Escola Nacional de Saúde Pública
UNIVERSIDADE NOVA DE LISBOA

Outdoor Air monitoring of atmospheric levels of $PM_{2.5}$ in Portugal over the years (2003-2021): First step for effectiveness evaluation of air quality policies in Portugal.

Curso de Mestrado em Saúde Pública

Lorena Falcão Lima

Setembro de 2023

Escola Nacional de Saúde Pública
UNIVERSIDADE NOVA DE LISBOA

Outdoor Air monitoring of atmospheric levels of $PM_{2.5}$ in Portugal over the years (2003-2021): First step for effectiveness evaluation of air quality policies in Portugal.

Dissertação apresentada para cumprimento dos requisitos necessários à obtenção do grau de Mestre em Saúde Pública, realizada sob a orientação científica de Professora Doutora Susana Viegas e Doutora Carla Martins.

Setembro de 2023

Disclaimer

The National School of Public Health, NOVA University Lisbon, is not responsible for the opinions expressed in this publication, which are solely the responsibility of the author.

Acknowledgments

Gostaria de expressar um agradecimento especial às minhas Professoras e Orientadoras, a Professora Doutora Susana Viegas e a Doutora Carla Martins, por terem confiado e apoiado o meu trabalho. Agradeço pela oportunidade que me deram, bem como pela acessibilidade e dinamismo durante meu processo educativo. Sou grata por valorizarem os diversos trajetos pessoais e profissionais, confiarem no meu trabalho e permitirem que eu explorasse áreas afins de interesse, desafiando-me a sair da minha zona de conforto. Acima de tudo, agradeço pelas oportunidades geradas para conhecer novos rumos na minha carreira. Serei eternamente grata.

Agradeço também ao João Santos, que me ensinou muito com sua empatia no processo de esclarecer aspetos relacionados à estatística e análise de dados. Por meio do seu carinho e sabedoria, ele demonstrou que o conhecimento deve ser compartilhado e que títulos não definem pessoas, mas sim o caráter e as escolhas de fazermos o bem todos os dias.

À Doutora Isabel Andrade, agradeço por me auxiliar na reta final do trabalho, oferecendo apoio e atenção para obter uma formatação da versão final da dissertação com sua expertice de qualidade nesta área.

À Sara, agradeço pelo acolhimento e apoio no início do projeto, mostrando-me que o trabalho pode ser leve e divertido (além de ensinar muitas receitas saudáveis e saborosas).

Aos meus amigos e equipa de trabalho no grupo de investigadores do Departamento de Saúde Ocupacional e Ambiental: À Mariana e ao Francisco, meus colegas e parceiros neste projeto, agradeço por conseguirmos aprender e apoiar-nos mutuamente todos os dias. À Ana Margarida, uma profissional incrível, amiga e empática, muito obrigado(a) por todos os ensinamentos, confiança e apoio incondicional. Ao José, por ser um profissional atencioso ao nosso trabalho conjunto. Ao Pedro e à Renata, que sempre foram carinhosos e empáticos nos momentos em que compartilhamos café e conhecimentos na área de Saúde Ambiental.

Aos meus amigos da ENSP, docentes e não docentes, que estiveram em algum momento ao meu lado para apoiar nessa caminhada, gratidão por tê-los encontrado... os levarei eternamente no meu coração.

Dedication

Ao meu Deus e Nossa Senhora, por me guiarem e mostrarem o quão bom podemos ser dia a dia, e que o FAZER O BEM é o caminho…

Aos meus familiares, em especial, ao Atyla, meu marido e parceiro de longos anos, e meus Pais, Afrânio e Verônica, minha Sis, Larissa e minha baby Helena (aind ano ventre, mas já tão amada)…Aos meu queridos Avôs, Stênio e Idel, pelo apoio emocional constante e por acreditarem em mim sempre, demostrando que os sonhos devem ser vividos, sem hipótese de desacreditar em si.

Minha grande família, tios e primos, que sempre fomos tão unidos e fortalecemos nossa corrente de amor e que nunca deixamos que a distância de percorrermos caminhos diferentes para alçar nossos vôos, modifique nossos laços de amor e união!

Vocês foram e são minha combustão diairiamente!

Amo vocês (infinitamente)!!!

Abstract

Introduction: Air pollution, specifically $PM_{2.5}$[1], is one of the leading environmental causes of global mortality, with significant negative effects on health, particularly on cardiovascular morbidity and mortality.

Aim: This study aimed to analyze $PM_{2.5}$ levels in Portugal from 2003-2021, considering different regions, emission influences, and environmental areas, and to compare them with the EU AQD 2008[2] and WHO AQG[3] 2006/2021.

Study Design: Data from the QualAr[4] Portugal platform was obtained for each monitoring station, including levels of $PM_{2.5}$, region, city, station, emission influence, and environment. The percentage of hourly exceedances of EU AQD and WHO AQG of 2006 and 2021 was determined.

Results: From 2003 to 2021, $PM_{2.5}$ levels decreased, with significant differences between Lisbon Metropolitan Area and other regions ($p \leq 0.05$). There were national-level differences in 2015-2016 and 2019–2021, including lockdown months in 2020 and 2021 ($p \leq 0.05$). Stations from urban and suburban areas, as well as traffic and background influence, showed higher significant $PM_{2.5}$ levels when compared to other stations ($p \leq 0.05$). In 2021, atmospheric $PM_{2.5}$ levels exceeded the EU AQD 2008 and WHO AQG 2006 and 2021, ranging from 0.07% to 11.70%, to 0.3% to 37%, and 6% to 72%, respectively.

Conclusion: Lisbon Metropolitan Area had differences among another regions. The flow of people due to restriction measures during COVID-19[5] pandemic caused a reduction in pollution by $PM_{2.5}$. Urban areas and traffic had the highest WHO AQG 2021 exceedance percentage. Despite environmental policies aiming to improve air quality, Portugal experienced periods of exceedance of references levels, making it challenging to meet the ongoing WHO AQD recommended limits.

Keywords: Outdoor air pollution; Particulate matter; Air Quality; Portugal.

Abbreviations:

1. $PM_{2.5}$ – Particulate Matter with less than 2.5 µm of diameters
2. EU AQD – Air Quality Directive 2008/50/EC of the European Parliament
3. WHO AQG - World Health Organization Global Air Quality Guidelines
4. QualAR – Online Air Quality Database
5. COVID-19 – Infectious disease caused by the SARS-CoV-2 virus

Resumo

Introdução: A poluição do ar, especificamente por $PM_{2.5}$[1], é uma das principais causas ambientais de mortalidade global, com efeitos significativos negativos na saúde, principalmente na morbidade e mortalidade cardiovascular.

Objetivo: Este estudo teve como objetivos analisar os níveis de $PM_{2.5}$ em Portugal de 2003 a 2021, considerando diferentes regiões, influências de emissões e áreas ambientais, e comparar com a EU AQD[2] 2008 e as WHO AQG[3] 2006/2021.

Desenho do estudo: Foram obtidos dados da plataforma QualAr[4] Portugal para cada estação de monitorização, incluindo concentrações, região, cidade, estação, influência de emissões e tipo de ambiente. Foi determinada a percentagem de excedências da EU AQD 2008 e das WHO AQG de 2006 e 2021.

Resultados: De 2003 a 2021, os níveis de $PM_{2.5}$ diminuíram, com diferenças significativas entre a Área Metropolitana de Lisboa e outras regiões ($p<0,05$). A nível nacional foram observadas diferenças em 2015-2016 e 2019-2021, incluindo os meses de confinamento em 2020 e 2021 ($p\leq 0,05$). As estações em áreas urbanas e suburbanas, assim como a influência do tráfego e do fundo, apresentaram níveis elevados de $PM_{2.5}$ em comparação com outras estações ($p<0,05$). Em 2021, os níveis atmosféricos de $PM_{2.5}$ excederam os limites estabelecidos pelo EU AQD 2008 e o WHO AQG 2006 e 2021, variando de 0,07% a 11,70%, de 0,3% a 37% e de 6% a 72%, respetivamente.

Conclusão: A Área Metropolitana de Lisboa apresentou diferenças em relação a outras regiões. O fluxo de pessoas como consequência das medidas restritivas durante a pandemia por COVID-19[5] provocou redução da poluição por $PM_{2.5}$. As áreas urbanas e o tráfego tiveram uma elevada percentagem de excedência do WHO AQG 2021. Apesar das políticas ambientais para melhorar a qualidade do ar, Portugal enfrentou períodos de níveis acima do EU AQD, tornando desafiante alcançar os limites recomendados da WHO AQD 2021.

Palavras-chave: Poluição do ar; Partículas Finas; Qualidade do Ar; Portugal.

Abreviaturas:

1. $PM_{2.5}$ – Matéria Particulada de diâmetro inferior a 2,5 µm
2. EU AQD – Diretiva 2008/50/EC da União Europeia sobre a Qualidade do Ar
3. WHO AQG – Diretrizes de Qualidade do Ar Global da Organização Mundial da Saúde – OMS
4. QualAR – Base de Dados Online sobre a Qualidade do Ar
5. COVID-19 – Doenças infeciosas causadas pelo vírus da SARS-CoV2

Table of Contents

List of Figures

List of Tables

List of acronyms

APA - Portuguese Environment Agency

COPD - Chronic Obstructive Pulmonary Disease

CVD - Cerebrovascular Diseases

EEA - European Environment Agency

EPA - Environmental Protection Agency

EU - European Union

GDP - Gross Domestic Product

NUTS - Statistical Territorial Units Nomenclature

PM2.5 - Particles with aerodynamic diameter ≤ 2.5 μm

QualAr – Base de Dados Online da Qualidade do Ar de Portugal

SDGs - Sustainable Development Goals

SPSS - Statistical Package for Social Science

WHO - World Health Organization

WHO AQG - Air Quality Guidelines

EU AQD

1. Introduction

The World Health Organization (WHO) estimates that 7 million deaths per year in the world are attributable to air pollution, with special emphasis on middle and low-income countries[1]. Therefore, air pollution is considered a serious threat to public health globally and is a significant risk factor for the occurrence of negatives outcomes and increasing mortality rates[2,3].

The increase in $PM_{2.5}$ (particles with aerodynamic diameter ≤ 2.5 μm) has been associated in several studies with hospitalization and death, and it has been found that reducing it by 3.9 μg/m^3 could prevent almost 8,000 hospital admissions each year in some countries[2,4,5]. In addition, studies show that air pollution is associated with increased risk of hospitalization rates for congestive heart failure, especially in cases of ischemic heart disease triggered by air pollution[6]. In addition, air pollution was the fourth most important risk factor for mortality, leading to more deaths than high LDL cholesterol, high body mass index, physical inactivity, or alcohol consumption. Worldwide, nearly 20% of cardiovascular disease deaths were attributable to air pollution[7].

Also, in Europe, air pollution is considered the greatest environmental risk, and according to the European Environment Agency[8], 307.000 premature deaths were attributed to chronic exposure to $PM_{2.5,}$ in 2021. In Portugal, particularly, 3.8% of deaths in the year 2017 were attributed to this pollutant, with the main health effects being ischemic heart disease (6.8%), stroke (4.4%), lung cancer (5.6%) and chronic obstructive pulmonary disease (22.6%)[9].

Besides the damage to public health, $PM_{2.5}$ emissions have significant economic impacts, such as the number of hospitalizations, which reduces productivity, because to the lost workdays[8,10,11], as well as the associated medical costs. Air pollution is the leading cause of stroke, cancer and diabetes in the EU, costing authorities an estimated €300-853 billion annually. New Air Quality standards cost less than 0.1% of Gross Domestic Product (GDP) and are at least seven times more effective, with total annual benefits estimated at between €42 billion and €121 billion in 2030, with annual costs less than €6 billion[12].

Considering the available scientific evidence of $PM_{2.5}$ health effects[13–17], in 2021, the WHO reviewed the Air Quality Guidelines (WHO AQG) and proposed the levels of $PM_{2.5}$ to be below than 5 μg/m^3 for the annual average and 15 μg/m^3 for the 24-hour average[18]. Efforts have been developed in Europe to design policies that meet the WHO recommendations, such as the Zero Pollution Action Plan aiming to reduce the number of premature deaths due to exposure to $PM_{2.5}$ by 55% by 2030, as compared to 2005[17]. Additionally, the Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008[19] on ambient air quality and cleaner air for Europe (EU AQD 2008) is currently under review to better align with the WHO recommendations[20].

The main objective of this study is to analyze the concentrations of $PM_{2.5}$ in Portugal between 2003 and 2021, in different regions (NUTS II), by emission influence and environmental areas, and to compare the air monitoring data with the EU AQD 2008 and the WHO AQG 2006 and 2021. This study was part of the project PMCardImpact, “The health and economic impact of $PM_{2.5}$-related cardiovascular diseases in Portugal”, funded by Foundation for Science and Technology (EXPL/SAU-PUB/0944/2017) and corresponded to the first aim of the project achieved. The contents of the present dissertation in what regards the sections Methodology, Results and Discussion, are part of a submission in the journal Heliyon (section Environment), currently under review.

2. Literature Review

2.1. Air Quality and Alignment with the Sustainable Development Goals (SDGs)

Air quality is a critical topic for achieving the Sustainable Development Goals (SDGs) of the United Nations' 2030 Agenda due to air pollution being a global problem to impacts human health and the environment[21]. The SDGs are a set of 17 global goals established in 2015 by the UN, aimed at eradicating poverty, protecting the planet, and promoting prosperity for all[22].

The main SDGs linked to air pollution are Health and Well-being (SDG 3), Affordable and Clean Energy (SDG 7), Sustainable Cities and Communities (SDG 11), Climate Action (SDG 13), and Life on Land (SDG 15)[23]. These goals indicate that reducing the emission of pollutants, particularly $PM_{2.5}$, are crucial for improving population health and can be achieved through various strategies, such as implementing clean energy sources (wind, solar, and hydropower) and promoting[24].

Another criterion that can be adopted is promoting the substitution of fossil fuels to use of public transportation, cycling, and walking to reduce pollutant emissions from fuel-powered vehicles[25]. For this, raising awareness among the population about the effects of air pollution on health and the environment is of great importance in fostering more sustainable practices and promoting a healthier environment[23,24].

Putting in place stricter limits for emissions from industries also significantly supports the reduction of $PM_{2.5}$ emissions, associated with increased incentives for sustainable production and consumption practices, as well as improved waste management[26,27]. Besides, the concern to create in Europe and legislation transposition to other countries shows a align to a world well-been going by according to the SDGs, despite different challenges and impacts[28].

2.2 Exposure to $PM_{2.5}$ and health effects

$PM_{2.5}$ are considered one of the most harmful pollutants to health, occupying the sixth ranking among all the risk factors leading to premature death in the world[10], and the biggest cause of premature death and disease and the major environmental health risk in Europe[29].

According to the World Health Organization (WHO), air pollution is responsible for about 7 million premature deaths worldwide every year. $PM_{2.5}$ in particular is responsible for about 4.2 million of these deaths[30]. Therefore, exposure to $PM_{2.5}$ particles increases hospitalizations[5], emergency department visits, and absenteeism from schools[31] and offices, especially in those with illnesses, the elderly, and children[32].

Fine particles known as $PM_{2.5}$, which are small enough to be inhaled into the lungs, are one of the leading causes associated with a range of health problems[7,33]. Epidemiological studies have linked serious health effects to particles as small as 2.5 microns in diameter, and exposure to air pollution is a leading cause of respiratory, such as cough, breathing, emphysema, pneumonia, lung cancer and chronic respirations infections[34,35]. In patients diagnosed with cardiac diseases, $PM_{2.5}$ can cause the exacerbation of pre-existing pulmonary, anxious, headaches, allergies, eye, noise and throat irritation and cardiovascular diseases[36–38]. $PM_{2.5}$ easily escapes from the nostrils and penetrates deep into the bronchi and alveoli[39,40], corroding the wall of the lung alveoli[41,42] , being responsible for pneumonia and bronchitis[43].

The effect of particles on the human body depends on the duration and concentration of exposure[44]. Particulate matter tends to carry toxic substances with them due to their smaller diameter and increased surface area[45]. Studies indicate that prolonged exposure to these air pollutants may lead to increased negative health outcomes, such as penetration to the respiratory system[16], cornea and conjunctiva, malignant effects[15], cerebrovascular diseases (CVD)[46], chronic obstructive pulmonary disease (COPD)[39], as well as mortality and morbidity in populations[35,47]. It is recognized that outcomes will be influenced by individuals' current health condition, age or genetic factors, duration of exposure, levels of exposure, and influences from different types of emissions[25,48–50].

In relation to cerebrovascular diseases, $PM_{2.5}$ can cross the blood-brain barrier, which is responsible for protecting the brain from harmful substances present in oxidative stress, leading to endothelial dysfunction, impairing the inner lining of blood vessels. This vascular dysfunction can increase the risk of blood clot formation and, consequently, stroke and other forms of cerebrovascular disease (CVD)[47]. Large cohort studies from both high- and low-income settings show increased cardiovascular disease incidence and mortality in association with particulate matter levels [41,51]. In addition, exposure to $PM_{2.5}$ can increase blood pressure, another important risk factor for CVD[52]. Elevated blood pressure damages the cerebral arteries, can lead to stroke or cerebral hemorrhage, and increases risk of atherosclerosis[3,53].

A study analyzed data from 114 epidemiological studies and concluded that short and long-term exposure to $PM_{2.5}$ was significantly associated with an increased risk of cardiovascular diseases, including ischemic heart disease, stroke, and peripheral arterial disease[21,48,54]. The study also showed that exposure to this pollutant was associated with a reduction in life expectancy in various populations. This evidence agreed with others showing that exposure to $PM_{2.5}$ is associated with a significant reduction in endothelial function, which is considered an important marker of cardiovascular health[42,55]. Endothelial dysfunction is a change in the function of endothelial cells that can occur in response to inflammation, oxidative stress, and other conditions. This can contribute to the development of cardiovascular diseases, as these cells are responsible for regulating blood flow and blood pressure[33].

$PM_{2.5}$ exposure is a significant risk factor for health outcomes in Europe as well. A study conducted in 41 European countries concluded that exposure to this pollutant was associated with a significant increase in the risk of death from pulmonary and cardiovascular diseases in all evaluated locations[56].

2.3. Measures in places to control $PM_{2.5}$ emissions

Due to the severe health consequences, many countries worldwide have taken measures to control the emission of PM in the environment, particularly $PM_{2.5}$, recognizing the health risks associated with this pollution. China has adopted measures to reduce air pollution with the aim of improving public health. These policies include emission limits for industries, programs for replacing old vehicles, and changes in the characteristics of fossil fuels used[53,57]. The United States, through the Environmental Protection Agency (EPA), has established national air quality standards for $PM_{2.5}$ and has worked with States to implement programs for reducing pollutants[21,45].

The European Union (EU) has set ambitious goals to improve air quality, which includes policies that determine emission limits for vehicles and industries[58]. The main goal of the directive is to improve ambient air quality in Europe, protect human health, and contribute to cleaner air for Europe as a whole. Considering the health effects triggered by pollution in Europe, the European Union has established maximum limits for $PM_{2.5}$ concentration in ambient air. According to the legislation, the annual average concentration of $PM_{2.5}$ should not exceed 25 micrograms per cubic meter (μg/m^3), and the daily average concentration not cited[19], as showed in Table 1.

Table 1. Recommended and legislated atmospheric levels for particulate matter in ambient air, respectively, from WHO AQG 2006 and 2021[18], and EU Directive 2008/50.[19]

$PM_{2.5}$ levels average		Annual average levels (μg/m³)	Daily average levels (μg/m³) (24 h) (μg/m³)
Recomendation WHO AQG	2006	10	25
	2021	5	15
EU Directive 2008/50		25	NA*

*NA: No application

Most European countries have adopted measures to control $PM_{2.5}$ emissions. An example is Poland, which has high levels of pollution due to its heavy reliance on fossil fuels for industrial activity and a significant coal industry[58]. Other European countries that have not yet effectively implemented air pollutant reduction measures are Bulgaria, Kosovo, and Bosnia and Herzegovina, which are also heavily dependent on fossil fuels and face challenges in modernizing their energy production infrastructure to reduce emissions[59].

It is important to emphasize that air pollution is a global problem and reducing $PM_{2.5}$ exposure can help prevent various diseases, mobility, and mortality. Furthermore, the implementation of public policies to improve air quality is a crucial measure to protect the health of populations. Therefore, to reduce emissions, the European Union has implemented several measures, including legislation on vehicle emissions, reduction of emissions from coal power plants, and promotion of renewable energy sources. Currently, European Union countries are required to present air quality plans to reduce air pollution[56].

In Portugal, the air quality is monitored by the Portuguese Environment Agency (APA), which has an extensive network of monitoring stations throughout the country.

These stations use advanced technology to measure concentrations of $PM_{2.5}$ and other air pollutants. Data is regularly shared with the public and authorities to improve air quality. Accurate measurement of $PM_{2.5}$ particles is essential to protect human health, as high concentrations of $PM_{2.5}$ particles are associated with various adverse effects mentioned above. Identifying problem areas and implementing corrective actions are key to protecting public health in Portugal. Is this way, APA develop an essential role in the coordination and management of air quality in Portugal. Its main functions include establishing norms and regulations for air quality, overseeing the implementation of the monitoring system, disseminating information to the public and coordinating cooperation with other entities to improve air quality and protect the health of the population.

The air quality policy in relation to fine particles in Portugal has gone through a timeline of development and transposition over the years. Initially, the country adopted internal measures to monitor and control the concentration of these particles in the air, seeking to protect the health of the population. With the advancement of environmental legislation in the European Union, Portugal has followed this path establishing guidelines and transposing regulations.

The transposition of European Union directives into national legislation has been a key step in the implementation of specific policies to control fine particles, including $PM_{2.5}$. These measures aim to ensure compliance with established air quality standards, as well as to promote continuous monitoring actions, dissemination of information and collaboration with other entities to improve air quality in Portugal.

3. Hypothesis

The main hypothesis of this work is that the levels of fine particulate matter ($PM_{2.5}$) decrease over the years in Portugal, with statistically significant differences observed across different regions from 2003 to 2021.

Thus, the second hypothesis is that Portugal have kept $PM_{2.5}$ levels within the parameters allowed by current European Union legislation (2008/50EC) but has had challenges in keeping up with the levels recommended by the AQG WHO.

4. Objectives

4.1. Main objective:

The main objective of this work is to analyze the concentrations trends of $PM_{2.5}$ in Portugal between 2003 and 2021, in different regions (NUTS II), by emission influence and environmental areas, and compared with the guidelines defined by the WHO and in the current legislation at the European Union.

4.2. Specific objectives:

- To analyze available data in Portugal regarding air monitoring parameters and develop a dedicated database in Excel;
- To collect air monitoring data from the monitoring platform QualAr for the years 2003-2021;
- To analyze differences of atmospheric levels of $PM_{2.5}$ by environmental areas and by emission influence by NUTS II;
- To analyze differences of atmospheric levels of $PM_{2.5}$ during different restriction periods due to COVID-19 in Portugal;
- To determine the exceedance of atmospheric levels of $PM_{2.5}$ in 2021, considering WHO AQG guidelines and European Union directive for $PM_{2.5}$.

5. Methodology

5.1. Study design

This is a descriptive observational and ecological study.

5.2. Study Area

The study area is Portugal, a country with highly heterogeneous environments, and it was considered the levels of $PM_{2.5}$ by areas according to the division proposed by the Statistical Territorial Units Nomenclature (NUTS), at NUTS II level, which divides the country into seven regions: North, Center, Lisbon Metropolitan Area, Alentejo, Algarve, Azores and Madeira Autonomous Region, as showed in the Figure 1[60].

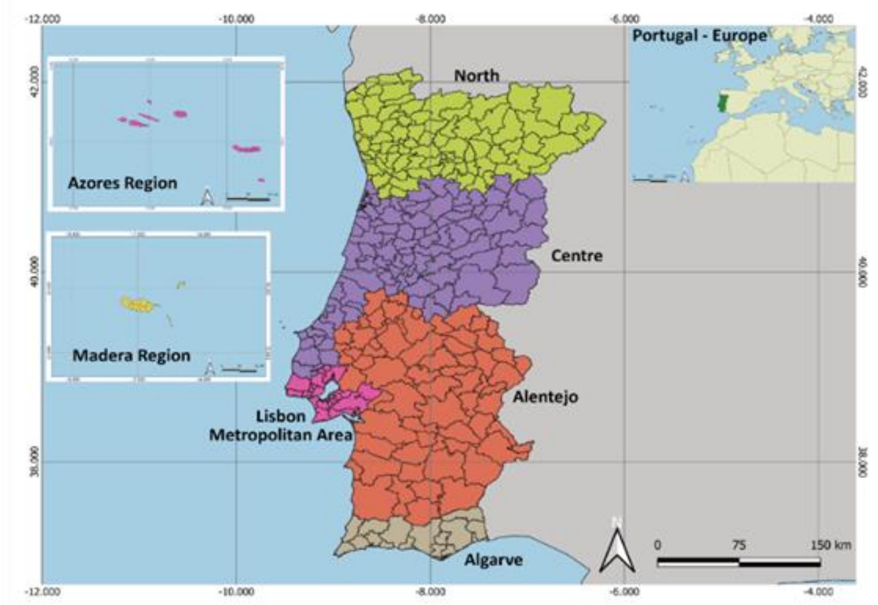

Figure 1. Map of location in the NUTS II subdivision of the regions used for air quality monitoring data. Map obtained from division from INE.

According to European Parliament and the Council Regulation nº 1059/2003[56], the division by these seven regions indicate the statistical areas and sub-regions into which the Portuguese territory is split.

5.3. Air Monitoring Database of $PM_{2.5}$

This study developed a population-level analysis of $PM_{2.5}$ levels in Portugal, by NUTS II, using concentration values obtained from the online database Informação sobre Qualidade do Ar (QualAr) from the Portuguese Environment Agency (APA), reported between January 2003 and December 2021.

The database QualAr provides hourly levels of different pollutants. The monitoring stations are classified according to the environmental area they are located (urban, suburban, and rural) and emission influence (traffic, industrial, and environment).

The daily average (24-hour) concentrations of $PM_{2.5}$ were calculated based on the hourly observed values at the stations. The annual average of $PM_{2.5}$ was estimated based on the concentrations of each station, using the previous 365 days. The study design is described in Figure 2.

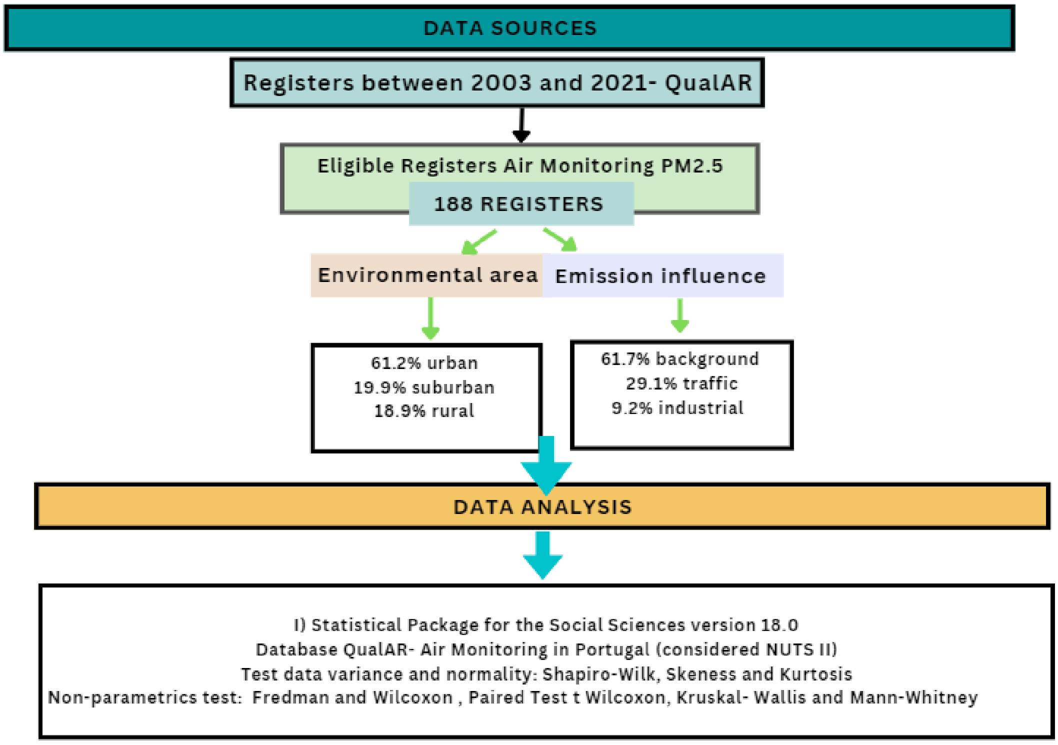

Figure 2. Study Design Flow Chart.

The Ambient Air Quality Directive[19] sets, for compliance purposes, the objective of a minimum data capture of 90 % for monitoring stations. For this assessment purposes and in line with previous publications coverage of 75 % allows more stations to be considered without a significant increase in monitoring uncertainties[59]. Thus, to ensure the uniformity of the results, only data obtained with a minimum reporting percentage of 75% were used in this study. The monitoring stations were distributed across the seven regions and covered 72 registered cities in total.

For the year 2021, it was calculated the exceedance of the reference values of Air Quality Directive and WHO Air Quality Guidelines. The $PM_{2.5}$ exceedance values were calculated hourly during the year for the 30 stations considered on a qualitative basis, excluding the stations without levels registered.

5.4. Statistical analysis

The descriptive analysis of all variables was performed, considering exposure results expressed as mean, median, maximum and percentiles for quantitative variables. The distributions were subjected to the Shapiro-Wilk normality test and Kurtosis. Non-parametric tests were considered due to the non-normal distribution of the sample. Data were described by NUTS II, environmental area, and emission influence. Kruskal-Wallis test and Mann-Whitney U test were used for independents samples, and Friedman and Wilcoxon tests were used for paired samples. The software used was Statistical Package for Social Science (SPSS) version 28. It considered the statistically significant differences between the groups and different years, with a significant level of 5% (p-value ≤ 0.05).

5.5 Spatial analysis

The locations of the stations within each region were identified and managed in the QGIS software version 3.16.0 (2019). For the maps of location and spatial distribution of exposure levels to $PM_{2.5}$, shapefiles with format obtained from the European Environment Agency (EEA) website and executed in QGIS version 3.16.0 (2019) software were used, following the WGS84 system with EPSG: 4326 reference code, which adds Greenwich as the starting point for longitude (0°). The maps were assigned a color intensity gradient according to the atmospheric levels of $PM_{2.5}$ for each region evaluated, drawing a comparison between data obtained in 2009 and 2019 (ten-year interval for the period pre-COVID 19 pandemic). The legend of the maps considered the average annual limit values according to the current EU Air Quality Directive (25 μg/m^3), WHO Air Quality Guidelines 2006 (10 μg/m^3) and WHO Air Quality Guidelines 2021 (5 μg/m^3).

6. Results and Discussion

6.1. Characteristics of the eligible air monitoring data in Portugal

A total of 188 eligible air monitoring data considered. We obtained the results of seven monitoring stations, considered at least ≥75% of the registers. These registers were obtained from the seven stations as the main criterion for evaluating $PM_{2.5}$ concentrations, between 2003 and 2021, and represented approximately 14% of the total records in the period. The distribution by environmental influence was 61.2% urban, 19.9% suburban and 18.9% rural, and by emission influence was 61.7% background, 29.1% traffic and 9.2% industrial (Figure 3).

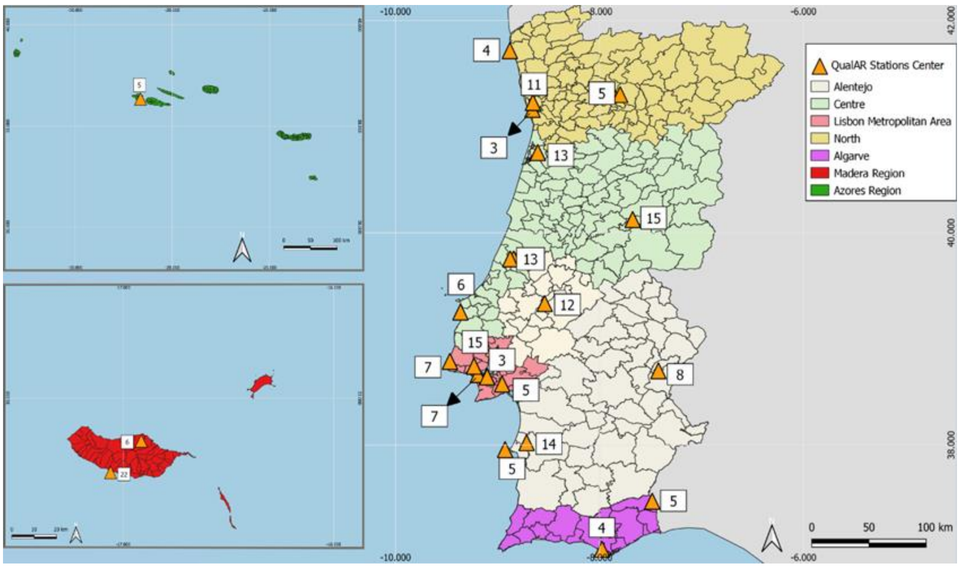

Figure 3. Geographic distribution of eligible air monitoring data. Data represents distribution by NUTS II, according to INE.

Despite the exclusion of registries, similarities were observed between the eligible and non-eligible stations, mainly in relation to the number of records related to each area and influence, confirming the robustness of the dataset. Regarding the non-eligible monitoring stations due to registrations <75% (n=1172), they presented the following proportional distribution: 53% urban stations, 14% suburban stations, and 11% rural stations (environmental influence), and 75% of background, 15% of traffic, and 10% of industrial influence (emission influence).

6.2. Trends of atmospheric levels of $PM_{2.5}$ over 2003-2021 in Portugal

Figure 4 shows that, over the years 2003-2021, the number of air monitoring data with registries with at least 75% has progressively increased, thus representing an effort at national level to increment the air monitoring data available, as a response to the implementation of European air quality policies. It is also observed that, globally for Portugal, the mean levels of $PM_{2.5}$ have been decreasing throughout the years.

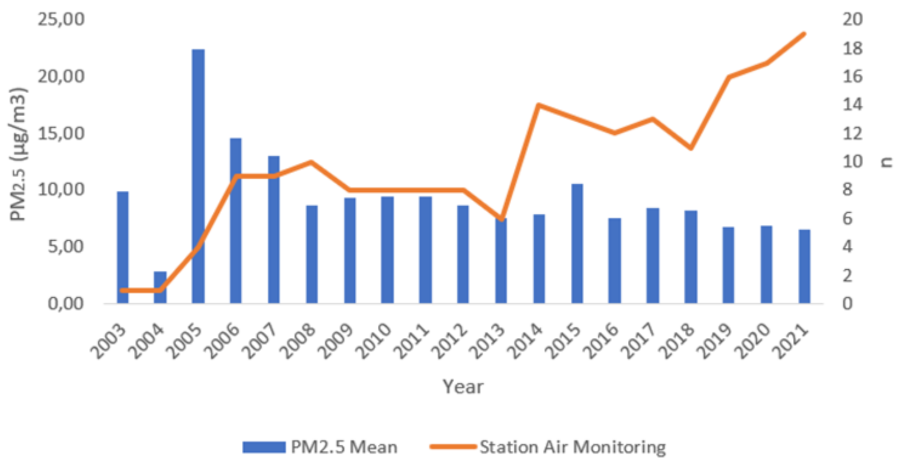

Figure 4. Air monitoring data considering atmospheric levels of $PM_{2.5}$ (µg/m^3) (primary Y-axis) and number of eligible stations (n) (secondary Y-axis) in Portugal, in the period 2003-2021. Data represents yearly mean values collected from QualAr.

Over the years (2003-2021) it is perceived an increment in air monitoring and control measures for $PM_{2.5}$ as a response to the implementation of European air quality policies. This resulted in a reduction of $PM_{2.5}$ average levels over the years.

The European Environment Agency (EEA) yearly-reports on air quality in Europe, which include data on $PM_{2.5}$ levels across the EU Member States, showing that Portugal has implemented air quality, control, and monitoring strategies[53]. The most recent EEA report for 2021 indicates that $PM_{2.5}$ levels across the EU have been gradually declining in recent years, and Portugal follows this trend[61]. Other authors have previously referred this reduction trend of atmospheric pollutants in Portugal over the years[9,62].

Driving factors like the Ambient Air Quality and Clean Air for Europe Directive 2008/50/EC (currently under review)[19], Directive 2016/2284 on the reduction of national emissions[26], the Paris Agreement[61], the European Green Deal[63] and the Zero Pollution Action Plan[64], have been of utmost importance for promoting the decrease of air pollution in Portugal, such as the levels of $PM_{2.5}$.

In addition, Portugal has invested in energy efficiency programs to reduce energy consumption in buildings and the transportation sector, such as the creation of a National Hydrogen Strategy[65]. Moreover, in 2020, about 60% of the electricity produced in Portugal came from renewable sources such as solar, wind, and hydropower[66].

However, the atmospheric levels of $PM_{2.5}$ over the years are variable as demonstrated by the monitoring results. The heterogeneity of concentrations demonstrates that different areas and different emissions must have specific and dedicated policies and be continuously monitored.

Table 2 shows the mean and median levels of $PM_{2.5}$ through the years for Portugal by NUTS II regions.

Table 2. Atmospheric levels of $PM_{2.5}$ (μg/m^3), in Portugal, in the period 2003—2021, by NUTS II regions, according to data obtained from QualAr. Results presented as mean (median) atmospheric levels of $PM_{2.5}$.

Year	Portugal	North [a]	Center	Area Metropolitan Lisbon [a]	Alentejo [a]	Algarve [a]	Azores [a]	Madeira [a]
2003	9.96 (8.00)				9.96 (8.00)			
2004	2.82 (2.00)	2.82 (2.00)						
2005	22.44 (15.89)	27.15 (18.00)	23.35 (17.67)		14.97 (12.00)			
2006	14.63 (10.83)	14.59 (10.33)	16.45 (11.33)					12.84(11.00)
2007	13.00 (10.12)	14.24 (11.00)	14.51 (11.00)		11.17 (10.00)			9.80 (8.50)
2008	8.71 (6.83)	6.72 (4.00)	10.14 (7.00)		9.72 (8.00)			8.93 (8.33)
2009	9.38 (7.25)	8.95 (4.50)	10.98 (8.50)		9.61(8.00)			8.52 (8.00)
2010	9.44 (7.58)	8.44 (4.50)	9.34 (6.50)		12.89 (11.00)			9.03 (8.33)
2011	9.52 (6.66)	5.55 (3.00)	13.21 (9.00)		9.94 (8.00)			
2012	8.70 (6.11)	5.07 (2.67)	12.46 (8.67)		8.54 (7.00)			
2013	7.61 (4.16)	4.7 (1.00)	9.92 (6.67)		8.42 (7.00)		2.68(2.00)	
2014	7.87 (6.25)	7.11 (5.00)	7.42 (4.67)	8.83 (6.67)	9.48 (6.67)	7.93 (6.50)	2.92 (8.00)	
2015	10.60 (7.96)*		9.24 (6.00)	12.78 (10.54)	9.19(7.67)	9.28 (7.65)		
2016	7.56 (5.96)*		6.70 (5.00)	10.52 (8.87)	7.69 (6.13)	6.85 (5.00)	8.79(8.00)	3.03 (2.80)
2017	8.51 (6.94)		6.06 (4.50)	11.38 (9.23)	8.23 (6.50)	8.30 (6.50)		5.84 (8.00)
2018	8.19 (5.16)		7.19 (6.00)	11.35 (9.45)	6.88 (6.00)			4.99 (4.35)
2019	6.82 (4.53)*		7.56 (5.50)	9.22 (6.72)	5.38 (4.70)		3.44(2.90)	3.94 (2.85)
2020	6.95 (5.07)*		5.99 (4.33)	9.89 (8.13)	5.68 (4.50)			4.30(3.33)
2021	6.53 (4.29)*		9.10 (6.25)	8.02 (6.06)	5.75 (5.00)	3.98 (3.33)	2.50(2.00)	4.55(3.13)

[a] Statistically significant differences considering results for each region by all years, $p \leq 0.05$.

*Statistically significant differences considering yearly mean results for each region by year, $p \leq 0.05$.

Data represents yearly mean (median) values of $PM_{2.5}$ (μg/m^3) collected from QualAr.

The annual mean levels of $PM_{2.5}$ observed in the present study showed several fluctuations over the years and regions. Statistically significant differences were found between Lisbon Metropolitan Area, and the other regions of the North, Algarve, Alentejo, Madeira and Azores ($p<0.05$).

The levels in the Lisbon Metropolitan Area are justified by the fact that it is an urban area with a high population density, and with intense emissions due to human activities and mobility. In addition, in other studies[9,67] there were also fluctuations in the mean levels by different emissions sources and environmental area, in Portugal. Thus, the civil construction services and traffic flow presented higher impact, as also highlighted in study from Zhao et al.[11], where it was concluded that the impacts of urban size and form varied by city size and geographical location. This emphasizes the need of investments in green space, green routes, and outdoor recreational areas, for a more sustainable mobility management, as referred by Reche et al[68].

Figure 5 showed the spatial changes over time in different NUTS II regions, represented as a Kernel map, where it is observed a transition and reduction in air pollution over 2009 and 2019 (ten years after the first year), being considered normal periods of $PM_{2.5}$ emissions in Portugal, and before COVID pandemic. The Figure 5 showed atmospheric levels by NUTS II for the year 2009 (North, Center, Alentejo, Madeira) and the year 2019 (Center, Lisbon Metropolitan Area, Alentejo, Azores, Madeira) considering an average central point in each region.

a) b)

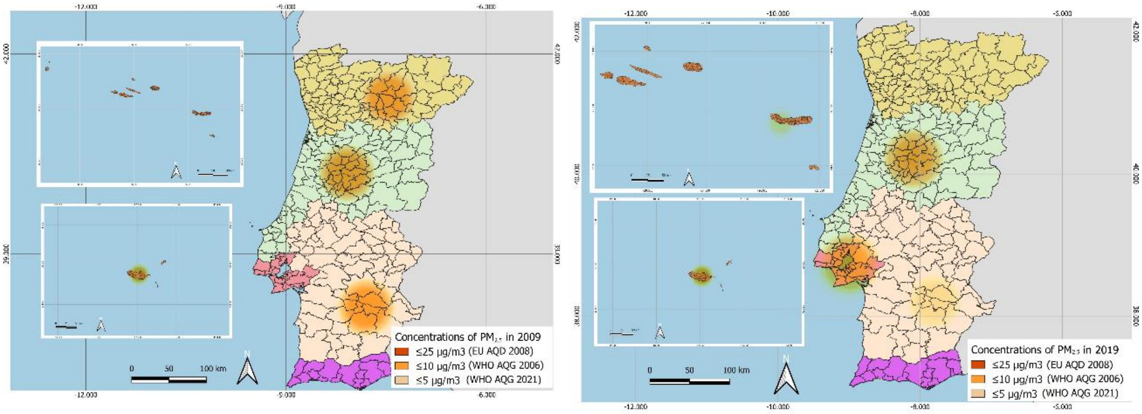

Figure 5. Kernel map showing the yearly-mean levels of $PM_{2.5}$ ($\mu g/m^3$) for each NUTS II, for the year 2009 (a), 2019 (b), in Portugal, according to data obtained from QualAr.

The heat maps allow visualizing this reduction for the regions Center, Alentejo and Madeira showing that, within the country, efforts to achieve a better air quality are producing results that can be beneficial to environment and population health. For the other regions, data were only available for one of the years considered (Table 1). The anthropogenic emissions of air pollutants are the key drivers behind atmospheric levels of PM and other atmospheric pollutants. In 2009, just the Center region showed levels of $PM_{2.5}$ above 10 $\mu g/m^3$ (Figure 3, a). The map (b) shows that, in 2019, all regions kept levels below 10 $\mu g/m^3$ (Figure 3, b), as can be verified by the intensity of coloring on the maps that corresponds to the average concentration values.

Regarding the years, at the national level, there were statistically significant differences between 2015 - 2016 and 2019 - 2021 ($p \leq 0.05$). Also, the year 2005 marked the highest average emission of $PM_{2.5}$ recorded (Figure 5, a), as well as shown in Table 1.

For the years 2020 and 2021, there were statistically significant differences between the months of lockdowns (March, April, May of 2020) and of restrictions (January, February, March of 2021), when compared with similar periods of the years 2017, 2018 and 2019($p \leq 0.05$). As mentioned by Heydari et[50,69], these periods showed a reduction in traffic-related emissions when compared to 2019, attributed to the adoption of “stay at home” policies due to the COVID-19 pandemic. Due to the COVID-19 pandemic restriction and lockdown imposed worldwide, air quality data improved in many European cities[14], thus demonstrating well the importance of traffic as an emission source.

6.3. Comparation of atmospheric levels of $PM_{2.5}$ with EU Air Quality Directive (2008) and WHO Air Quality Guidelines (2006 and 2021)

Figure 6 showed the distribution of mean atmospheric levels of $PM_{2.5}$ considering the environment area and the emission influence, and the comparison with the limits of EU Air Quality Directive 2008 (25 µg/m^3), WHO AQG 2006 (10 µg/m^3) and WHO AQG 2021 (5 µg/m^3) for $PM_{2.5}$. The decreasing trend of mean levels of $PM_{2.5}$ through the years for the different environmental areas and emission influences is demonstrated.

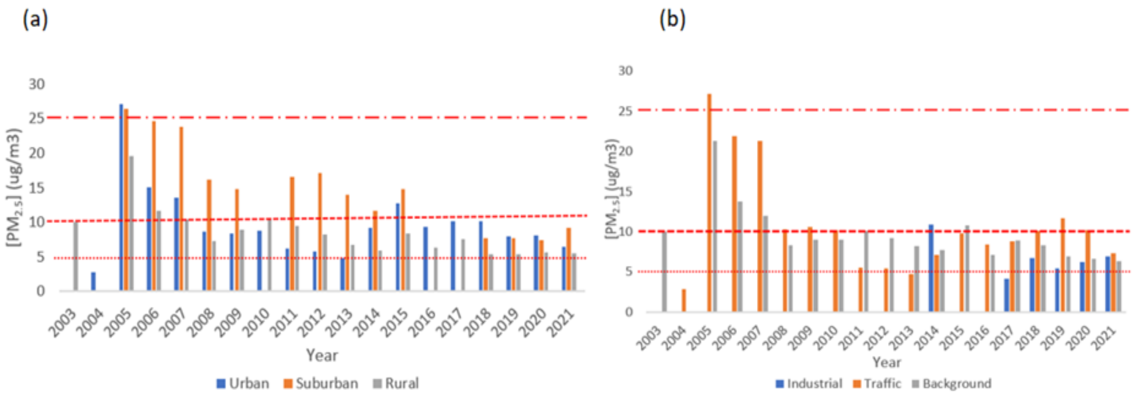

Figure 6. Mean levels of $PM_{2.5}$, by environmental area (urban, suburban and rural) (a) and emission influence (industrial, traffic, background) (b) in Portugal, in the period 2003–2021. Data represents yearly mean values collected from QualAr. Limits of EU Air Quality Directive (25 µg/m^3), WHO Air Quality Guidelines 2006 (10 µg/m^3) and WHO Air Quality Guidelines 2021 (5 µg/m^3) for $PM_{2.5}$ are included as dashed lines.

Urban and suburban areas presented statistically significant higher levels of $PM_{2.5}$ when compared to rural areas ($p<0.05$). These areas are influenced by human activities, being confirmed by the higher levels of $PM_{2.5}$ registered by stations of traffic influence when compared with background and industrial influence ($p<0.05$). The study by Błaszczak et al[70] was conducted in Poland and showed the same differences namely that, in general, urban and suburban stations present higher concentrations of $PM_{2.5}$ than rural stations.

Between 2019 and 2020, the reduction in $PM_{2.5}$ levels was observed in urban areas while an increase was observed in suburban and rural areas. This modification could be justified by COVID-19 pandemic and by people moving to rural areas. Consequently, urban zones experienced higher reductions than rural zones, a pattern also observed in other countries[71]. Moreover, the oscillations in different areas can be explained by the positioning of the monitoring stations, which may be closer to industries or other sources of pollution, as well as may have been influenced by dust winds or even fires in the period considered[72].

Considering the emission influence, statistically significantly differences were found for atmospheric levels of $PM_{2.5}$ among stations ($p<0.05$), with traffic influence presenting the higher levels of $PM_{2.5}$. Regarding traffic and background influences, other studies developed in Portugal demonstrated the significant relation between human activities and traffic and background emissions[62,67].

The decreasing of $PM_{2.5}$ levels of traffic stations from 2003-2010 and from 2011 onwards can be justified with the several policies that increased monitoring action, such as the EU Directive 2001/8 [73] 1 and the Industrial Emissions Directive 2010/75/EU[26], implemented from 2014 onwards.

As referred by Ferreira[27], these Directives were drivers for the implementation of environmental protection policies, pushing industries to implement evaluation plans aiming to modify processes and reduce emissions. For the reduction of emissions of traffic influence, the implementation of Euro 6 in 2015 (aimed to limit the emission of polluting gases into the atmosphere produced by road vehicles, making the manufacture of diesel cars and trucks mandatory, and combining pollutant reduction systems) was a major advance.

The decrease in traffic-related pollutant emissions was already observed in the period 2011-2015, probably related with the rejuvenation of the circulating cars, which has led to a reduction on the emissions, especially in urban and suburban areas since 2016, although still insufficient to comply with the ongoing WHO AQG 2021 (Figure 6).

When considering other EU countries, Bulgaria, Poland, Romania, and Croatia, a similar pattern was registered, with higher atmospheric levels of $PM_{2.5}$ in urban areas[74]. In a global perspective and in line with the results obtained under the present study, the EEA Report 2021 showed that residential, commercial, and institutional energy consumption was the principal source of coarse particulate matter (PM_{10}) and fine particulate matter ($PM_{2.5}$) in 2019[75]. Road transport and industry were also significant sources for both pollutants, although, these concentrations have been decreasing by 29% since 2005.

A detailed description of levels of $PM_{2.5}$ for the period considered (2003-2021) by environmental area and emission influence is presented as Attachments: Table S1, Table S2 e Table S3. A more detailed description of levels of $PM_{2.5}$ by environmental area and emission influence in the eligible stations is presented in sequence of figures: Figure S4, Figure S5, and Figure S6.

Portugal has presented air monitoring registries below the legislation established in the European Union since 2006. However, only in 2016 certain areas presented levels below the former WHO AQG recommendation for $PM_{2.5}$ (10 $\mu g/m^3$), and in 2021 the compliance with the current WHO recommended values for $PM_{2.5}$ (5 $\mu g/m^3$) still encompasses a great challenge. Figure 7 represents the exceedance (%) of the EU AQD 2008 and WHO AQG of 2006 and 2021, for the hourly registers in Portugal for the year 2021, disaggregated by environmental area and emission influence, and considering all monitoring stations (n=30).

Figure 7 showed that atmospheric levels of $PM_{2.5}$ in 2021 exceeded the permitted levels of EU Air Quality Directive 2008 and the recommended levels of WHO AQG 2006 and 2021. Considering all monitoring stations, the exceedance ranged from 0.07% (Terena) to 11.70% (Estarreja), from 0.3% (Cerro) to 37% (Estarreja), and from 6% (Faial) to 72% (Mem Martins) when compared to the EU Air Quality Directive, WHO AQG 2006 and WHO AQG 2021, respectively. The following stations presented an exceedance of WHO AQG 2021 above 70% of registries: Mem-Martins (urban and traffic), Alfragide and Mem-Martins (urban and background), and Monte Velho (rural and background). Results showed also that all stations registered atmospheric levels above 25 $\mu g/m^3$, except for Joaquim Magalhães Station and Cerro Station, urban and rural stations, respectively.

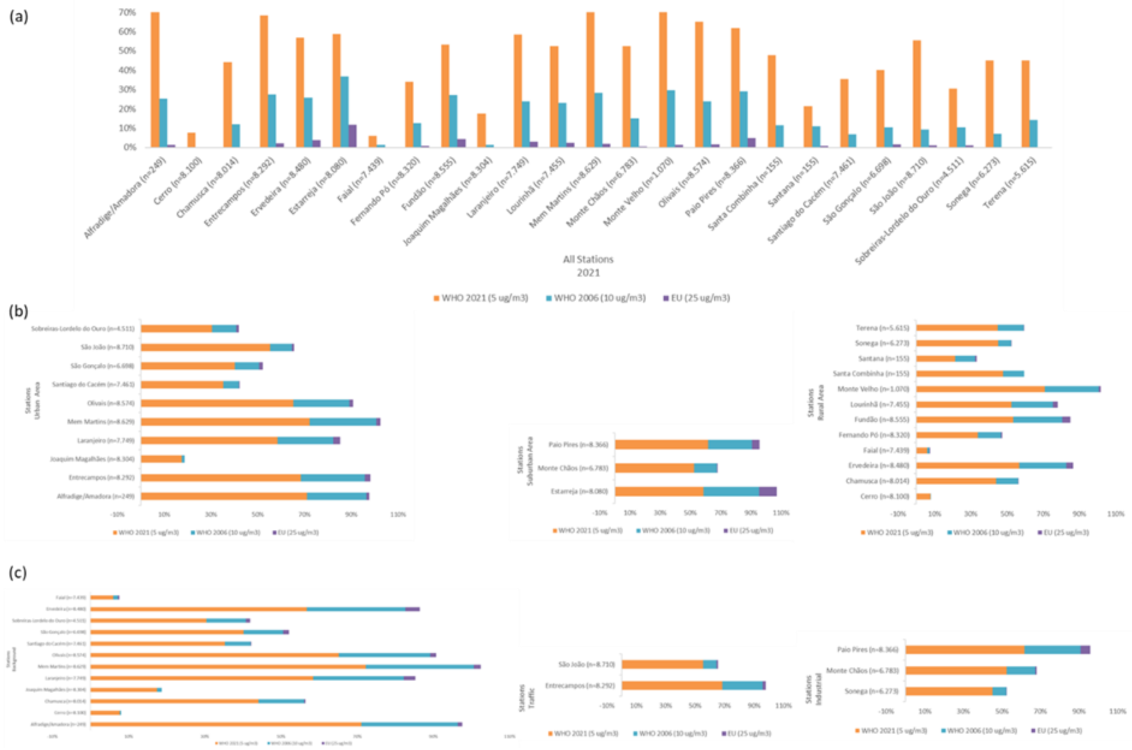

Figure 7. Percentage of hourly exceedance of $PM_{2.5}$ of WHO air quality guidelines (2006 and 2021) and of EU Air Quality Directive (2008) registered by monitoring stations (a), by environmental area (b), and emission influence (c), in 2021, Portugal. According to According to data obtained from QualAr*.

*For this analysis, five stations were not considered due to the absence of registries for the year 2021. For this, four were urban and one was rural by environmental area and, by emission influence, four were background and one was traffic.

Despite the efforts of environmental policies aimed at controlling and encouraging a society with better air quality, Portugal registered at some time points levels above the EU Air Quality Directive, being even more challenging to keep within the recommended limits of WHO Air Quality Guidelines (2006 and 2021). It should be also noted that several episodes of sand dust from North Africa affected the air quality in Portugal during the year 2021. Therefore, some of the exceedances registered might be due to unavoidable climatic events[76].

These results agree well with the ones presented in EEA 2023 report, where it is referred that 94% of the European population is exposed to atmospheric levels of $PM_{2.5}$

above the recommended levels of WHO AQG 2021 and only 1% is exposed to atmospheric levels of $PM_{2.5}$ above the levels of EU AQD 2008[77]. Considering that exposure to $PM_{2.5}$ is linked to adverse health effects, such as respiratory and cardiovascular diseases, and cancer, efforts should be considered to decrease the levels of atmospheric $PM_{2.5}$[71].

The concentrations of $PM_{2.5}$ are a global issue, according to Yu et al.[78], being one of the main pollutants causing several health outcomes. Several studies indicate that long-term exposure to $PM_{2.5}$ is associated with an increase in the long-term risk of cardiopulmonary mortality by 6–13% per 10 $\mu g/m^3$ of $PM_{2.5}$[54,79,80]. The exposure is ubiquitous and involuntary and there is no evidence of a safe level of exposure or a threshold below which no adverse health effects occur, therefore, increasing the significance of this risk factor[20,81].

It was verified reduced levels of $PM_{2.5}$ in 2021 when compared to 2020, especially in suburban areas and industrial emission[82,83]. The present study confirmed this trendin the recent years of 2020-2021.

The reduction of atmospheric pollutants by $PM_{2.5}$ in Portugal is directly related to the achievement of the United Nations' Sustainable Development Goals (SDGs), especially SDG 3 (good health and well-being), SDG 13 (climate action) and SGD 15 (life on land). Considering the above-mentioned causal associations with health outcomes, it is fundamental to develop all efforts to achieve the WHO targets and therefore contribute for the attainment of SDGs at a global level. Portugal, although being a country with a continuous record of decreasing levels of air pollutants, is not exempt from this challenge; the targets recommended by WHO in 2021 are ambitious and require continuous efforts.

In view of this exemption, the implementation is crucial for directives to yield results, and concrete action plans involving society, companies, and governmental entities are essential for effective implementation.

6.4. Strengths and Limitations

Results presented here with showed a detailed analysis of atmospheric levels of $PM_{2.5}$ for Portugal during the last two decades. Although previous assessments were available, the present study included the years 2020-2021 where a different scenario of exposure was experienced due to the COVID-19 pandemic. In addition, the comparison of results before and after the implementation of important EU policies and legislative measures allows us to evaluate their effectiveness, being an important asset for regulators and policymakers. The results of this research will be made available to different stakeholders aiming to support policy decisions regarding air quality in Portugal.

The present study considered only air monitoring data available for $PM_{2.5}$, thus not including data estimated using conversion factors from levels of PM_{10}. Although incurring data loss, this option was preferred to avoid data assumptions, decrease the associated uncertainty, and to allow presenting the real scenario provided by monitoring stations. In daddition, the characteristics of the eligible monitoring stations that were considered for this study were similar to the non-eligible stations, regarding environmental area and emission influence.

7. Conclusions

It has been demonstrated for the period 2003-2021 that Portugal has implemented environmental policies which have been important to reduce atmospheric levels of $PM_{2.5}$. Efforts have been made to guarantee that pollutant levels are below the mean levels established by the applicable EU Directives. However, the comparison of the registered atmospheric levels of $PM_{2.5}$ with the WHO AQG 2021 recommended values highlights the challenge ahead and the need for further and continuous measures.

This study highlighted some improvement opportunities for the atmospheric levels of $PM_{2.5}$ in the light of environmental policies and legislative measures implemented in Portugal along these years. Also, a comparison with the new WHO AQG 2021 is provided, thus highlighting the need for further environmental policies to comply with the recommended values.

In this study, it was considered for the first time, the period 2003-2021, including a detailed analysis of the period 2020-2021 to assess the possible influence of COVID-19 related lockdowns. Additionally, the study analysed the fluctuations in $PM_{2.5}$ levels across various regions, considering environmental factors and emission sources.

Portugal has a proven good record of decreasing air pollution. Some measures are important to keep up in this path: implement cleaner and more efficient technologies in sectors such as transportation and industry; support electric vehicles and efficient public transportation systems and create adequate infrastructures to support these alternatives. Additionally, continuous monitoring of air quality is fundamental to assess human and environmental exposure to atmospheric pollutants and to evaluate the effectiveness of the policy measures implemented. The integrated and continuous efforts of all the stakeholders (e.g., academia, companies, policy makers) are of utmost importance for the persecution of improving human health and well-being through the decrease of the air pollution impact, as a relevant environmental determinant of health.

7.1. Recommendations for future research studies and public health intervention

- This study was conceived as a first relevant step towards a comprehensive assessment that provides a complete understanding of the air quality situation in Portugal. Thus, it reflects on the possible risks to health due exceedance levels from $PM_{2.5}$, and provides opportunities to relate negative outcomes with the appropriate dose response to assess disease burden and associated costs.
- This study design can be applied to the other atmospheric pollutants, thus contributing to have a broader perspective for Portugal.
- This study design can support to analysis of different periods, being able to evaluate situations of exceeding the limits allowed by the regulations and WHO recommendations. These data may be correlated with situations of emergency care, hospitalizations, deaths due to climatic events, due to sandstorms, floods or other natural disasters that are occurring more and more frequently due to climate change.

8. References

1. Toscano D, Murena F, Costabile F, Hussein T, Massimi L. The historical trend of Air Pollution and its impact on Human Health in Campania Region (Italy). Atmosphere 2021, Vol 12, Page 553 [Internet]. 2021 Apr 25 [cited 2022 Nov 5];12(5):553. Available from: https://www.mdpi.com/2073-4433/12/5/553/htm

2. Niu Z, Liu F, Yu H, Wu S, Xiang H. Association between exposure to ambient air pollution and hospital admission, incidence, and mortality of stroke: an updated systematic review and meta-analysis of more than 23 million participants. Environ Health Prev Med [Internet]. 2021 Dec 1 [cited 2023 May 17];26(1). Available from: https://pubmed.ncbi.nlm.nih.gov/33499804/

3. Tian M, Zhao J, Mi X, Wang K, Kong D, Mao H, et al. Progress in research on effect of PM2.5 on occurrence and development of atherosclerosis. J Appl Toxicol [Internet]. 2021 May 1 [cited 2023 May 17];41(5):668–82. Available from: https://pubmed.ncbi.nlm.nih.gov/33263192/

4. Gorr M, Aslaner D, Saldaña T, Mackenzie D, Falvo M, Wold L. Short-term PM exposure and social defeat cause reduction in pulmonary and right ventricle function. The FASEB Journal [Internet]. 2021 May 14 [cited 2023 May 17];35(S1):fasebj.2021.35.S1.05207. Available from: https://onlinelibrary.wiley.com/doi/10.1096/fasebj.2021.35.S1.05207

5. Liu H, Tian Y, Xu Y, Huang Z, Huang C, Hu Y, et al. Association between ambient air pollution and hospitalization for ischemic and hemorrhagic stroke in China: A multicity case-crossover study. Environ Pollut [Internet]. 2017 [cited 2023 May 17];230:234–41. Available from: https://pubmed.ncbi.nlm.nih.gov/28654881/

6. Wang M, Zhou T, Song Y, Li X, Ma H, Hu Y, et al. Joint exposure to various ambient air pollutants and incident heart failure: a prospective analysis in UK Biobank. Eur Heart J [Internet]. 2021 Apr 21 [cited 2023 May 17];42(16):1582–91. Available from: https://pubmed.ncbi.nlm.nih.gov/33527989/

7. Brauer M, Casadei B, Harrington RA, Kovacs R, Sliwa K. Taking a Stand Against Air Pollution—The Impact on Cardiovascular Disease. Circulation [Internet]. 2021 Apr 6;143(14):E800–4. Available from: https://www.ahajournals.org/doi/10.1161/CIRCULATIONAHA.120.052666

8. EEA. Briefing no. 19/2021: Health impacts of air pollution in Europe, 2021 — European Environment Agency. 2021 [cited 2023 May 17]; Available from: https://www.eea.europa.eu/publications/health-risks-of-air-pollution/health-impacts-of-air-pollution

9. Brito J, Bernardo A, Gonçalves LL. Atmospheric pollution and mortality in Portugal: Quantitative assessment of the environmental burden of disease using the AirQ+ model. Science of The Total Environment [Internet]. 2022 Apr 1 [cited 2023 May 17];815:152964. Available from: https://linkinghub.elsevier.com/retrieve/pii/S0048969722000535

10. Amoushahi S, Bayat R, Sanaei A, Szyszkowicz M, Faridi S, Hassanvand MS. Health and economic impacts of ambient fine particulate matter in Isfahan, Iran. Urban Clim [Internet]. 2022 Jan 1 [cited 2023 May 17];41:101048. Available from: https://linkinghub.elsevier.com/retrieve/pii/S2212095521002789

11. Zhao X, Zhou W, Wu T, Han L. The impacts of urban structure on PM2.5 pollution depend on city size and location. Environmental Pollution. 2022 Jan 1;292:118302.

12. Commission proposes rules for cleaner air and water [Internet]. [cited 2023 May 26]. Available from: https://ec.europa.eu/commission/presscorner/detail/en/ip_22_6278

13. Kelly FJ, Fussell JC. Air pollution and public health: emerging hazards and improved understanding of risk. Environ Geochem Health [Internet]. 2015 Aug 29 [cited 2023 May 17];37(4):631–49. Available from: https://pubmed.ncbi.nlm.nih.gov/26040976/

14. Sharma S, Chandra M, Kota SH. Health Effects Associated with PM2.5: a Systematic Review. Current Pollution Reports 2020 6:4 [Internet]. 2020 Jul 17 [cited 2023 May 17];6(4):345–67. Available from: https://link.springer.com/article/10.1007/s40726-020-00155-3

15. $PM_{2.5}$ levels, air pollution, and our health – Howard J. Luks, MD [Internet]. [cited 2023 May 25]. Available from: https://www.howardluksmd.com/pm-2-5-levels-air-pollution-and-our-health/

16. Wyatt LH, Devlin RB, Rappold AG, Case MW, Diaz-Sanchez D. Low levels of fine particulate matter increase vascular damage and reduce pulmonary function in young healthy adults. Part Fibre Toxicol [Internet]. 2020 Dec 1 [cited 2023 May 25];17(1). Available from: https://www.howardluksmd.com/pm-2-5-levels-air-pollution-and-our-health/

17. Health impacts of air pollution in Europe, 2021 — European Environment Agency [Internet]. [cited 2023 May 26]. Available from: https://www.eea.europa.eu/publications/air-quality-in-europe-2021/health-impacts-of-air-pollution

18. WHO global air quality guidelines: particulate matter ($PM_{2.5}$ and PM_{10}), ozone, nitrogen dioxide, sulfur dioxide and carbon monoxide [Internet]. [cited 2023 May 17]. Available from: https://apps.who.int/iris/handle/10665/345329

19. Directive 2008/50/EC, air quality — European Environment Agency [Internet]. [cited 2023 May 26]. Available from: https://www.eea.europa.eu/policy-documents/directive-2008-50-ec-of

20. New WHO Global Air Quality Guidelines aim to save millions of lives from air pollution [Internet]. [cited 2023 May 17]. Available from: https://www.who.int/news/item/22-09-2021-new-who-global-air-quality-guidelines-aim-to-save-millions-of-lives-from-air-pollution

21. Abbafati C, Abbas KM, Abbasi-Kangevari M, Abd-Allah F, Abdelalim A, Abdollahi M, et al. Global burden of 87 risk factors in 204 countries and territories, 1990–2019: a systematic analysis for the Global Burden of Disease Study 2019. The Lancet [Internet]. 2020 Oct 17 [cited 2023 May 26];396(10258):1223–49. Available from: http://www.thelancet.com/article/S0140673620307522/fulltext

22. Qualidade do Ar | ONU News [Internet]. [cited 2023 May 17]. Available from: https://news.un.org/pt/tags/qualidade-do-ar

23. Objetivos de Desenvolvimento Sustentável - ONU Portugal [Internet]. [cited 2023 May 26]. Available from: https://unric.org/pt/objetivos-de-desenvolvimento-sustentavel/

24. Jaiswal KK, Chowdhury CR, Yadav D, Verma R, Dutta S, Jaiswal KS, et al. Renewable and sustainable clean energy development and impact on social, economic, and environmental health. Energy Nexus. 2022 Sep 1;7:100118.

25. Jing QL, Liu HZ, Yu WQ, He X. The Impact of Public Transportation on Carbon Emissions—From the Perspective of Energy Consumption. Sustainability 2022, Vol 14, Page 6248 [Internet]. 2022 May 20 [cited 2023 May 26];14(10):6248. Available from: https://www.mdpi.com/2071-1050/14/10/6248/htm

26. Industrial Emissions Directive [Internet]. [cited 2023 May 17]. Available from: https://environment.ec.europa.eu/topics/industrial-emissions-and-safety/industrial-emissions-directive_en

27. Santos MA, Ferreira N. Implementação da Diretiva 2010/75/EU-Emissões Industriais Estudo de Caso: Refinaria de Sines Mestrado Integrado em Engenharia da Energia e do Ambiente. 2015;

28. Directorate-General For External Policies, Policy Department. Europe's approach to implementing the Sustainable Development Goals: good practices and the way forward [Internet]. 2019. [cited 2023 May 27]. Available from: https://www.europarl.europa.eu/cmsdata/160360/DEVE%20study%20on%20EU%20SDG%20implementation%20formatted.pdf

29. EEA. Briefing no. 19/2021: Health impacts of air pollution in Europe, 2021 — European Environment Agency. 2021 [cited 2023 May 28]; Available from: https://www.eea.europa.eu/publications/health-risks-of-air-pollution/health-impacts-of-air-pollution

30. Ambient (outdoor) air pollution [Internet]. [cited 2023 May 27]. Available from: https://www.who.int/news-room/fact-sheets/detail/ambient-(outdoor)-air-quality-and-health

31. Sunyer J, Esnaola M, Alvarez-Pedrerol M, Forns J, Rivas I, López-Vicente M, et al. Association between Traffic-Related Air Pollution in Schools and Cognitive Development in Primary School Children: A Prospective Cohort Study. PLoS Med [Internet]. 2015 Mar 1 [cited 2023 May 25];12(3):e1001792. Available from: https://journals.plos.org/plosmedicine/article?id=10.1371/journal.pmed.1001792

32. Weuve J, Kaufman JD, Szpiro AA, Curl C, Puett RC, Beck T, et al. Exposure to Traffic-Related Air Pollution in Relation to Progression in Physical Disability among Older Adults. Environ Health Perspect [Internet]. 2016 Jul 1 [cited 2023 May 25];124(7):1000–8. Available from: https://ehp.niehs.nih.gov/doi/10.1289/ehp.1510089

33. Varieur BM, Fisher S, Landrigan PJ. Air Pollution, Political Corruption, and Cardiovascular Disease in the Former Soviet Republics. Ann Glob Health [Internet]. 2022 [cited 2023 May 17];88(1). Available from: https://pubmed.ncbi.nlm.nih.gov/35854924/

34. Health effects of particulate matter: policy implications for countries in eastern Europe, Caucasus and central Asia [Internet]. [cited 2023 May 27]. Available from: https://apps.who.int/iris/handle/10665/344854

35. Lelieveld J, Evans JS, Fnais M, Giannadaki D, Pozzer A. The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature [Internet]. 2015 Sep 16 [cited 2023 May 17];525(7569):367–71. Available from: https://www.nature.com/articles/nature15371

36. Li T, Yu Y, Sun Z, Duan J. A comprehensive understanding of ambient particulate matter and its components on the adverse health effects based from epidemiological and laboratory evidence. Part Fibre Toxicol [Internet]. 2022 Nov 29 [cited 2023 May 17];19(1):67. Available from: https://particleandfibretoxicology.biomedcentral.com/articles/10.1186/s12989-022-00507-5

37. Feng S, Gao D, Liao F, Zhou F, Wang X. The health effects of ambient PM2.5 and potential mechanisms. Ecotoxicol Environ Saf. 2016 Jun 1;128:67–74.

38. Basith S, Manavalan B, Shin TH, Park CB, Lee WS, Kim J, et al. The Impact of Fine Particulate Matter 2.5 on the Cardiovascular System: A Review of the Invisible Killer. Nanomaterials 2022, Vol 12, Page 2656 [Internet]. 2022 Aug 2 [cited 2023 May 27];12(15):2656. Available from: https://www.mdpi.com/2079-4991/12/15/2656/htm

39. Xing YF, Xu YH, Shi MH, Lian YX. The impact of PM2.5 on the human respiratory system. J Thorac Dis [Internet]. 2016 [cited 2023 May 17];8(1):E69. Available from: /pmc/articles/PMC4740125/

40. Yang L, Li C, Tang X. The Impact of PM2.5 on the Host Defense of Respiratory System. Front Cell Dev Biol. 2020 Mar 4;8.

41. Bevan GH, Al-Kindi SG, Brook R, Rajagopalan S. Ambient Air Pollution and Atherosclerosis: Recent Updates. Curr Atheroscler Rep [Internet]. 2021 Oct 21 [cited 2023 May 17];23(10):63. Available from: https://link.springer.com/10.1007/s11883-021-00958-9

42. Münzel T, Sørensen M, Gori T, Schmidt FP, Rao X, Brook J, et al. Environmental stressors and cardio-metabolic disease: part I–epidemiologic evidence supporting a role for noise and air pollution and effects of mitigation strategies. Eur Heart J [Internet]. 2016 Jul 26 [cited 2023 May 17];550–6. Available from: http://eurheartj.oxfordjournals.org/lookup/doi/10.1093/eurheartj/ehw269

43. Thurston GD, Kipen H, Annesi-Maesano I, Balmes J, Brook RD, Cromar K, et al. A joint ERS/ATS policy statement: What constitutes an adverse health effect of air pollution? An analytical framework. European Respiratory Journal. 2017 Jan 1;49(1).

44. Zhang Q, Meng X, Shi S, Kan L, Chen R, Kan H. Overview of particulate air pollution and human health in China: Evidence, challenges, and opportunities. The Innovation [Internet]. 2022 Nov 11 [cited 2023 May 25];3(6):100312. Available from: /pmc/articles/PMC9490194/

45. Cai S, Ma Q, Wang S, Zhao B, Brauer M, Cohen A, et al. Impact of air pollution control policies on future PM2.5 concentrations and their source contributions in China. J Environ Manage [Internet]. 2018 Dec 1 [cited 2023 May 17];227:124–33. Available from: https://pubmed.ncbi.nlm.nih.gov/30172931/

46. Park J, Kim HJ, Lee CH, Lee CH, Lee HW. Impact of long-term exposure to ambient air pollution on the incidence of chronic obstructive pulmonary disease: A systematic review and meta-analysis. Environ Res [Internet]. 2021 Mar 1 [cited 2023 May 17];194:110703. Available from: https://linkinghub.elsevier.com/retrieve/pii/S0013935120316029

47. Cohen AJ, Brauer M, Burnett R, Anderson HR, Frostad J, Estep K, et al. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015. Lancet [Internet]. 2017 May 5 [cited 2023 May 27];389(10082):1907. Available from: /pmc/articles/PMC5439030/

48. Lim SS, Vos T, Flaxman AD, Danaei G, Shibuya K, Adair-Rohani H, et al. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. The Lancet [Internet]. 2012 Dec 15 [cited 2023 May 17];380(9859):2224–60. Available from: http://www.thelancet.com/article/S0140673612617668/fulltext

49. European Environment Agency. Sources and emissions of air pollutants in Europe. Air quality in Europe 2021 [Internet]. 2019 [cited 2023 May 17];1–15. Available from: https://www.eea.europa.eu/publications/air-quality-in-europe-2021/air-quality-in-europe-2021

50. Heydari S, Asgharian M, Kelly FJ, Goel R. Potential health benefits of eliminating traffic emissions in urban areas. PLoS One [Internet]. 2022 Mar 1 [cited 2023 May 27];17(3):e0264803. Available from: https://journals.plos.org/plosone/article?id=10.1371/journal.pone.0264803

51. Yusuf S, Joseph P, Rangarajan S, Islam S, Mente A, Hystad P, et al. Modifiable risk factors, cardiovascular disease, and mortality in 155 722 individuals from 21 high-income, middle-income, and low-income countries (PURE): a prospective cohort study. The Lancet [Internet]. 2020 Mar 7 [cited 2023 May 17];395(10226):795–808. Available from: https://linkinghub.elsevier.com/retrieve/pii/S0140673619320082

52. Di Q, Wang Y, Zanobetti A, Wang Y, Koutrakis P, Choirat C, et al. Air Pollution and Mortality in the Medicare Population. New England Journal of

Medicine [Internet]. 2017 Jun 29 [cited 2023 May 17];376(26):2513–22. Available from: http://www.nejm.org/doi/10.1056/NEJMoa1702747

53. Liu Q, Huang K, Liang F, Yang X, Li J, Chen J, et al. Long-term exposure to fine particulate matter modifies the association between physical activity and hypertension incidence. J Sport Health Sci. 2022 Nov 1;11(6):708–15.

54. Alexeeff SE, Liao NS, Liu X, Van Den Eeden SK, Sidney S. Long-Term PM2.5 Exposure and Risks of Ischemic Heart Disease and Stroke Events: Review and Meta-Analysis. J Am Heart Assoc [Internet]. 2021 Jan 5 [cited 2023 May 27];10(1):1–22. Available from: https://www.ahajournals.org/doi/abs/10.1161/JAHA.120.016890

55. Schraufnagel DE, Balmes JR, Cowl CT, De Matteis S, Jung SH, Mortimer K, et al. Air Pollution and Noncommunicable Diseases: A Review by the Forum of International Respiratory Societies' Environmental Committee, Part 2: Air Pollution and Organ Systems. Chest [Internet]. 2019 Feb 1 [cited 2023 May 17];155(2):417–26. Available from: https://pubmed.ncbi.nlm.nih.gov/30419237/

56. Burnett R, Chen H, Szyszkowicz M, Fann N, Hubbell B, Pope CA, et al. Global estimates of mortality associated with longterm exposure to outdoor fine particulate matter. Proc Natl Acad Sci U S A [Internet]. 2018 Sep 18 [cited 2023 May 17];115(38):9592–7. Available from: https://www.pnas.org/doi/abs/10.1073/pnas.1803222115

57. Gimbrone MA, García-Cardeña G. Endothelial Cell Dysfunction and the Pathobiology of Atherosclerosis. Circ Res [Internet]. 2016 Feb 19 [cited 2023 May 17];118(4):620–36. Available from: https://www.ahajournals.org/doi/abs/10.1161/CIRCRESAHA.115.306301

58. Kobza J, Geremek M, Dul L. Characteristics of air quality and sources affecting high levels of PM10 and PM2.5 in Poland, Upper Silesia urban area. Environ Monit Assess [Internet]. 2018 Sep 1 [cited 2023 May 17];190(9):1–13. Available from: https://link.springer.com/article/10.1007/s10661-018-6797-x

59. European Environmental Agency (EEA). Air quality in Europe 2022. Report no. 05/2022. 2022;

60. Portal do INE [Internet]. [cited 2023 May 27]. Available from: https://www.ine.pt/xportal/xmain?xpid=INE&xpgid=ine_faqs&FAQSfaq_boui=63614948&FAQSmodo=1&xlang=pt

61. The Paris Agreement | UNFCCC [Internet]. [cited 2023 May 17]. Available from: https://unfccc.int/process-and-meetings/the-paris-agreement

62. Torres P, Ferreira J, Monteiro A, Costa S, Pereira MC, Madureira J, et al. Air pollution: A public health approach for Portugal. Science of The Total Environment. 2018 Dec 1;643:1041–53.

63. The European Green Deal — European Environment Agency [Internet]. [cited 2023 May 17]. Available from: https://www.eea.europa.eu/policy-documents/com-2019-640-final

64. Zero pollution action plan [Internet]. [cited 2023 May 28]. Available from: https://environment.ec.europa.eu/strategy/zero-pollution-action-plan_en

65. Portugal seeks to become major exporter of green hydrogen | Reuters [Internet]. [cited 2023 May 17]. Available from: https://www.reuters.com/business/energy/portugal-seeks-become-major-exporter-green-hydrogen-2022-11-10/

66. Portugal Climate Change Data | Emissions and Policies | Climate Watch [Internet]. [cited 2023 May 27]. Available from: https://www.climatewatchdata.org/countries/PRT?end_year=2019&start_year=1990

67. Gama C, Monteiro A, Pio C, Miranda AI, Baldasano JM, Tchepel O. Temporal patterns and trends of particulate matter over Portugal: a long-term analysis of background concentrations. Air Qual Atmos Health [Internet]. 2018 May 1 [cited 2023 May 17];11(4):397–407. Available from: https://link.springer.com/article/10.1007/s11869-018-0546-8

68. Reche C, Tobias A, Viana M. Vehicular Traffic in Urban Areas: Health Burden and Influence of Sustainable Urban Planning and Mobility. Atmosphere (Basel) [Internet]. 2022 Apr 1 [cited 2023 May 27];13(4):598. Available from: https://www.mdpi.com/2073-4433/13/4/598/htm

69. Giugliano M, Lonati G, Butelli P, Romele L, Tardivo R, Grosso M. Fine particulate (PM2.5–PM1) at urban sites with different traffic exposure. Atmos Environ. 2005 Apr 1;39(13):2421–31.

70. Błaszczak B, Widziewicz-Rzońca K, Ziola N, Klejnowski K, Juda-Rezler K. Chemical Characteristics of Fine Particulate Matter in Poland in Relation with Data from Selected Rural and Urban Background Stations in Europe. Applied Sciences 2019, Vol 9, Page 98 [Internet]. 2018 Dec 28 [cited 2023 May 27];9(1):98. Available from: https://www.mdpi.com/2076-3417/9/1/98/htm

71. Ródenas M, Soler R, Borrás E, Vera T, Diéguez JJ, Muñoz A. Assessment of COVID-19 Lockdown Impact on the Air Quality in Eastern Spain: PM and BTX in Urban, Suburban and Rural Sites Exposed to Different Emissions. Atmosphere (Basel) [Internet]. 2022 Jan 1 [cited 2023 May 17];13(1):97. Available from: https://www.mdpi.com/2073-4433/13/1/97/htm

72. Aghababaeian H, Ostadtaghizadeh A, Ardalan A, Asgary A, Akbary M, Yekaninejad MS, et al. Global Health Impacts of Dust Storms: A Systematic Review. Environ Health Insights [Internet]. 2021 [cited 2023 May 27];15. Available from: https://pubmed.ncbi.nlm.nih.gov/34103932/

73. Directive 2001/81/EC of the European Parliament and of the Council of 23 October 2001 on national emission ceilings for certain atmospheric pollutants [Internet]. 2011 [cited 2023 May 28]. Available from: https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex%3A32001L0081

74. How polluted is the air in urban areas? - Products Eurostat News - Eurostat [Internet]. [cited 2023 May 17]. Available from: https://ec.europa.eu/eurostat/web/products-eurostat-news/-/edn-20210603-1

75. Sources and emissions of air pollutants in Europe — European Environment Agency [Internet]. [cited 2023 May 28]. Available from: https://www.eea.europa.eu/publications/air-quality-in-europe-2021/sources-and-emissions-of-air

76. IPMA - Detalhe noticia [Internet]. [cited 2023 May 17]. Available from: https://www.ipma.pt/pt/media/noticias/news.detail.jsp?f=/pt/media/noticias/arquivo/2020/poeiras_saara_21062020

77. ETC HE Report 2023/2: Status report of air quality in Europe for year 2022, using validated and up-to-date data — Eionet Portal [Internet]. [cited 2023 May 28]. Available from: https://www.eionet.europa.eu/etcs/etc-he/products/etc-he-products/etc-he-reports/etc-he-report-2023-2-status-report-of-air-quality-in-europe-for-year-2022-using-validated-and-up-to-date-data

78. Yu W, Ye T, Zhang Y, Xu R, Lei Y, Chen Z, et al. Global estimates of daily ambient fine particulate matter concentrations and unequal spatiotemporal distribution of population exposure: a machine learning modelling study. Lancet Planet Health [Internet]. 2023 Mar 1 [cited 2023 May 17];7(3):e209–18. Available from: http://www.thelancet.com/article/S2542519623000086/fulltext

79. ETC HE Report 2022/11: Estimating the morbidity related environmental burden of disease due to exposure to PM2.5, NO2 and O3 in outdoor ambient air — Eionet Portal [Internet]. [cited 2023 May 27]. Available from: https://www.eionet.europa.eu/etcs/etc-he/products/etc-he-products/etc-he-reports/etc-he-report-2022-11-estimating-the-morbidity-related-environmental-burden-of-disease-due-to-exposure-to-pm2-5-no2-and-o3-in-outdoor-ambient-air

80. Wolf K, Hoffmann B, Andersen ZJ, Atkinson RW, Bauwelinck M, Bellander T, et al. Long-term exposure to low-level ambient air pollution and incidence of stroke and coronary heart disease: a pooled analysis of six European cohorts within the ELAPSE project. Lancet Planet Health [Internet]. 2021 Sep 1 [cited 2023 May 17];5(9):e620–32. Available from: https://pubmed.ncbi.nlm.nih.gov/34508683/

81. Health risks of air pollution in Europe – HRAPIE project. Recommendations for concentration–response functions for cost–benefit analysis of particulate matter, ozone and nitrogen dioxide [Internet]. [cited 2023 May 17]. Available from: https://apps.who.int/iris/handle/10665/153692

82. Akan AP, Coccia M. Changes of Air Pollution between Countries Because of Lockdowns to Face COVID-19 Pandemic. Applied Sciences (Switzerland) [Internet]. 2022 Dec 1 [cited 2023 May 17];12(24):12806. Available from: https://www.mdpi.com/2076-3417/12/24/12806/htm

83. Wijnands JS, Nice KA, Seneviratne S, Thompson J, Stevenson M. The impact of the COVID-19 pandemic on air pollution: A global assessment using machine learning techniques. Atmos Pollut Res. 2022 Jun 1;13(6):101438.

9. Appendix

9.1 Submitted article: Heliyon Q1 (Impact factor, 4.0)

Heliyon

Concentrations of PM2.5 in Portugal from 2003 until 2021 - Detailed analysis for policy uptake

--Manuscript Draft--

Manuscript Number:	HELIYON-D-23-32174
Article Type:	Original Research Article
Section/Category:	Physical and Applied Sciences
Keywords:	Outdoor air pollution; Particulate matter; Air Quality; Portugal
Manuscript Classifications:	90.100: Atmospheric Science; 90.100.130: Air Quality; 90.150: Environmental Assessment; 90.210: Environmental Health; 90.240: Environmental Pollution; 130.100: Public Health
Corresponding Author:	Carla Martins New University of Lisbon National School of Public Health PORTUGAL
First Author:	Lorena Lima
Order of Authors:	Lorena Lima
	Susana Viegas
	Carla Martins
Abstract:	Air pollution is the main environmental cause of mortality worldwide. Particulate matter < 2.5 µm (PM2.5) has adverse health effects, especially on cardiovascular morbidity and mortality. This study analyzed levels of PM2.5 in Portugal from 2003-2021 across regions, emission influences, and environmental areas, comparing results with the European Union Air Quality Directive 2008 (EU AQD 2008) and the World Health Organization Air Quality Guidelines of 2006 and 2021 (WHO AQG 2006/2021). Data available in the platform QualAR Portugal were obtained and for each monitoring station, on region, city, station, emission influence (traffic, background, industrial), and environment (urban, suburban, rural) were collected. The percentage of hourly exceedances of EU AQD (25 µg/m3) and WHO AQG of 2006 and 2021 (10 µg/m3 and 5 µg/m3, respectively) was determined. From 2003 to 2021, a reduction of the annual mean levels of PM2.5 was observed, with differences between Lisbon Metropolitan Area, and the other regions ($p<0.05$). There were significant differences at national level for the years 2015-2016 and 2019–2021, and for the months of lockdown in 2020 and 2021 ($p \leq 0.05$). Regarding environmental area and emission influence, urban and suburban areas and traffic and background influence, respectively, showed higher statistically significant levels of PM2.5 than the remaining stations ($p<0.05$). The atmospheric levels of PM2.5 in 2021 exceeded the permitted levels of EU AQD 2008 and the recommended levels of WHO AQG 2006 and 2021, in percentages ranging from 0.07% to 11.70%, from 0.3% to 37%, and from 6% to 72%, respectively. The

10. Attachments

Table S1. Atmospheric levels of $PM_{2.5}$ (µg/m^3) (mean, median, maximum, and percentiles 25 and 75) for urban monitoring stations by emission influence, per year, for Portugal.

	Urban																	
	Industrial						Traffic						Background					
Year	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean
2004							5,00	11,00	1,00	2,00	4,00	2,82						
2005							0,00	229,00	6,00	18,00	38,00	27,15						
2006							0,00	126,00	15,00	19,00	25,00	21,89	0,00	196,33	4,33	9,00	15,67	12,87
2007							0,00	319,00	5,00	14,00	28,00	21,30	0,00	131,00	6,50	8,50	12,00	9,80
2008							0,00	176,50	5,00	6,50	11,50	10,25	0,00	70,50	4,50	6,50	9,00	7,04
2009							0,00	129,00	5,50	7,00	12,00	10,63	0,50	28,50	4,00	5,50	7,00	6,03
2010							0,00	95,67	5,00	7,33	12,67	10,22	0,00	60,50	4,50	6,00	8,00	6,65
2011							0,00	129,00	1,00	1,00	5,00	5,49	0,00	40,00	3,00	5,00	9,00	6,84
2012							0,00	96,00	1,00	1,00	4,00	5,44	0,00	49,00	2,00	4,00	8,00	6,09
2013							4,00	20,00	1,00	1,00	5,00	4,78						
2014							10,50	20,50	3,00	5,00	9,00	7,11	33,63	46,10	5,10	8,33	13,50	10,50
2015							1,00	97,00	8,00	12,50	19,40	14,95	0,73	89,68	5,98	10,05	16,20	12,26
2016							0,20	76,13	5,87	8,57	12,37	9,84	0,55	65,05	3,80	6,95	11,25	8,61
2017							0,00	70,00	6,00	8,00	11,00	8,79	8,80	44,60	4,78	8,48	13,75	10,44
2018	0,00	47,00	5,00	8,00	12,00	8,82	0,35	57,95	5,80	8,80	12,75	10,05	0,10	63,10	4,93	8,70	13,97	10,64
2019	0,00	34,00	3,00	5,50	8,50	6,32	0,60	81,30	6,20	9,60	14,20	11,66	0,63	75,15	2,68	5,35	10,18	7,99
2020	0,00	30,00	3,00	4,00	7,00	4,88	0,30	29,30	6,10	9,00	12,85	10,19	0,98	70,85	3,63	6,30	10,40	7,86
2021	0,00	4,80	2,00	4,00	6,00	4,80	4,50	7,35	4,20	6,13	8,75	7,35	0,88	6,48	3,16	4,90	8,12	6,48

Table S2. Atmospheric levels of $PM_{2.5}$ (µg/m^3) (mean, median, maximum, and percentiles 25 and 75) for suburban monitoring stations by emission influence, per year, for Portugal.

	Suburban																	
	Industrial						Traffic						Background					
Year	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean
2005													0,00	258,00	9,00	19,00	34,00	26,39
2006													0,00	163,00	9,00	17,00	31,00	24,67
2007													0,00	149,00	8,00	18,00	31,00	23,87
2008													0,00	146,00	5,00	11,00	21,00	16,14
2009													0,00	111,00	5,00	11,00	20,00	14,81
2011													0,00	127,00	5,00	11,00	21,00	16,59
2012													0,00	158,00	5,00	11,00	22,00	17,13
2013													22,00	33,00	5,00	10,00	17,00	14,01
2014	7,00	7,00	3,00	6,00	11,00	10,89							32,00	105,00	3,00	8,00	16,00	12,49
2015													0,00	151,00	5,00	10,00	17,00	14,75
2018	0,00	34,00	4,00	7,00	10,00	7,65												
2019	0,00	32,00	3,00	6,00	9,00	6,48							0,00	85,00	2,00	5,00	11,00	8,99
2020	0,10	73,70	3,40	6,15	10,35	7,69							0,00	106,00	3,00	6,00	11,00	6,76
2021	0,10	8,00	3,80	6,30	9,55	8,00							0,00	11,80	3,00	7,00	15,00	11,80

Table S3. Atmospheric levels of $PM_{2.5}$ (µg/m^3) (mean, median, maximum, and percentiles 25 and 75) for rural monitoring stations by emission influence, per year, for Portugal.

	Rural																	
	Industrial						Traffic						Background					
Year	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean	Minimum	Maximum	1°qt_25%	Median	3°qt_75%	Mean
2003													0,00	57,00	4,00	8,00	13,00	9,96
2005													0,33	317,67	8,67	15,33	25,00	19,55
2006													0,00	147,75	5,00	8,75	14,75	11,62
2007													0,20	68,60	5,40	8,80	13,60	10,44
2008													0,00	48,80	3,40	5,80	9,60	7,28
2009													0,00	194,67	4,33	7,33	12,00	8,97
2010													0,00	120,67	3,33	8,00	14,33	10,52
2011													0,00	105,20	3,40	7,00	12,60	9,45
2012													0,00	93,20	3,00	6,40	10,80	8,19
2013													4,50	10,00	2,50	4,75	7,75	6,71
2014													4,89	14,29	2,36	5,14	9,76	5,87
2015							0,00	44,00	3,00	4,00	6,00	4,72	0,00	97,17	3,67	7,05	12,33	9,05
2016							0,00	60,00	2,00	3,00	5,00	4,16	0,17	95,77	2,63	5,35	9,15	6,64
2017	0,00	60,00	2,00	3,00	5,00	4,16							4,14	65,54	6,83	6,92	12,00	7,98
2018	0,00	44,00	2,00	3,00	5,00	3,81							0,00	49,97	2,40	4,93	8,10	5,93
2019	0,00	14,00	1,00	2,00	4,00	2,69							0,00	45,35	2,77	4,67	7,28	5,79
2020	0,00	22,00	2,00	4,00	6,00	4,71							0,37	39,72	2,30	4,10	7,47	5,76
2021													0,14	7,19	2,51	4,05	6,98	5,55

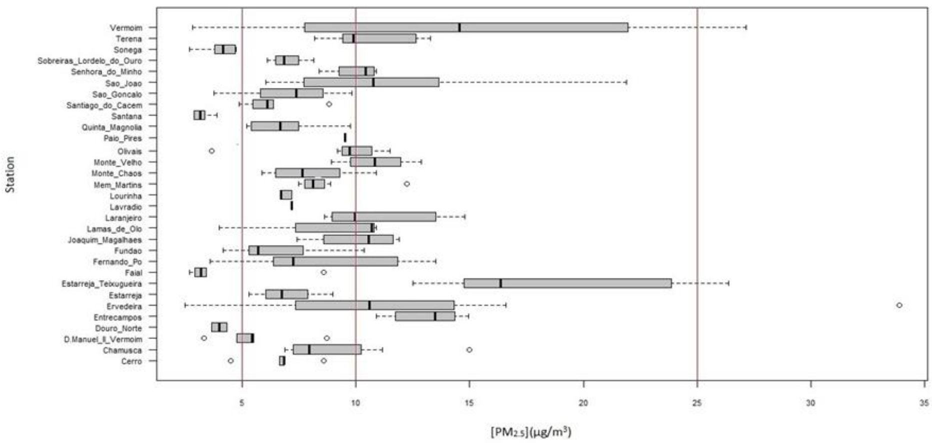

Figure S1. Concentrations of particulate matter ($PM_{2.5}$) (mean) in Portugal, in the period 2003–2021. Data represents average values collected from stations of the Air Quality Network of the Portuguese Environment Agency (QualAr). Limits of EU Air Quality Directive (25 µg/m^3), WHO Air Quality Guidelines 2006 (10 µg/m^3) and WHO Air Quality Guidelines 2021 (5 µg/m^3) for $PM_{2.5}$ are included as red lines.

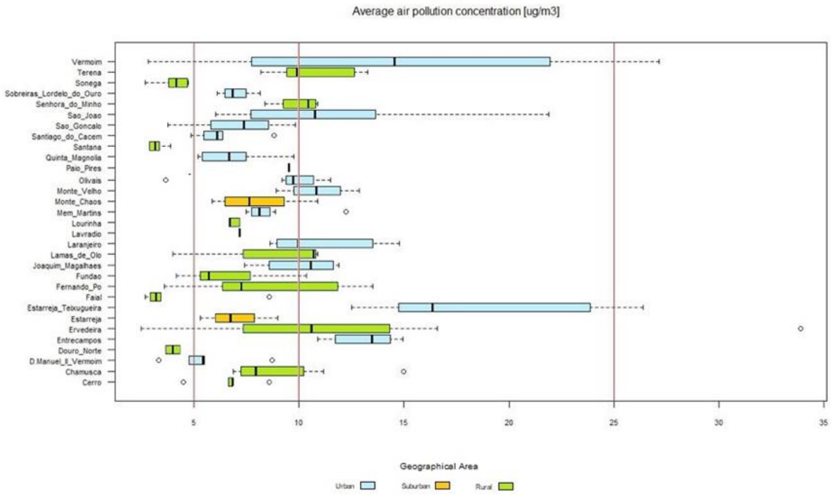

Figure S2. Concentrations of particulate matter ($PM_{2.5}$) (mean), by geographical area (urban, suburban and rural) and stations, in Portugal, in the period 2003–2021. Data represents average values collected from stations of the Air Quality Network of the Portuguese Environment Agency (QualAr). Limits of EU Air Quality Directive (25 μg/m^3), WHO Air Quality Guidelines 2006 (10 μg/m^3) and WHO Air Quality Guidelines 2021 (5 μg/m^3) for $PM_{2.5}$ are included as red lines.

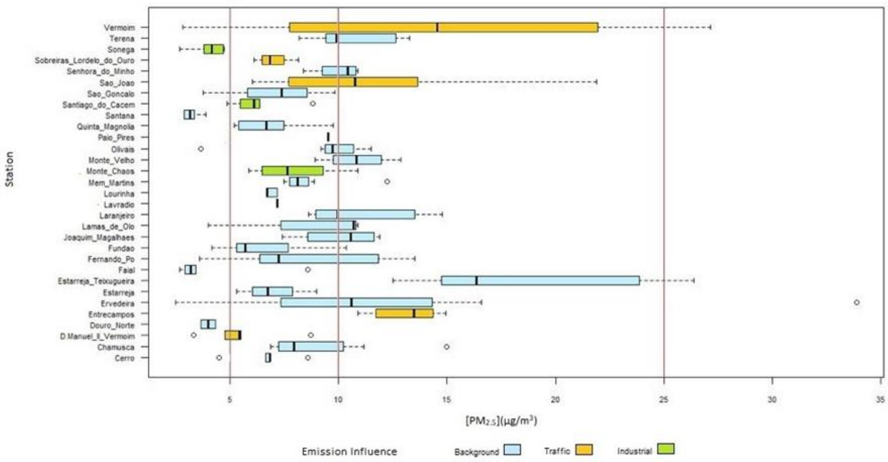

Figure S3. Concentrations of particulate matter ($PM_{2.5}$) (mean), by emission influence (background, traffic and industrial) and stations, in Portugal, in the period 2003–2021. Data represents average values collected from stations of the Air Quality Network of the Portuguese Environment Agency (QualAr). Limits of EU Air Quality Directive (25 μg/m^3), WHO Air Quality Guidelines 2006 (10 μg/m^3) and WHO Air Quality Guidelines 2021 (5 μg/m^3) for $PM_{2.5}$ are included as red lines.

11. Acknowledgements

This work is funded by FCT/MCTES through national funds to PMCardImpact (EXPL/SAU-PUB/0944/2021).