime correlation coefficients between hourly means of individual pollutants (CO, NO, NO₂, NO_x and O₃) and the meteorological parameters for winter and spring. The values in bold were those that show a significant correlation at the 0.05 level (2-tailed). Similar to Agudelo–Castaneda et al. [22], a positive correlation was found between nitrogen oxides and carbon monoxide during daylight, indicating that both gases have the same source, mainly related to combustion processes (e.g motor vehicles and industries).

Ozone showed a significant positive correlation with solar radiation and temperature during the daylight hours. This relation reverses during the night, due to the lack of solar radiation for ozone synthesis. This trend is observed in other studies [23, 24], showing that a favorable temperature and high solar radiation increase the photochemical reactions that generate ozone [25]. Moreover, based on reactions 1 to 3, an inverse relationship between ozone and nitrogen oxides is also observed, which can be confirmed by the negative correlation between these gases.

The results also showed a negative correlation between CO and O_3 , since carbon monoxide reacts with hydroxyl radicals (R4) producing HO₂ radicals (R5) and, consequently, NO₂ (R6) which is photo-dissociated to produce O₃ through the reactions 2 and 3.

				Winter I	Daytime			
	CO	NO	NO_2	<i>O</i> ₃	Т.	<i>R.H.</i>	W.S.	S.R.
CO	1	0,780	0,693	-0,688	-0,685	0,629	-0,559	-0,246
NO		1	0,800	-0,622	-0,543	0,502	-0,410	-0,154
NO_2			1	-0,356	-0,281	0,238	-0,380	-0,251
O 3				1	0,852	-0,901	0,715	0,518
Т.					1	-0,895	0,586	0,485
<i>R.H</i> .						1	-0,554	-0,543
<i>W.S.</i>							1	0,474
S.R.								1
				Winter N	ighttime			
	CO	NO	NO_2	O 3	Т.	<i>R.H</i> .	W.S.	S.R.
CO	1	0,771	0,840	-0,791	-0,568	0,608	-0,358	0,160
NO		1	0,802	-0,672	-0,391	0,443	-0,346	0,134
NO_2			1	-0,518	-0,283	0,304	-0,300	0,076
<i>O</i> ₃				1	0,739	-0,860	0,458	-0,295
Т.					1	-0,702	0,328	-0,177
<i>R.H</i> .						1	-0,288	0,392
<i>W.S.</i>							1	0,088
S.R.								1

 Table 1. Spearman correlation coefficients between hourly mean CO, NO, NO2, NOx, O3 and meteorological parameters for winter nighttime and daytime

T. - Temperature R.H. - Relative Humidity W.V. - Wind Speed S.R. - Solar Radiation

It is also observed in Tables 1 and 2 that the wind speed has a positive correlation with ozone concentration and a negative correlation with nitrogen oxides and carbon monoxide. Agudelo–Castaneda et al. [22] explain that this phenomenon may occur due to high wind speeds, which leads to the dispersion and mixing the gases from local sources, favoring ozone transport and formation reactions taking place in the atmosphere. Moreover, Markovic and Markovic [26] also justify the positive correlation between wind speed and ozone levels due to the transport of ozone produced on the main roads to the measurement point. There is also an expected negative correlation between ozone and relative humidity, since as relative humidity increases the major photochemical paths of O₃ removal will be lowered [27]. High humidity levels are associated with cloudy days and less sunshine, thus reducing photochemical processes [27].

 Table 2. Spearman correlation coefficients between hourly mean CO, NO, NO2, NOx, O3 and meteorological parameters for spring nighttime and daytime

Spring Daytime									
	СО	NO	NO ₂	<i>O</i> ₃	Т.	<i>R.H</i> .	<i>W.V.</i>	S.R.	
CO	1	0,675	0,781	-0,091	-0,375	0,330	-0,031	-0,223	
NO		1	0,906	-0,402	-0,340	0,369	0,001	-0,211	
NO_2			1	-0,211	-0,325	0,296	-0,096	-0,327	
O 3				1	0,531	-0,706	0,313	0,328	
Т.					1	-0,825	0,159	0,498	
R.H.						1	-0,175	-0,528	
W.V.							1	0,160	
S.R.								1	
				Spring N	ighttime				
	CO	NO	NO2	03	Т.	<i>R.H.</i>	<i>W.V.</i>	S.R.	
СО	1	0,142	0,458	-0,032	-0,302	0,164	-0,069	-0,009	
NO		1	0,684	-0,369	0,071	0,178	-0,193	0,403	
NO_2			1	-0,392	-0,033	0,113	-0,377	0,200	
O 3				1	0,450	-0,715	0,702	-0,059	
Т.					1	-0,681	0,345	-0,013	
R.H.						1	-0,482	0,057	
W.V.							1	0,058	
S.R.								1	

T. - Temperature R.H. - Relative Humidity W.V. - Wind Speed S.R. - Solar Radiation

3.3 Local and Regional Contributions to Ozone Formation

To determine the local and regional contribution to ozone formation, potential ozone levels $(O_3 + NO_2)$, also called total oxidant levels (OX), were related to NOx, following the same analysis used in other studies [19, 28, 29]. For this purpose, daily average values (day and night) of OX were evaluated against the values of NO_X. For each data distribution, a linear regression was applied, thus providing an equation, in which the slope represents the local contribution (NO_x-dependent), while the intersection represents the regional contribution (NO_x-independent). Figure 3 presents the linear regressions lines obtained for each of the studied months. The regional contribution represents the background OX concentration, while the local contribution is related to the local production/destruction [28].

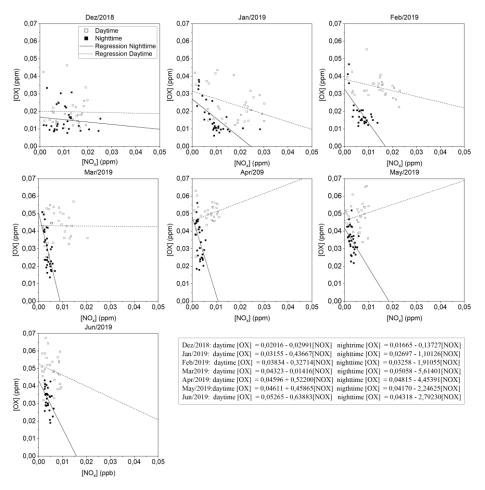


Fig. 3. Daily variation means of regional [OX] with NOx during the months of winter and spring for daytime and nighttime.

When OX levels increase as a function of NO_X , the NO_X contributes mainly to ozone production; when the OX levels decrease, means that NO_X influences the processes of ozone depletion; and when OX levels remain relatively constant as a function of NO_X , indicate that NO_X contributes in equal parts to the production and depletion of ozone.

Based on the regression analysis from December 2018 to June 2019, it was possible to obtain the slope and intersection for each period. These results are presented in Figure 4 showing the monthly local and regional dependence of OX. It is noticeable that values of the local contribution are higher than the values of the regional contribution, thus indicating that for the industrial park of Mirandela OX production occurs mainly locally due to primary pollutants emissions.

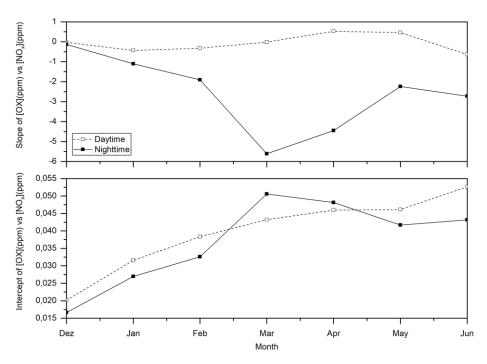


Fig. 4. Monthly variation of regional (intercept) and local (slope) OX in the Industrial Park of Mirandela.

For the regional component, there is a tendency to an increase in the concentration of OX, because the intensification of solar radiation helps photochemical processes. This same tendency was observed in the study of Notario et al. [29]. Regarding the local component, for daytime, an increase in OX concentration is observed from January to May, indicating that during this period NO_X favors ozone production. For the nighttime the relationship is inverse, suggesting that that NO_X is mainly related to ozone depletion mechanisms.

3.4 Noise monitoring

Figure 5 shows the hourly averages values during the spring months. Between Monday and Friday, the noise profile throughout the day is very similar, with the lowest averages observed between 1:00 and 3:00 at night. Subsequently, noise rises until 8:00, when activities start in the industrial zone, remaining relatively stable until 12:00. At his time, there is a decrease in noise levels due to lunchtime, where most activities stop in the industrial zone. After 18h00 the noise level decreases due to the finish of the work hours. During the weekend, noise levels are lower than during working days, although Saturday levels are higher than those registered on Sundays because some activities operate on Saturday.

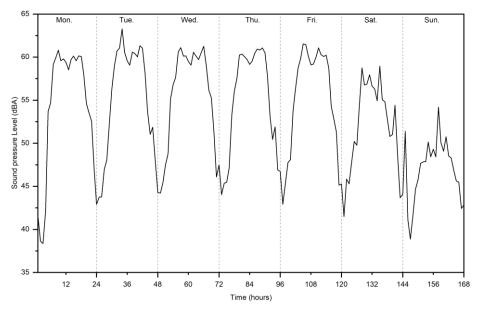


Fig. 5. Average hourly variation of sound pressure levels true for spring weeks.

Figure 6 presents the daily average night noise levels (Ln) and the day-evening-nightnoise (Lden) levels during the spring days. The Portuguese Decree-Law No. 9/2007 of 17 January establishes for Lden a limit of 65 dBA and for Ln a limit of 55 dBA for mixed zones. During the evaluation time the night indicator was exceeded on April 4th, 15th, and 24th, and May 8th and 24th, reaching the values of 57.0, 56.7, 56.4. 56.0 and 55.6 dBA respectively. As the values of Ln are very close to the established limit and observing that only occurred five days in three months, it can be inferred that a singular event resulted in this phenomenon, so they are values not very relevant for the acoustic characterization of the industrial zone.

It should also be noted that although there are no legal limits for industrial parks, the observed values are within the legal limits for mixed zones, meaning that noise generated by activities developed in industrial park of Mirandela, including traffic road have little impact on the acoustic environment of the sensitive receivers located in its vicinity.

During working days, noise remains relatively stable, close to 60 dBA, thus indicating that the noise sources in the industrial park have a typical behavior over the week.

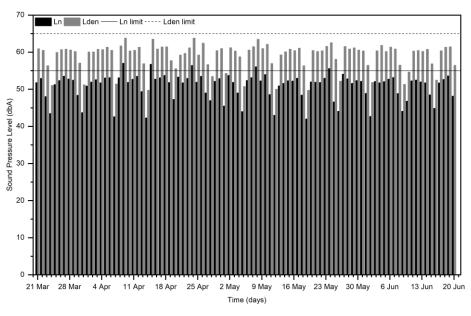


Fig. 6. Ln and Lden levels for Spring

It is noteworthy that only one fixed noise sensor was used in this study, and, for a better acoustic characterization of the industrial zone, it would be necessary to install more noise sensors, considering that the sound is attenuated due to the obstacles and the distance traveled. Additional monitoring points would also add useful information on the diversity of noise sources.

4 Conclusions

The monitoring of atmospheric pollution in the industrial park of Mirandela was useful for the identification of the main components related to the local ozone production, and it was possible to verify that besides the ozone precursors, the meteorological variables have a great influence on its production, especially the solar radiation.

CO and NO emissions were also related mainly to urban traffic because their concentration increased at the same time during the rush hours in the industrial zone. Regarding the analysis of OX vs NO_x, it was found that the ozone present in the region derives mainly from local influence, and the negative relationship found for the local component indicates that NO_x favors ozone depletion mechanisms. Thus, in future studies, it is suggested to monitor VOCs and hydrocarbons to know the different ozone production routes and verify which pollutant has the highest impact on ozone synthesis. Also, by monitoring noise, it was possible to create the daily and weekly sound pressure level profiles and, verify that during the observation period in five occasions the night limit was exceeded. Continuous monitoring will continue in the industrial park of Mirandela, in order to obtain the ozone and noise profile for all seasons of the year.

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