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AIR POLLUTION IN THE NORTH OF PORTUGAL: THE IMPLICATION OF 2020 LOCKDOWN

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ABSTRACT

Infectious disease COVID-19 caused by a new coronavirus SARS-CoV-2 was identified in Wuhan, China, in 2019 and became a public health emergency of international concern. To control its propagation, governments around the world have implemented various restrictive measures that have led to reduction of people movement, decrease in transport (road and aviation), and even suspended industrial activities. Lockdowns due to COVID-19 have had both social and economic consequences but also impacted the environment and air quality.

This study aimed to analyze the impacts of restrictions implemented in Portugal (March- May) to control the spread of COVID-19 on air pollution in the North of Portugal focusing three main pollutants, namely nitrogen dioxide (NO_2), particulate matter (PM_{10}) and ozone (O_3).

The hourly concentrations of NO_2, PM_{10} and O_3 were retrieved for 22 stations (urban, suburban, and rural) that belong to the monitoring network of northern region. Three different periods of time were included: pre-lockdown (January 1–March 14), lockdown (March 15–June 13) and post-lockdown (June 14–September 30).

The results showed that restrictions implemented to mitigate effects of the COVID-19 pandemic in Portugal affected all the studied pollutants. In all three periods analyzed levels of NO_2 were higher at stations with strong influence of traffic emissions. However, more significant decreases during the lockdown occurred at sites with indirect influence of traffic emissions: 50% at rural stations, 46% at urban background stations and 38% at urban traffic stations. For PM_{10}, industrial and traffic stations showed the most significant decrease during lockdown (43% and 37%), followed by background and suburban areas where concentrations decreased at a lesser degree (33% and 30%). Finally, the O_3 concentration trends differed from NO_2 and PM_{10}. Rural zones exhibited the highest levels of O_3. Rural and suburban sites also exhibited very similar evolution trends with no significant changes during and after lockdown. Concentrations at urban background stations, although lower than at rural zones, showed more temporal trends during (24% increase) and after lockdown (26% decrease).

Though lockdown during the COVID-19 pandemic led to some direct positive impacts on air quality in the studied area of Portugal, these seem to be temporary. Furthermore, as air pollution might aggravate the consequences of COVID-19, it is now of outmost importance to continue with strategies to reduce emissions of harmful pollutants in the atmosphere and to maintain good air quality for the respective citizens.

RESUMO

A COVID-19 é uma doença infeciosa causada por um novo coronavírus denominado SARS-CoV-2. Foi inicialmente identificada em Wuhan, China em 2019 e rapidamente tornou-se uma emergência de saúde pública a nível internacional.

Para controlar a sua propagação, governos de diversos países implementaram várias medidas restritivas que levaram à redução da circulação, diminuição do transporte (rodoviário e aéreo) e até à suspensão de atividades industriais. O isolamento devido ao COVID-19 além de ter consequências sociais e económicas, causou impacto também no ambiente e na qualidade do ar. Este trabalho teve como objetivo analisar os impactos das restrições implementadas para controlar a propagação da COVID-19 em Portugal (Março-Maio), na poluição atmosférica no Norte do país. A análise é feita tendo em foco três principais poluentes, nomeadamente dióxido de azoto (NO_2), matéria particulada (PM_{10}) e ozono (O_3). Foram consideradas as concentrações horárias de NO_2, PM_{10} e O_3 de 22 estações (urbanas, suburbanas e rurais) pertencentes à rede de monitorização da qualidade do ar da região norte. Foram definidos três diferentes períodos para análise: pré-isolamento (1 de Janeiro a 14 de Março), isolamento (15 de Março a 13 de Junho) e pós-isolamento (14 de Junho a 30 de Setembro).

Os resultados mostraram que as restrições implementadas afetaram todos os poluentes analisados. Nos três períodos analisados os níveis de NO_2 foram maiores nas estações com forte influência do tráfego rodoviário. No entanto, durante o isolamento, as diminuições mais significativas ocorreram em estações com influência indireta do tráfego: 50% nas estações rurais, 46% nas estações urbanas de fundo e 38% nas estações urbanas de tráfego. Para a PM_{10}, as estações industriais e de tráfego apresentaram diminuições mais significativas durante o isolamento (43% e 37%), seguidas por estações urbanas de fundo e suburbanas onde as concentrações diminuíram em menor grau (33% e 30%). Finalmente, para o O_3 as tendências de concentração foram diferentes do NO_2 e PM_{10}. As zonas rurais exibiram os níveis mais elevados de O_3 e apresentaram tendências de evolução muito semelhantes às estações suburbanas, sem mudanças significativas durante e após o isolamento. As concentrações nas estações urbanas de fundo, embora mais baixas do que nas zonas rurais, mostraram variações mais significativas durante (aumento de 24%) e após o isolamento (diminuição de 26%).

Embora o isolamento devido à pandemia da COVID-19 tenha levado a alguns impactos positivos diretos na qualidade do ar na área estudada, estes parecem ser temporários. Além disso, tendo em conta que a poluição do ar pode agravar as consequências da COVID-19, é de extrema importância continuar com estratégias para reduzir as emissões de poluentes nocivos na atmosfera e manter a boa qualidade do ar para os cidadãos.

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List of Publications

Some of the content presented in this thesis was submitted and accepted for publication and oral communication in the following events:

1. **14th University of Porto Young Researchers Meeting (IJUP), Porto, May 2021 (Book of Abstracts: page 159)**
   Leidy Tavares, Maria do Carmo Pereira, Klara Slezakova. *Impacts of the COVID-19 lockdown on air pollution in the North of Portugal in 2020.*

2. **4th Doctoral Congress in Engineering (DCE21), Symposium on Environmental Engineering, Porto, June 2021 (Book of Abstracts: page 49)**
   Leidy Tavares, Maria do Carmo Pereira, Klara Slezakova. *Traffic related pollution due to COVID-19 lockdown in the North of Portugal in 2020.*

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NOMENCLATURE

ABBREVIATIONS

APA Agência Portuguesa do Ambiente

AQG Air quality guidelines

BC Black carbon

CCDR Comissão de Coordenação e Desenvolvimento Regional

CCDR-N Comissão de Coordenação e Desenvolvimento Regional do Norte

COVID-19 Disease caused by the coronavirus SARS-COV-2

DGS Direção Geral da Saúde

EEA European Environment Agency

EPA Environmental Protection Agency

EU European Union

GHG Greenhouse gas

IRCEL Belgian Interregional Environment Agency

NUTS Nomenclature of Territorial Units for Statistical Purposes

OMA Oporto metropolitan area

PAH Polycyclic aromatic hydrocarbons

PM Particulate matter

PM_{10} Particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 10 mm aerodynamic diameter

$PM_{2.5}$ Particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 2.5 mm aerodynamic diameter

QualAr Air quality information system

VOC Volatile organic compounds

WHO World Health Organization

ELEMENTS AND COMPOUNDS

CH_4 Methane

C_6H_6 Benzene

CO Carbon monoxide

CO_2 Carbon dioxide

NH_3 Ammonia

NO Nitric oxide

NO_X Nitrogen oxides

NO_2 Nitrogen dioxide

O_3	Ozone
SO_2	Sulphur dioxide

1 THESIS OUTLINE

This thesis is divided into 5 chapters and the content associated to each chapter is detailed bellow.

Chapter 1 is the present chapter and explains how the thesis is structured.

Chapter 2 is the introduction and provides an overview of the problematic of air pollution and its impacts focusing on the pollutants considered relevant to this work. A brief description of the legislation applicable to ambient air pollution is presented, as well as a summarized timeline of COVID-19 pandemic in order to help understand the events that led to the scenario we are currently experiencing. The chapter also presents the main objectives of the work.

Chapter 3 presents the materials and methods used to carry out the present dissertation including aspects such as the description of the study region, characterization of the data analyzed, and the period defined to perform the analysis.

Chapter 4 contains the results and the discussion of the work. It consists of the analysis of the da5 summarizes the main conclusions of this work and presents suggestions for future studies

2 Introduction

2.1 Air pollution

The World Health Organization (WHO) defines ambient air pollution as a complex mixture of different substances that occur in the gas, liquid or solid phase. Several potentially harmful components have been found in the troposphere and to characterize air quality, the 2005 "WHO Air quality guidelines" offer global guidance on thresholds and limits for key air pollutants that pose health risks. These guidelines apply worldwide and are based on expert evaluation focusing on four indicators of air pollution: particulate matter (PM), ozone (O_3), nitrogen dioxide (NO_2) and sulfur dioxide (SO_2) (WHO, 2005). Indoor and outdoor air pollution, even at low doses, are considered a threat to human health and have been associated with increased mortality and morbidity worldwide (WHO, 2015).

There are many different types of air pollutants. These include various gases, divided solids, or finely dispersed liquid aerosols that may reach rates that exceed the natural capacity of the environment to dissipate and dilute or absorb them (Nathanson, 2020).

With emissions and air concentrations increasing worldwide, air pollution is a global threat with large impacts on both human health and ecosystems (EEA, 2019). Recent European Environment Agency (EEA) reports inform that although air quality has slowly improved in several European cities due to robust air quality policies, introduction of targeted measures and technological improvements, some regions still experience exceedances of the regulated limits of air pollution (EEA, 2018). Especially in urban areas air pollution continues to have significant impacts on the health of the European population with PM_{10}, NO_2 and ground-level O_3 considered the most serious pollutants in terms of human health in Europe (EEA, 2020).

Air pollution is currently the most important environmental risk to human health, and it is considered the second greatest environmental concern for Europeans, after climate change. As a result, there is a strong political, media and public interest in all air quality issues and, consequently, increased public support for action (EEA, 2019).

EEA reports that in 2017, around 10% of all the reporting stations in European Union (EU) countries recorded concentrations above the annual limit for NO_2, which is equivalent to the WHO air quality guidelines (AQGs) limit value. In total, 86% of concentrations above this limit value were observed at traffic stations. In the same year concentrations of PM_{10} exceeded both the EU limit values and the WHO AQGs. PM_{10} concentrations above the EU daily limit value were registered at 22% of the reporting stations while long-term WHO AQGs for PM_{10} was exceeded at 51% of the stations. A total of 17% of the EU urban population was exposed to PM_{10} levels above the daily limit value and 44% was exposed to concentrations exceeding the stricter WHO AQGs value for PM_{10} in 2017. For O_3, 20% of stations registered concentrations above the EU target value for the protection of human health while the WHO AQGs value for O_3 was exceeded in 95 % of all the reporting stations. This represents about 14% of the EU urban population being exposed to O_3 concentrations above the EU target value threshold and 96% population exposed to O_3 levels exceeding the WHO AQGs value (EEA, 2019).

To effectively reduce air pollution and its impacts, it is necessary to have a good understanding of its causes, how pollutants are transported and transformed in the atmosphere, how the chemical composition of the atmosphere changes over time and how pollutants affect humans, ecosystems, the climate and subsequently society and the economy.

2.2 IMPACTS OF AIR POLLUTION

The health impacts of exposure to air pollution are unquestionable. Numerous epidemiological studies have found an association between air pollution and a wide range of adverse health effects, from subtle subclinical effects to premature death

(WHO, 2016). According to the WHO seven million people annually prematurely dies worldwide due to poor air quality. WHO data shows that 9 out of 10 people breathe air that exceeds WHO limits, with low and middle-income countries suffering from the highest exposures (WHO, 2014).

Exposure to air pollution and its effects on human health have motivated several studies over the past years. Increases in mortality and hospital admissions due to respiratory and cardiovascular diseases have been associated with exposure to PM and O_3 even at very low levels, and it is still unclear if there is a threshold concentration below which there is no concerning effects on human health (Brunekreef & Holgate, 2002).

Air pollution is a risk factor for many mortalities and adverse health effects including heart disease, stroke, lower respiratory infections, lung cancer, diabetes and chronic obstructive pulmonary disease. There is also emerging evidence that exposure to air pollution is associated with type 2 diabetes in adults and it may be linked to obesity, systemic inflammation, Alzheimer's disease, and dementia (EEA, 2020). The effects of air pollution on health does not depend only on type and extents of exposure but also on the susceptibility of individuals. Air pollution affects the whole population, but certain groups, such as children, elderly people, pregnant women, and those with pre-existing health problems have more susceptibility to its adverse effects (Ritchie & Roser, 2017).

Hooper and Kaufman describe the concept of susceptibility as the characteristics (external and intrinsic) that increase the risk of experiencing adverse health outcomes in response to air pollution exposure. Examples are the presence of underlying disease, age, and genetic background but also sex, race, ethnicity, epigenetic changes, socioeconomic position, nutritional status, and personal behaviors such as physical activities or smoking (Hooper & Kaufman, 2017).

Air pollution has also several important environmental impacts. It directly affects natural ecosystems and biodiversity. The most harmful air pollutants in terms of damage to ecosystems are ozone, ammonia and nitrogen oxides. Nitrogen oxides and ammonia emissions disrupt terrestrial and aquatic ecosystems by introducing excessive amounts of nitrogen nutrient, which causes eutrophication (i.e. an oversupply of nutrients that can lead to changes in species diversity and to invasions of new species). Air pollution can also contribute to the acidification of soil, lakes, and rivers, causing loss of biodiversity. Ground-level ozone damages agricultural crops, forests and plants by reducing their growth rates and has negative impacts on biodiversity and ecosystem services (EEA, 2020).

Air pollution is highly linked with climate change. Pollutants, such as carbon dioxide, ozone, nitrogen oxides, non-methane volatile organic compounds, ammonia and sulfur dioxide are also climate forcers, which have a potential impact on climate and global warming in the short term (Fu et al., 2020).

Changes in weather patterns due to climate change may then affect the transport, dispersion, deposition, and formation of air pollutants in the atmosphere while higher temperatures may lead to increased ozone formation. As greenhouse gases and air pollutants share the same emission sources, benefits can arise from limiting emissions of one or the other (EEA, 2019).

Air pollution also has considerable economic impacts, due to the shortening of life expectancy, increasing medical costs and reduce productivity through working days lost across the economy (EEA, 2019).

Although the association of air pollution with multiple adverse health outcomes is very well established, its negative economic impact is less well measured. A study in India estimated the economic impact of air pollution as the cost of lost output due to premature deaths and morbidity in consequence of exposure to air pollution. According to this study death and disease due to air pollution and its adverse economic impact from loss of output could impede India's aspiration to be a $5 trillion economy by 2024. Thus, implementing successful specific strategies to reduce air pollution would lead to substantial benefits for both the health of the population and the economy (Pandey et al., 2021).

2.3 Sources and pollutants

In general, air pollutants can be classified as primary or secondary (EEA, 2019). Primary emissions occur when pollutants are released directly into the atmosphere from a source such as a factory chimney or exhaust pipe, or through suspension of contaminated dusts by the wind. So, in theory, it is possible to measure the amounts emitted at the source itself (WHO, 2005).

However, pollutants can also be formed as a result of chemical interactions involving precursor substances (i.e. secondary formation). There are complex links between pollutants emissions to air and the respective air quality, including emission heights, chemical transformations, reactions to sunlight, additional natural and hemispheric contributions and the impact of weather and topography (EEA, 2020b). Therefore, reductions of emissions do not necessarily result in similar decrease in ambient air concentrations, though significant decrease of emissions are essential for improving air quality (EEA, 2019).

Air pollutants may have a natural, anthropogenic, or mixed origin, depending on their sources or the sources of their precursors. Burning of fossil fuels to generate electricity, industrial processes, transport, household heating, solvent use in chemical and mineral industries, agriculture and waste treatment represent the main anthropogenic sources. Volcanic eruptions, windblown dust, sea-salt spray, and emissions of volatile organic compounds from plants are examples of natural emission sources (EEA, 2020b).

Key primary air pollutants include particulate matter (PM), black carbon (BC), sulfur dioxide (SO_2), nitrogen oxides (NO_X) (including nitrogen monoxide–NO, and nitrogen dioxide–NO_2), ammonia (NH_3), carbon monoxide (CO), methane (CH_4), volatile organic compounds (VOCs), including benzene (C_6H_6), certain metals and polycyclic aromatic hydrocarbons (PAHs). The secondary air pollutants are represented by PM (formed in the atmosphere), ozone (O_3), NO_2 and several oxidized VOCs. Depending on the pollutant, the main sectors responsible for air pollution in Europe are transport (road and non-road), residential, commercial, institutional, energy supply (fuel production and processing and energy production) manufacturing and extractive industry, agriculture and wastewater management (EEA, 2020).

2.3.1 Nitrogen oxides (NO_x)

The combustion of fossil fuels is one of the activities responsible for the emission of large quantities of pollutants to the environment. Among these pollutants are the nitrogen oxides (NO_x) which are the source of severe environmental problems such as acid rain, smog formation, global warming, and ozone layer weakening (Gómez-Garcia et al., 2005).

Nitrogen oxides are a group of nitric oxide gas (NO), nitrogen dioxide (NO_2) and other gaseous oxides containing nitrogen. NO_x is a strong oxidizing agent that is produced by combustion processes. Nitrogen oxides are emitted as NO that oxidizes to NO_2 or reacts with ozone or radicals in the atmosphere forming NO_2 (Kampa & Castanas, 2008).

In urban areas, traffic exhausts (i.e. personal automobiles, trucks, and various non-road vehicles–construction equipment, boats, etc.) are the main sources of NO_x. EEA informs that more than 40% of EU NO_x emissions originate from road transport (EEA, 2020b).

NO_x has been associated with adverse health effects while exposure to high levels of nitrogen dioxide can interfere with the ability of blood to carry oxygen, leading to dizziness and shortness of breath (EPA, 2021). According to WHO there are epidemiological studies providing evidence that long-term NO_2 exposure may decrease lung function and increase the risk of respiratory symptoms (WHO, 2003).

2.3.2 Particulate matter (PM)

Particulate matter is a mixture of solid particles and/or liquid droplets found in the air. The size of these particles is a significant parameter as it determines the place of the PM deposition in the respiratory system. Upon inhalation, PM_{10}, inhalable particles – those with aerodynamic diameters below 10 μm, typically deposit in upper respiratory airways; $PM_{2.5}$, fine PM – i.e. particles with aerodynamic below 2.5 μm, can enter the lungs. The smallest fraction of PM, the ultrafine particles, then penetrates the bloodstream, thus causing serious health problems to almost all human organs (Harrison & Yin, 2000; Kampa & Castanas, 2008).

While natural sources of PM include sea salts and resuspended dust, or emissions from volcanic eruptions, the majority of PM is originated from man-made activities. The main primary anthropogenic sources of PM are fossil fuel combustions and industrial process (such as power plants, waste incinerations, construction activities, etc.). The secondary PM are formed in the atmosphere as a result of complex reactions of key precursors, namely SO_2, NO_2, NH_3, and VOCs (WHO, 2013).

There are several studies connecting particle exposure to the source of various health problems including premature death in people with heart or lung disease, nonfatal heart attacks, irregular heartbeat, aggravated asthma, decreased lung function, and increased respiratory symptoms such as irritation of the airways, coughing, or difficulty breathing (Ki-Hyun et al., 2015; Cadelis et al., 2014).

The composition of PM is variable, as they can absorb and transfer a variety of pollutants such as metals, organic compounds, material of biologic origin, ions, reactive gases, and the particle carbon core. There is evidence supporting that ultra-fine and fine particles can be more dangerous than larger ones in terms of mortality and cardiovascular and respiratory effects. In addition, the metal content, the presence of PAHs and other organic components can increase PM toxicity (Kampa & Castanas, 2008).

2.3.3 Ozone (O_3)

Tropospheric, or ground level ozone, is a colorless and highly irritating gas. It is a secondary pollutant which is formed by the reactions of its precursors, mainly NO_X, VOCs, CO and CH_4 in the presence of strong solar radiation (EEA, 2021; Duan et al., 2008). Ozone has a strong oxidizing power that accelerates the degradation of materials, promotes the loss of vegetation productivity and as one of the greenhouse gases, it contributes to the global warming of the troposphere (APA, 2019).

Studies investigating short-term exposures to O_3 showed positive associations with morbidity, mortality, exacerbation of respiratory illness, and increased hospital admissions. There is also strong evidence that short-term O_3 exposure is associated with cardiovascular disease and premature death. However, epidemiological studies of long-term O_3 exposure are scarce (Turner et al., 2015).

The rate of ozone production is a nonlinear function of the mixture of VOC and NO_x in the atmosphere and much of the difficulty in reducing ozone can be traced to its complex chemistry (Cardelino & Chameides, 1995).

In rural areas NO_x emissions are mainly responsible for ozone formation, while VOCs are responsible in urban areas. In less polluted environments, ozone is produced through the photolysis of NO_2 in the presence of solar radiation. In the absence of other chemical species this cycle reaches a photo-stationary equilibrium without effective ozone formation. In polluted environments the photochemical production of ozone is influenced by the additional presence of VOCs. The VOC oxidation cycle disturbs the natural equilibrium NO_x–O_3, allowing alternative paths for the oxidation of NO to NO_2 without consumption of O_3 (Alvim-Ferraz et al., 2006).

As several studies have reported growing levels of background ozone, to obtain its acceptable levels, abatement of a major fraction of both NO_x and VOCs emissions is necessary (IRCEL, 2021).

According to WHO, tropospheric ozone is one of the most widespread global air pollution problems today. Ozone and some of its precursors are also transported long distances in the atmosphere and are therefore considered a transboundary problem. In urban areas, near strong emission sources of nitrogen oxides, where there is an abundance of nitric oxide, ozone levels are lower as it reacts with nitric oxide. As a result, the ozone concentrations are often lower in busy urban areas and higher in suburban and adjacent rural areas. (WHO, 2005).

2.4 LEGISLATION

The EU policies aim to reduce exposure to air pollution by reducing the emissions and setting limits and target values for air pollutants in ambient air. EEA is the air pollution data center for European Union. It supports the implementation of EU legislation linked to air emissions and air quality.

The EEA also contributes to the evaluation of EU air pollution policies and to the development of long-term strategies to improve air quality in Europe. The EU acts at many levels to reduce exposure to air pollution: through legislation, through co-operation with sectors responsible for air pollution, through national, regional authorities and non-government organizations and through research.

EU Directive on Ambient Air Quality and Cleaner Air for Europe addresses air pollution. It describes the basic principles concerning the assessment and management of air quality and sets pollutant concentrations thresholds that shall not be exceeded (Directive 2008/50/CE).

Through this directive, the EU has developed an extensive body of legislation which establishes health-based standards and objectives for a number of pollutants present in the air. Those standards are applied over different time duration because the observed health impacts associated with the various pollutants occur over different exposure times. These standards and objectives are summarized in Table 1, focusing on PM_{10}, NO_2 and O_3, which are the pollutants considered in this work.

Table 1: Air quality standards defined by European Union (Directive 2008/50/CE)

Pollutant	Averaging period	Legal nature and concentration	Comments
PM_{10}	24 h	Limit value: 50 µg/m^3	Not to be exceeded more than 35 times per year
	1 year	Limit value: 40 µg/m^3	
NO_2	1 h	Limit value: 200 µg/m^3	Not to be exceeded more than 18 times per year
	1 year	Limit value: 40 µg/m^3	
O_3	Maximum daily 8-hour mean	Target value: 120 µg/m^3	Not to be exceeded more than 25 days averaged over 3 years

2.5 COVID-19 PANDEMIC

In December 2019, authorities in Wuhan, China, alerted the WHO that dozens of people were being treated for pneumonia from an unknown source. What started as a mystery disease was first referred to as 2019-nCoV and then named COVID-19 (Ravelo & Jerving, 2021). Coronavirus disease (COVID-19) is an infectious disease caused by a newly discovered coronavirus that spreads in the air mainly through droplets of saliva or discharge from the nose when an infected person coughs or sneezes (WHO, 2021).

In January 2020, Chinese state media reported the first death from novel coronavirus. China imposed aggressive containment measures in Wuhan, the epicenter of the outbreak, suspending flights and trains and shutting down subways, buses and ferries in an attempt to stem the spread of the virus. Later that month, for the sixth time in history, the WHO declared a public health emergency of international concern, a designation reserved a highest level of alarm for extraordinary events and WHO's highest level of alarm (Schumaker, 2021). On March 2020, with the disease spreading rapidly around the world, WHO made the assessment that the COVID-19 outbreak could be characterized as a pandemic (Cucinotta & Vanelli, 2020).

Europe faced its first major outbreak in February 2020 as the number of reported cases in Italy started to increase dramatically. In the Lombardy region, officials locked down several towns after a cluster of cases suddenly emerged in the southeast of Milan, with schools closing and sporting and cultural events being canceled (The New York Times, 2021). As of May 16th 2021, WHO has reported over 162 million cases and 3.3 million deaths all over the world with the virus spreading to nearly every country and affecting society in multiple ways (WHO, 2021b).

To limit the spread of pandemic, governments in countries around the world have imposed various restrictions, which included reduction of people movements, decrease in transport (road and aviation), and even suspended industrial activities.

In Portugal, the first two recorded cases were confirmed on March 2020 in Porto (SIC, 2020). On March 12, Prime Minister António Costa announces that all schools should close and six days later the President of the Republic Marcelo Rebelo de Sousa decrees the first state of emergency due to the new coronavirus. On that date, 194 new daily cases were registered, and the country had already accumulated total 642 cases and two deaths. When Portugal declared its State of Emergency, most of European countries had already enforced various protective actions. The general measures included social distancing and teleworking whenever possible; all non-essential commercial establishments were closed; restaurants could only operate with take-away service. Measures were taken for three groups of citizens. Firstly, infected patients or suspected cases became obliged to isolate themselves. People at risk, such as the elderly or the chronically ill, should not leave their residence unless exceptional circumstances. The third group composed of all other citizens, who were subject to a general duty of recollection (Santos & Piteira, 2021; Portuguese Republic, 2020).

The state of emergency was in effect in Portugal until May 2nd, 2020. The country underwent a state of calamity, initiating a three-phase plan (May 4th, May 18th, and June 1st), allowing for a gradual reviving of various sectors of activity (Assembleia da República, 2021). However, at the end of the state of emergency not all restrictions were lifted. Circulation of citizens was limited at certain times, the use of a mask in confined public spaces was mandatory, and gatherings with more than ten people were prohibited (Santos & Piteira, 2021).

To this date (June 2021) there are 869 thousand confirmed cases and 17 thousand deaths (DGS, 2021).

2.6 THESIS OBJECTIVES

Recent studies have proved that although the pandemic has had impacts in economy, transport, health care and other sectors, restrictive measures imposed by countries have also caused lower air pollution emissions and consequent air quality improvement (Skirienè & Stasiškienè, 2021).

Despite the indirect negative impacts of contingency measures, such as the reduction in recycling and the increase in domestic and medical waste, there is also a significant association between contingency measures and improvement in air quality, clean beaches, environmental noise reduction as well as a reduction in emissions of some greenhouse gases (GHG) (Zambrano-Monserrate et al., 2020).

A study comparing data emissions from over 20 monitoring stations in Portugal during the lockdown period with data from the mirrored period of the previous five years, showed 41% reduction in NO_2 emissions and 18 % reduction in PM_{10} emissions (Gama et al., 2021). All the evidence and studies suggesting that restriction measures due to the COVID-19 pandemic can impact air pollution, set a precedent for the interest in analyzing the effects of such measures in the northern region of Portugal.

The main objective of this study was to evaluate the impacts of the first wave COVID-19 lockdown on air quality in the North of Portugal in the year of 2020. The specific objectives of this work include:

- Assessment of air pollution per type of pollutants, namely NO_2, PM_{10} and O_3;
- Analysis per type of monitoring stations (urban, suburban, rural) and emission sources (traffic, background, industrial);
- Data analysis divided in three periods of time (pre-lockdown, lockdown and post-lockdown);
- Comparison between concentrations of NO_2, PM_{10} and O_3 and the existing legislation for each pollutant.

3 MATERIALS AND METHODS

3.1 STUDY REGION

The data used in this study are related to the north region of Portugal that includes 86 municipalities, about 3.6 million inhabitants and concentrates almost 35% of the resident population in Portugal. Its area is approximately 21284 km^2 with a population density of 168 inhabitants per km^2 (CCDR-N, 2021). It is one of five regions of Mainland Portugal (NUTS II subdivisions) and includes the Oporto Metropolitan Area (OMA) that is located on the North Coast of Portugal with a population of around 1.7 million inhabitants (AMP, 2021).

3.2 DATA DESCRIPTION

Data for selected pollutants, namely PM_{10}, NO_2 and O_3 can be retrieved from the QualAr, an online database managed by Portuguese Environmental Agency. Although, it is important to emphasize that when this work was started (October 2020) the data used, which were kindly provided by the CCDR-N (Northern Regional Coordination and Development Commission), had not yet been validated and published on the QualAr database.

The database aims to centralize all air quality data measured in Portugal and provides information to the public (Portuguese Environmental Agency, 2021). Air quality assessment is carried out using networks of monitoring stations that are managed by the Regional Development and Coordination Commissions (CCDR) of the region where they operate. Data that is measured continuously at the different stations is transmitted, in almost real time, to regional concentrators and from these to the central information system based on the QualAr database; they are then made available to the public. The number of the stations in each region correspond to the number of inhabitants and population density, the characteristics of land occupation and air quality levels (QualAr, 2021).

This study considered the northern region monitoring network that includes 22 monitoring stations. Depending on the density and distribution of buildings in the surrounding area, monitoring stations are classified as: suburban – large built-up urban area; urban – continuously built-up urban area; and rural – all other areas. Furthermore, considering the emission sources the stations are characterized as (Table 2): (i) background one with pollution levels representative of the average exposure of the general population or vegetation; (ii) traffic one that are situated in a close proximity to a single major road; and (iii) industrial stations which are situated in close proximity to an industrial area or an industrial source (EEA, 2020).

Table 2: Classification of air quality monitoring stations in North region (EEA, 2020; QualAr, 2021b)

Station	Type	Emission sources
Frossos-Braga	Suburban	Background
Anta-Espinho	Suburban	Background
VNTelha-Maia	Suburban	Background
Custóias-Matosinhos	Suburban	Background
Meco-Perafita	Suburban	Industrial
Leça do Balio-Matosinhos	Suburban	Background
Mindelo-Vila do Conde	Suburban	Background
Fr Bartolomeu Mártires-S.Vitor	Urban	Traffic
Avintes	Urban	Background
Cónego Dr. Manuel Faria-Azurém	Urban	Traffic
D.Manuel II-Vermoim	Urban	Traffic
João Gomes Laranjo-S.Hora	Urban	Traffic
Seara-Matosinhos	Urban	Industrial
Pe. Moreira Neves-Castelões de Cepeda	Urban	Traffic
Paços de Ferreira	Urban	Background
Francisco Sá Carneiro-Campanha	Urban	Traffic
Sobreiras-Lordelo do Ouro	Urban	Background
Burgães-Santo Tirso	Urban	Background
Ermesinde-Valongo	Urban	Background
Santa Combinha	Rural	Background
Minho-Lima	Rural	Background
Douro Norte	Rural	Background

3.3 PERIOD OF ANALYSIS

This study analyzed concentrations of pollutants 1 January until 30 September 2020. The continuous measurements of PM_{10}, NO_2 and O_3 were provided by the Northern Regional Coordination and Development Commission (CCDR-N). To better assess the impact of lockdown and restrictive measures applied in Portugal, three periods of time were considered: (i) pre-lockdown (January 1–March 14); (ii) lockdown (March 15–June 13); and (iii) post-lockdown (June 14–September 30) when some of the restrictions still applied. Out of the 22 monitoring stations of the CCDR-N network, thirteen registered NO_2 and/or PM_{10} emissions, whereas O_3 was monitored at eleven stations. However, it is necessary to emphasize that not all these stations continuously monitored pollutants throughout the three analyzed periods. For NO_2 only 6 stations registered data during the three periods whereas it was 11 stations and 10 for PM_{10} and O_3, respectively. Table 3 summarizes the respective stations, their characterizations, and measured pollutants.

Table 3: Characterization of monitoring stations and pollutant measurements available (S – suburban, U – urban, R – rural, B – background, T –traffic, I – industrial; X – identifies pollutant measurements at a station)

Station codification	**Characterization**	**PM_{10}**	**NO_2**	**O_3**
Frossos-Braga	SB			X
Anta-Espinho	SB	X	X	X
VNTelha-Maia	SB	X	X	X
Custóias-Matosinhos	SB	-	-	-
Meco-Perafita	SI	X		
Leça do Balio-Matosinhos	SB	X		X
Mindelo-Vila do Conde	SB	X	X	X
Fr Bartolomeu Mártires-S.Vitor	UT		X	
Avintes	UB	X		X
Cónego Dr. Manuel Faria-Azurém	UT	X	X	
D.Manuel II-Vermoim	UT	-	-	-
João Gomes Laranjo-S.Hora	UT	X	X	
Seara-Matosinhos	UI	X		
Pe. Moreira Neves-Castelões de Cepeda	UT	X	X	
Paços de Ferreira	UB	X	X	X
Francisco Sá Carneiro-Campanha	UT		X	
Sobreiras-Lordelo do Ouro	UB			X
Burgães-Santo Tirso	UB	X	X	X
Ermesinde-Valongo	UB		X	
Santa Combinha	RB	-	-	-
Minho-Lima	RB		X	X
Douro Norte	RB	X	X	X

Note: Data was not available for three monitoring stations (14%);

X – identifies pollutant measurements at a station;

S – suburban, U – urban, R – rural; B – background, T –traffic, I – industrial;

4 RESULTS AND DISCUSSION

4.1 NO_2

The examples of weekly trends of NO_2 concentrations during the analysed period in the in urban traffic urban background, suburban and rural stations are shown in Figures (1–3). It is possible to observe that several monitoring stations exhibited discontinuance of the recorded data. Thus, stations where these periods without data were very long or recurrent were not considered in this analysis. Out of the thirteen stations that recorded NO_2 concentrations, six were included in the statistical assessment.

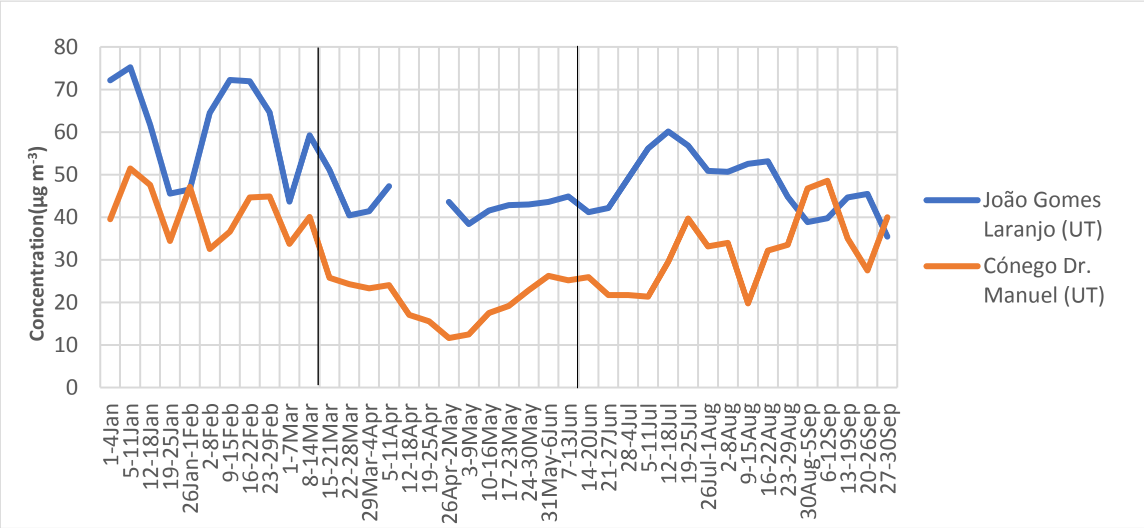

Figure 1: Examples of weekly profiles of NO_2 concentrations during the study period at urban traffic stations

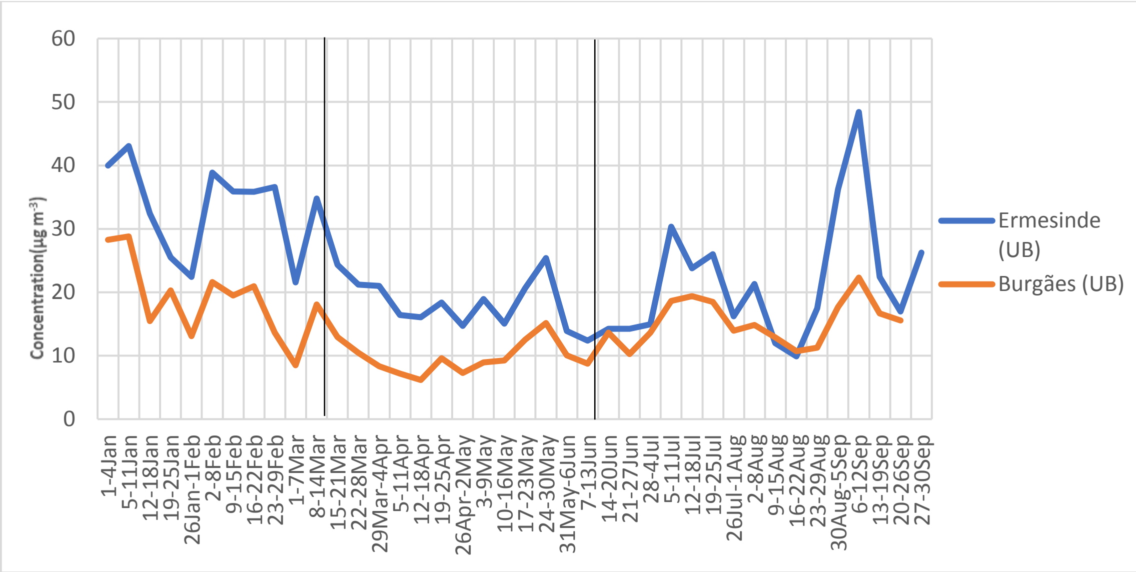

Figure 2: Examples of weekly profiles of NO_2 concentrations during the study period at urban background stations

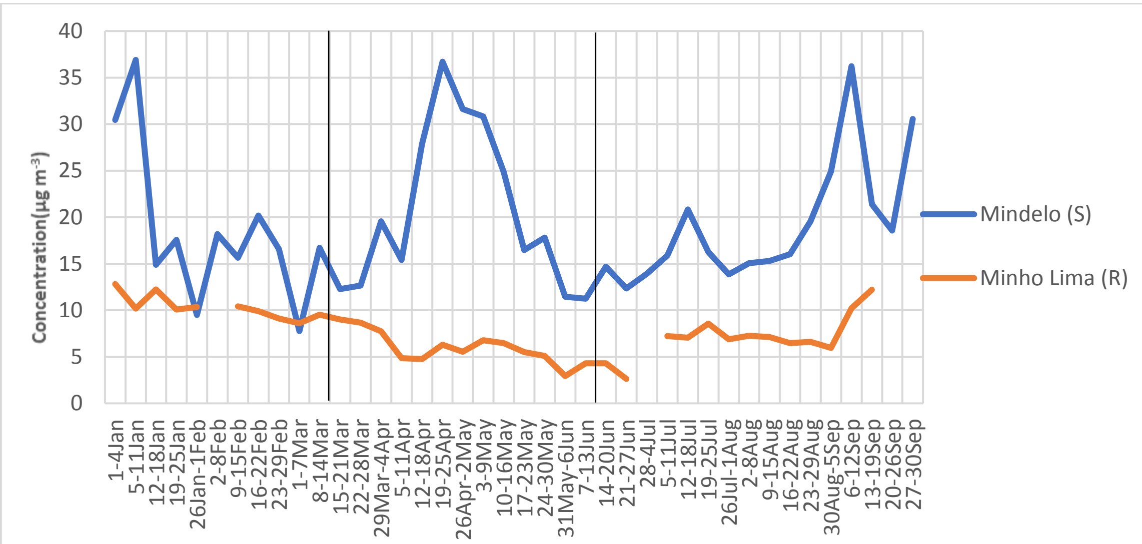

Figure 3: Examples of weekly profiles of NO_2 concentrations during the study period at suburban and rural stations

The statistics of NO_2 levels at all stations with data throughout the three analyzed periods are summarized in Figure 4. These data clearly show that the highest concentrations of NO_2 (during all three periods) were observed at urban sites with influence of traffic emissions. The respective levels were 2.1-1.4 times higher than at the suburban and rural zones. These results are in agreement considering that NO_2 mainly originates from fuel combustion of the road vehicles. Across all the urban traffic sites, the 24 h NO_2 mean was 52 µg m^{-3} (ranging between 41 µg m^{-3} and 62 µg m^{-3}) before lockdown, showing significant reductions during (mean 32 µg m^{-3}; range: 20– 44 µg m^{-3}) and after (mean: 40 µg m^{-3}; range: 32 – 48 µg m^{-3}) lockdown. A similar trend was observed at the urban background stations, where the average 24 h concentration was 26 µg m^{-3} (range: 19 µg m^{-3} and 33 µg m^{-3}) before lockdown, whereas it was 14 µg m^{-3} (9.7 – 18 µg m^{-3}) during lockdown and 34 µg m^{-3} (range: 15 – 53 µg m^{-3}) after lockdown. Overall, the lowest concentrations were observed are suburban sites. Further the results showed that estimated means were not significantly different ($p<0.05$) between the all three periods with the following values: 19 µg m^{-3} (range: 7.8–37 µg m^{-3}) pre-lock down, 21 µg m^{-3} (range: 11–37 µg m^{-3}) during lockdown, and 19 µg m^{-3} (range: 12–36 µg m^{-3}) after lockdown. Finally, at the rural station the average 24 h concentration was 41 µg m^{-3} (33 – 51 µg m^{-3}) before lockdown, 20 µg m^{-3} (range: 12–26 µg m^{-3}) during lockdown and 32 µg m^{-3} (range: 20 – 49 µg m^{-3}) after lockdown.

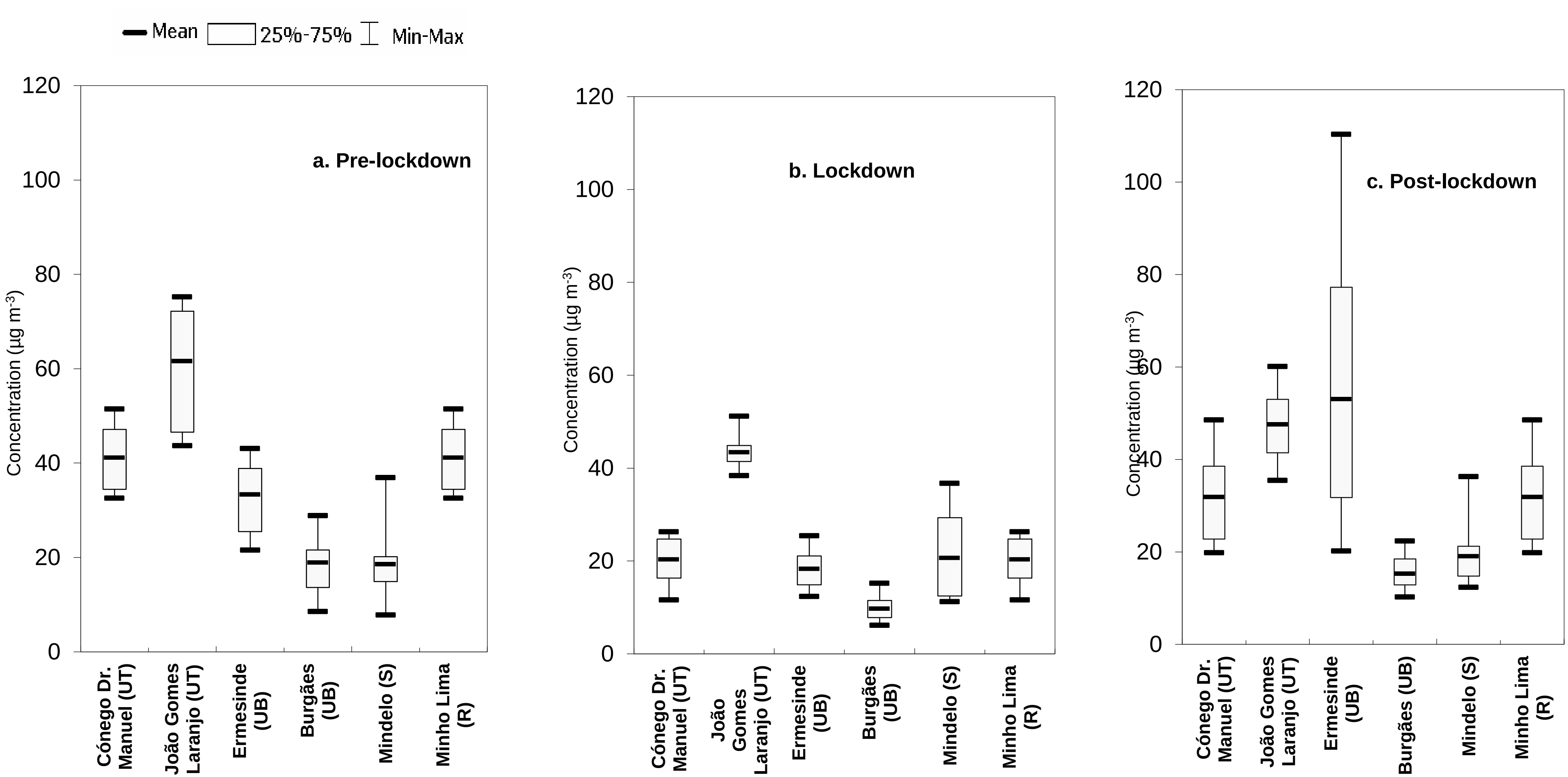

Figure 4: The levels of NO_2 (■ mean; □ 25–75%, and ⌶ max-min) at the monitoring stations during three periods of time: (a) pre-lockdown; (b) lockdown; and (c) post-lockdown). Note: only stations with data registered for all three periods were considered. U–urban, S–suburban, R–rural; B–background, T–traffic, I– industrial

Focusing on the change of levels during the lockdown, the results showed that NO_2 decreased 38% at urban traffic zones, whereas it was 46% decrease at urban background zones and 50% in rural zones (in a comparison with pre-lockdown period). While the decreased levels of NO_2 in the atmosphere of urban zones resulted from direct lesser vehicular traffic on a local level, reduction of NO_2 levels in remote sites were most likely due to the decrease of regional emissions (i.e., lesser traffic movement on national level, limited vehicle movements between municipalities due the imposed restrictions). It is necessary to point out that in post lockdown, after some restrictions were lifted, concentrations of NO_2 significantly increased: at rural stations (56%) and urban background (144%). At urban traffic stations, NO_2 levels increased to a lesser degree (25%), most likely due to the atypicality of that period.

As NO_2 originated from traffic emissions, the decrease of traffic should have resulted in reduction of NO_2 emissions due to the various restrictions imposed during the lockdown. However, it was not possible to observe such impact in a quantitative form in all stations. Inconsistency in data measurements could be the cause. Still, the weekly profiles as well as the overall comparison in NO_2 concentrations showed significant reduction in lockdown in all urban stations as well as the rural station. Similarly, decrease of NO_2 were reported in other European countries and in general, it is estimated that the lockdown in Europe has resulted in a significant decline in NO_2 emissions, corresponding to 37% (Hoang, Huynh, Nguyen, Nguyen, & Le, 2021). On the other hand, the suburban areas were impacted to much lesser degree, though weekly profile of NO_2 (Figure 3) shows some influences of the local restrictions. For example, as demonstrated on Figure 3 sharp increase in NO_2 concentrations occurred on week around 25^{th} of April (holiday in Portugal) with extremely sharp reduction of the respective levels immediately after. This trend was most likely caused by local increase in road transport during the respective weekdays; to limit the pandemic, the circulation between municipalities was prohibited on the respective weekend (corresponding to May 1^{st}) with many citizens traveling outside cities before the weekend.

In the period corresponding to the post lockdown, overall, it was possible to observe an increasing trend of NO_2 concentrations. In majority of the areas (Figures 1-3) NO_2 levels rise in the beginning of September most likely due to the reoccurring daily transport to work and school activities (presential school education, and face-to-face office work and dealing in many sectors).

Evaluating the existent legislation during this study 1 h limit values (200 µg/m^3; 18 exceedances per year permitted) were surpassed on 2 occasions at an urban traffic station (in September 2020, i.e., corresponding to the post-lockdown period). The annual limit of 40 µg/m^3 was surpassed at 3 stations (2 urban traffic stations and 1 suburban station). Though it is necessary to point out that these results have to be implicated carefully, considering that the included period consists of 9 months, i.e., 75% of the year.

4.2 PM_{10}

The examples of weekly trends of PM_{10} concentrations at different zones are presented on Figures 5–7. Similarly to NO_2, several of the monitoring stations exhibited discontinuance of the recorded data. Thus, out of the thirteen stations that recorded PM_{10} concentrations, only eleven were included in further statistical assessment.

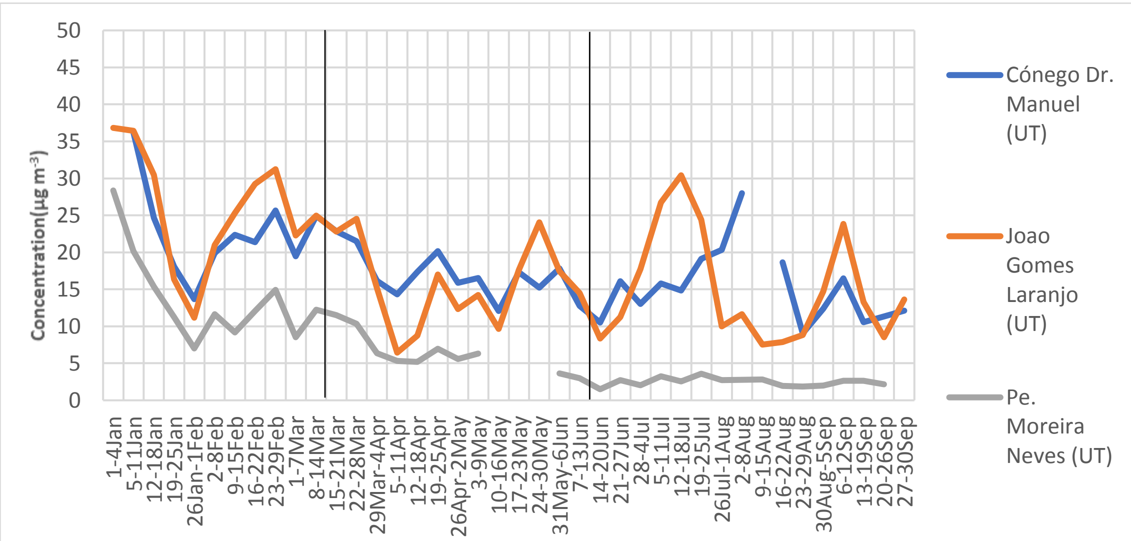

Figure 5: Examples of weekly profiles of PM_{10} concentrations during the study period at urban traffic stations

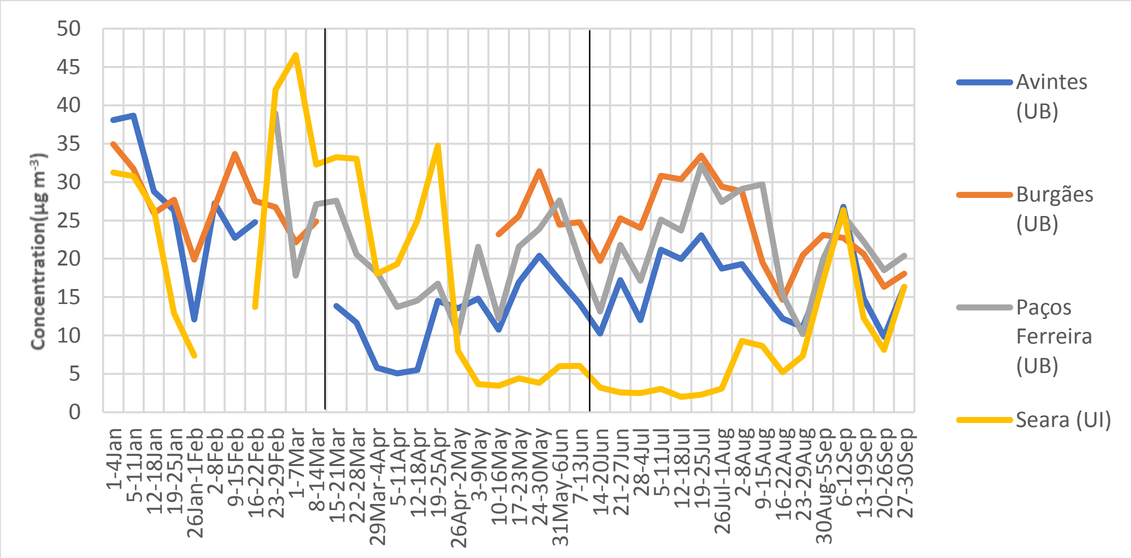

Figure 6: Weekly profile of 24 h PM_{10} during the study period at urban background and industrial stations

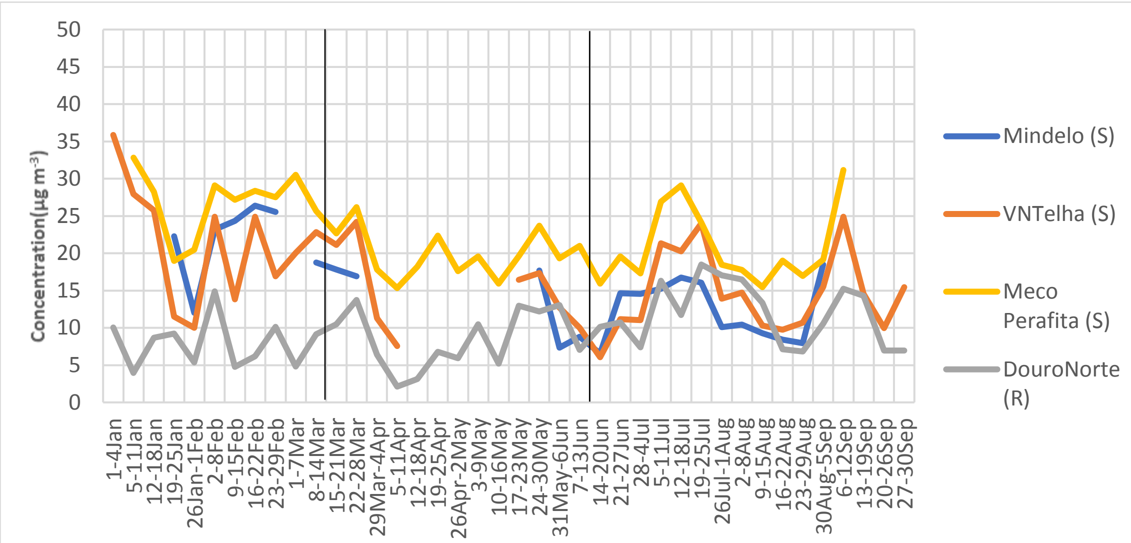

Figure 7: Weekly profile of 24 h PM_{10} during the study period at suburban and rural stations

The statistical summary of PM_{10} levels at all stations throughout the three analyzed periods is summarized in Figure 8. The PM_{10} results were different from those of NO_2. Before lockdown, the highest PM_{10} concentrations were observed at urban background zones (mean of 29 μg m^{-3}; range: 27–31 μg m^{-3}), being followed by levels at industrial (27 μg m^{-3}; range: 7.4–47 μg m^{-3}) and suburban sites (23 μg m^{-3}; range: 21–27 μg m^{-3}). These findings are in agreement with the existing data regarding PM_{10} pollution. On European level the main sources of PM_{10} emissions are commercial, institutional and households' sector (39%) and industrial processes (20%) while road transport contributes only ~11 % (EEA, 2019). Rural areas exhibited the lowest levels (7.9 μg m^{-3};

range: 3.9–15 µg m^{-3}), being approximately 3.6 times lower in a comparison with urban traffic sites.

When the restrictions took place, as well for the post lockdown, PM_{10} levels significantly decreased at the urban industrial site, with mean of 15 µg m^{-3} (range: 3.4– 35 µg m^{-3}) and 8.1 µg m^{-3} (range: 2.0 – 26 µg m^{-3}), respectively. These results showed that at the industrial site, PM_{10} reductions represented 43 and 47% drop, most likely due to restricted industrial production. A similar trend was observed at traffic sites, where the PM_{10} means decreased to 9.8 µg m^{-3} (range: 6.4– 17 µg m^{-3}; 37% drop) during lockdown and to 8.2 µg m^{-3} (range: 3.5 – 15 µg m^{-3}; 17% less) after the lockdown. However, considering urban background site, PM_{10} decreased during lockdown (mean 19 µg m^{-3}; range: 13– 26 µg m^{-3}) but no significant changes were observed after lockdown ended (21 µg m^{-3}; range: 17 – 24 µg m^{-3}). Similar trend occurred for PM_{10} at suburban sites, with significant (30%) reductions during lockdown (16 µg m^{-3}; range: 14– 20 µg m^{-3}) but not significant levels (mean: 16 µg m^{-3}; range: 12 – 21 µg m^{-3}) in atmosphere in the post lockdown. Finally, PM_{10} levels observed at rural sites were not significantly different during lockdown (8.4 µg m^{-3}; range: 2.1–14 µg m^{-3}), but after lockdown ended, the levels increased by 40% (12 µg m^{-3}; range: 6.8–19 µg m^{-3}), most likely due to recurring motor vehicles transport on national level.

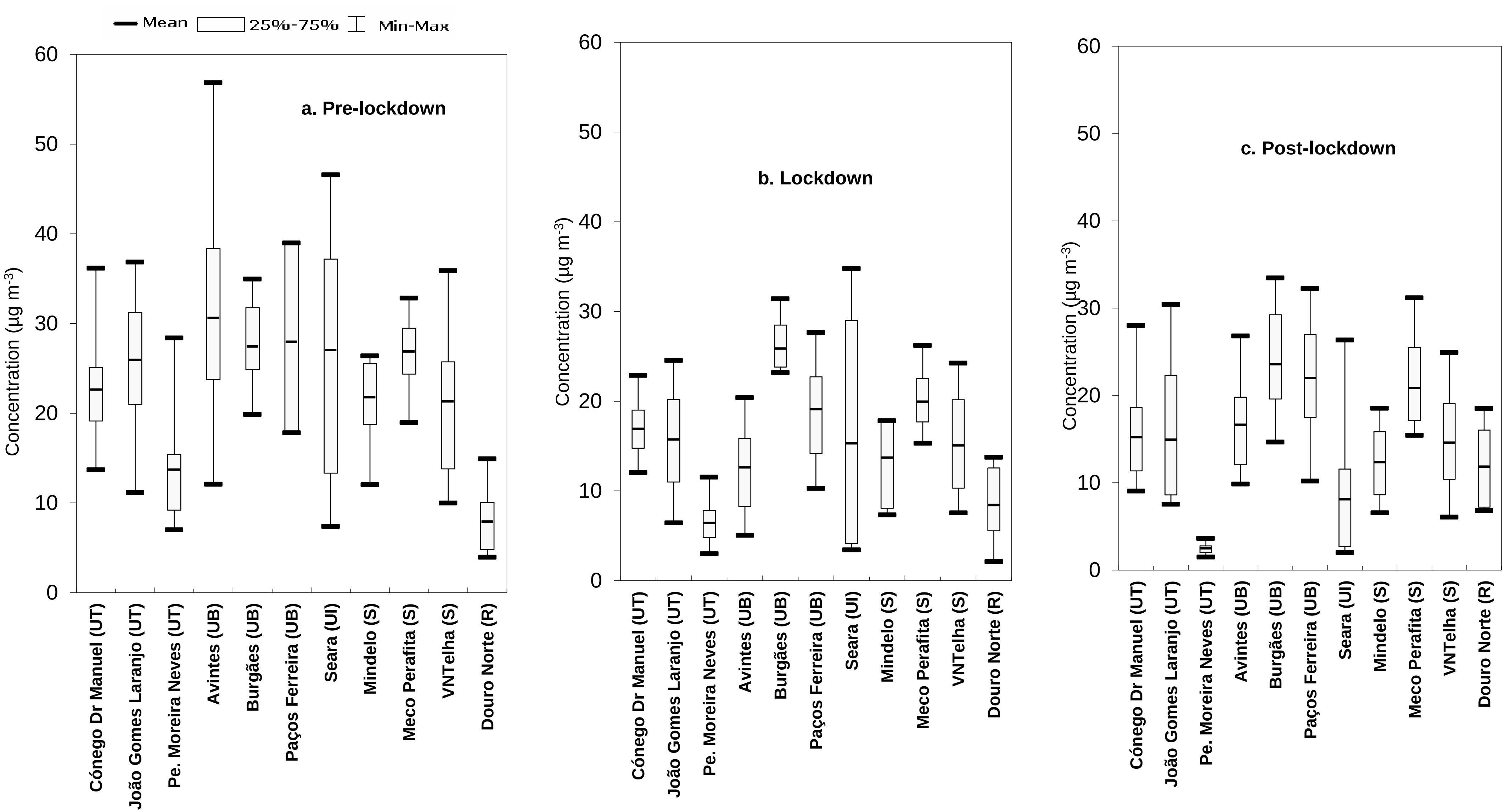

Figure 8: levels of PM_{10} (■ mean; □ 25–75%, and ⌶ max-min) at the monitoring stations during three periods: (a) pre-lockdown; (b) lockdown; and (c) post-lockdown. Note: only stations with data registered for all three periods were considered. U–urban, S–suburban, R–rural; B–background, T–traffic, I– industrial.

Thus, the results showed that the highest decrease in PM_{10} levels is due to lockdown restrictions occurred for the urban industrial sites (43%), with significant reductions also at urban traffic (37%) and background sites (33%), as well suburban sites (30%). In the post-lockdown period, PM_{10} concentration kept decreasing at both traffic (17%) and industrial stations (47%), the latter being most likely caused by the partial recovery of the industrial sector.

Regarding legislative limits for PM_{10}, over the analyzed periods 16 exceedances of 24 h limit occurred, all in the pre-lockdown period. Further, 15 exceedances (i.e. 94%) occurred at urban sites (38 and 31% at urban traffic and background zones, respectively, and 25% at urban industrial) and 1 at suburban areas. These results thus show that PM urban pollution in the North region requires further attention, though the period analyzed in this study consisted of only 9 months, thus implying a careful interpretation.

4.3 O_3

Figures 9–11 present the weekly trends of O_3 concentration at ten monitoring stations that recorded O_3 continuously throughout the whole analyzed period. It is possible to observe that O_3 levels were higher at rural stations (approximately 1.4–1.7 times higher than at urban background and suburban sites, respectively). Tropospheric ozone is a secondary pollutant and therefore is not emitted directly into the air. Higher levels of ozone in rural areas might be caused by lesser O_3 degradation (due to atmospheric reactions and presence of ozone precursors, such as NO, NO_2 and VOCs (WHO, 2005). In addition, as demonstrated by Duan et al., higher solar radiation in summer months favors ozone formations (Duan et al., 2008).

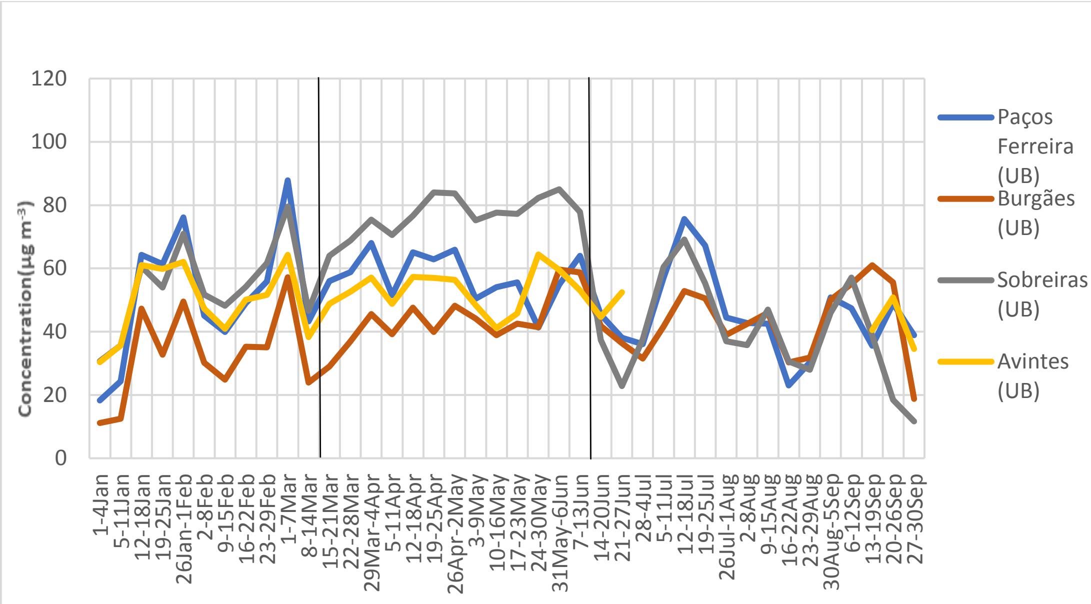

Figure 9: Examples of weekly profiles of O_3 concentrations during the study period at urban background stations

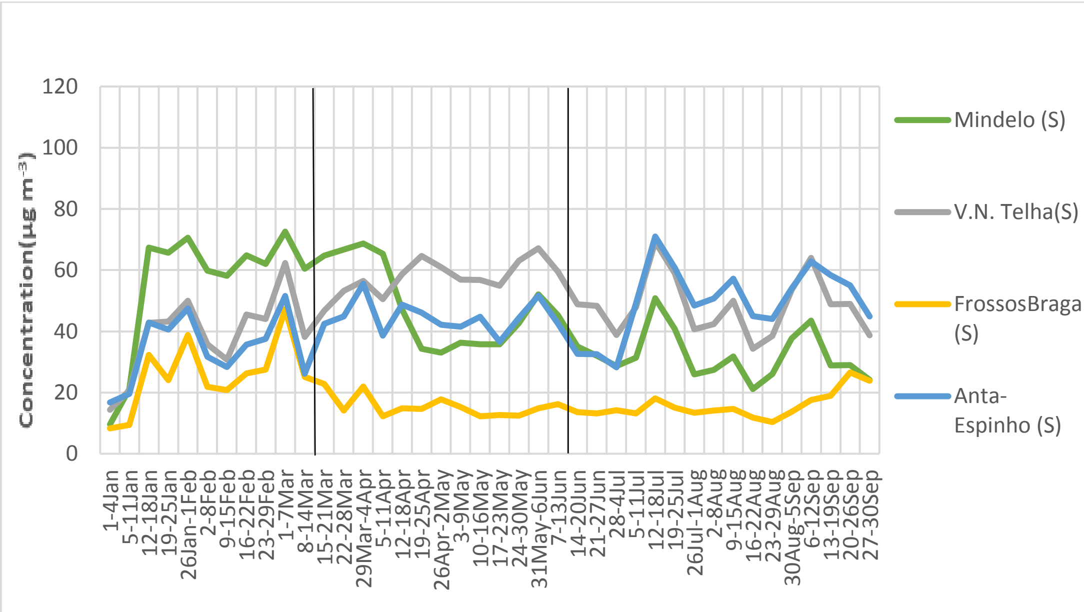

Figure 10: Examples of weekly profiles of O_3 concentrations during the study period at suburban stations

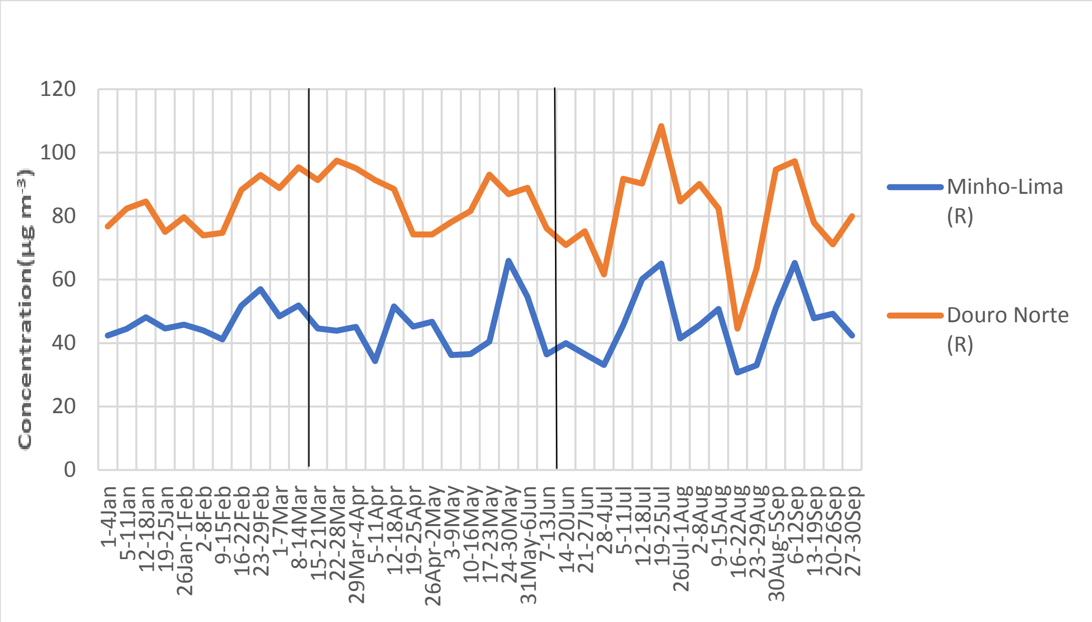

Figure 11: Examples of weekly profiles of O_3 concentrations during the study period at rural stations

The statistical summary of O_3 levels at all stations throughout the three analyzed periods is presented in Figure 12. In agreement with previous results, the highest levels of ozone were observed at rural sites, with a mean of 65 µg m^{-3} (ranging between 47 µg m^{-3} and 83 µg m^{-3}) before lockdown. During the lockdown, no significant changes were observed ($p<0.05$; mean: 65 µg m^{-3}; range: 45–86 µg m^{-3}). Same occurrence was observed for the post lockdown period, (mean 63 µg m^{-3}; range: 46–80 µg m^{-3}), though much larger variation of the levels was observed. The evolution of ozone at the suburban sites during the three period also did not alter. The estimated mean before lockdown was 39 µg m^{-3} (26 µg m^{-3} – 56 µg m^{-3}), with no significant changes during lockdown (42 µg m^{-3}; range: 16–58 µg m^{-3}) and after lockdown (37 µg m^{-3}; range: 16–50 µg m^{-3}). Finally, at urban background stations before lockdown the concentration was approximately 40% lower than at rural sites (47 µg m^{-3}; 33 – 54 µg m^{-3}), increasing 24% during lockdown (mean: 58 µg m^{-3}; range: 44–77 µg m^{-3}) and decreasing 26% after lockdown (mean: 43 µg m^{-3}; range: 40–45 µg m^{-3}). This increase during lockdown period could be due to the lower NO_2 concentrations at urban background stations in that period.

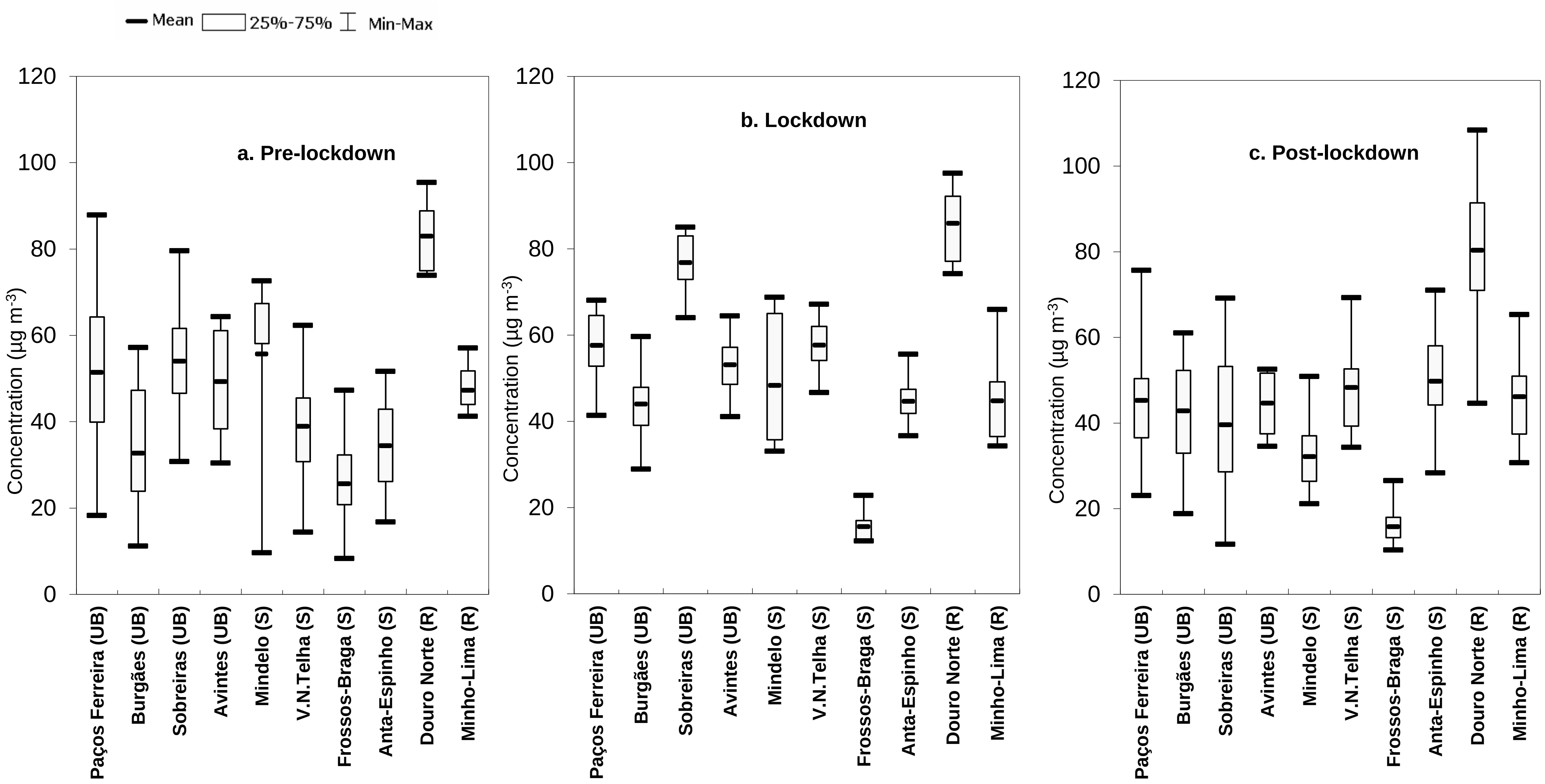

Figure 12: Levels of O_3 (■ mean; □ 25–75%, and ⌶ max-min) at the monitoring stations during three periods: (a) pre-lockdown; (b) lockdown; and (c) post-lockdown. Note: only stations with data registered for all three periods were considered. U–urban, S–suburban, R–rural; B–background, T–traffic, I– industrial

For O_3 the air quality standard for protection of human health is 120 µg m^{-3} (maximum daily 8 h mean), not to be exceeded on more than 25 days/year averaged over 3 years. Over the period of the study (January until September 2020) 64 exceedances of the limit value were registered, with the majority (50%) occurring at rural stations. Suburban and urban background sites exceed the limit value for health protection considerably less: 8% and 42% at suburban and urban background sites, respectively. The Directive 2008/50/CE also sets 1 h information threshold (180 µg m^{-3}) that was exceeded on 11 occasions (6 exceedances at rural sites and 5 at urban background stations). It is noteworthy that levels of ozone at urban traffic sites would be precious due to the traffic restrictions during COVID-19 pandemic but these type of monitoring data were unavailable in the North district (Figure 12).

5 Conclusions and future work

5.1 Conclusions

The results of this study showed that the restrictions implemented to contain the COVID-19 pandemic in Portugal affected all studied pollutants but in a different manner. For NO_2, the urban sites (namely those with strong influence of traffic emissions) showed the highest pollution during all three analyzed periods. The imposed restrictions led to significant decrease of NO_2: at rural stations (50%), urban background zones (46%) and urban traffic stations (38%). As the restrictions were lifted, concentrations of NO_2 significantly increased: at urban background (144%) and rural stations (56%); urban traffic sites seemed to be impacted too (25 %). These values showed that while NO_2 is a pollutant that is associated with traffic emissions, the lockdown restrictions impacted more strongly sites with indirect influence of traffic emissions. Finally, at suburban stations, NO_2 concentrations showed no significant differences between the periods analyzed.

For PM_{10} the trends of pollution during the analyzed periods were different. Firstly, urban background stations were the highest polluted ones throughout the three periods. Secondly, urban industrial site was the most impacted by locked down restrictions with PM_{10} decrease of 43%, being followed by urban traffic sites (37%), urban background sites (33%) and suburban sites (30%). Thirdly, contrary to NO_2, in the post-lockdown period, PM_{10} concentration continued to decrease at both traffic (17%) and industrial stations (47%). Finally, rural areas showed a different trend with no significant changes during lockdown and 41% increase of concentrations in post-lockdown period.

As O_3 is a secondary pollutant its concentration trends differed from both NO_2 and PM_{10}. Firstly, the highest levels of O_3 (1.4-1.7 times than at urban background and suburban sites, respectively) occurred at rural stations; out of the total 50% of the exceedances of O_3 limit value occurred at rural sites. Secondly, rural and suburban sites exhibited similar evolution trends with no significant changes during lockdown and after lockdown. Concentrations at urban background stations, although lower than at rural zones, showed more changes during (24% increase) and after lockdown (26% decrease).

5.2 FUTURE WORK

An important observation made during this study was the discontinuity of the pollutant's measurements in several stations in the North region, which highly limited the number of stations considered in assessment of the pollutants. Thus, in order to provide a more comprehensive understanding about the impacts of the implemented restrictions, the data of this work should include various regions of Portugal, both continent and islands and during the whole calendar year. Furthermore, the analysis of the previous years may help to provide better understanding of the seasonal trends. In addition, the levels and trends of other air pollutants, such as $PM_{2.5}$, CO, SO_2 should be addressed. Future work needs to also focus on countries with different air pollution levels as well as to provide a comparison with those geographical regions that implemented different or less restrictive lockdown measures.

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