U.PORTO FEUP FACULDADE DE ENGENHARIA UNIVERSIDADE DO PORTO

Integrated Master's in Environmental Engineering

2016/2017

AIR POLLUTION: THE PORTUGUESE SCENARIO AND ITS PUBLIC HEALTH IMPACTS

Pedro Nuno Leal Ribeiro Torres

Dissertation for the Degree of:

MASTER IN ENVIRONMENTAL ENGINEERING

Supervisor FEUP: Maria do Carmo da Silva Pereira, PhD

Supervisor ISPUP: Ana Sofia Estevão Mendes, PhD

Porto, September 2017

The work presented in this thesis was performed at the Institute of Public Health of the University of Porto (ISPUP), Portugal, and the Environmental Health Department of the National Institute of Health, Porto, Portugal.

This work was supported by FEDER through the NO_2 Operational Program Competitiveness and Internationalization and national funding from the Foundation for Science and Technology – FCT (Portuguese Ministry of Science, Technology and Higher Education) (POCI-01-0145-FEDER-016752), under the project "FUTURAR - Air quality in Portugal in 2030: a policy support" (PTDC/AAG-MAA/2569/2014).

“Science seeks the truth. And it does not discriminate. For better or worse it finds things out. Science is humble. It knows what it knows and it knows what it doesn’t know. It bases its conclusions and beliefs on hard evidence -- evidence that is constantly updated and upgraded. It doesn’t get offended when new facts come along. It embraces the body of knowledge. It doesn’t hold on to medieval practices because they are tradition.”

- Ricky Gervais

ACKNOWLEDGEMENTS

I would like to express my sincere gratitude for the motivation and guidance of the supervisor of the Institute of Public Health of the University of Porto (ISPUP) and the Department of Environmental Health of the National Institute of Health (INSA), Ana Sofia Mendes, for all comments, observations and guidance during the whole process of this dissertation, as well as for the patience and availability, which not only enabled the execution of my work, but also helped my development as an engineer and even as a person.

I would also like to thank my supervisor at the Faculty of Engeny of the University of Porto, Professor Maria do Carmo Pereira (LEPABE), for providing the opportunity to work on this dissertation.

To all the staff of INSA and ISPUP, a huge word of gratitude, for welcoming me in their midst and for receiving me so well, for providing all the necessary conditions for this work. The presence of all these people not only helped me to feel at home, but also helped to improve my work and my professional performance throughout the process.

To the entities involved in this work, (FEUP, LEPABE, INSA and ISPUP), for having ensured all the necessary conditions for the elaboration of this work.

For Dr. Joana Ferreira, from the Center for Environmental and Marine Studies of the University of Aveiro, for advice and help in the development of the theme.

For my family and friends, thank you very much for the help given in every possible way, during all this work. Only with the support, dedication and patience received by them, was it possible to deal with all the problems faced with the work and to finish the dissertation.

A special thanks to Laura Correia for her unconditional and constant support during these months, which undoubtedly was an immense help, dedicating much of her time, patience and vacations, to support me and help me through the most difficult moments in this work .

ABSTRACT

To mitigate the effects of air pollution and reduce emissions and consequent concentrations of atmospheric pollutants, several tools have been developed to monitor the atmospheric pollution emissions and concentrations. Several monitoring stations are distributed around the world to provide a global, national and regional analysis of the existing air quality status. The Portuguese air quality index specifies the state of pollution in each territorial area and future projections of pollution.

This work comprises a literature review, critical analysis and Case Study of air quality indicators and its impacts in the field of public health. The main outcome of this study was to analyze the health impacts regarding the air quality indicators and pollutant emission scenario for Portugal, between the chosen years of 2009 and 2015.

This study was conducted between February and September 2017, focusing on the literature from 1995 to 2017, using the Web of Science, PubMed and ScienceDirect databases. The principal method of the Case Study was an emissions analysis, air quality pollutant concentrations analysis and health effects expressed by indicators and to discuss their possible relationships. Nevertheless, a direct analysis of the relations between these three inputs (emissions, air quality pollutant concentrations and health effects) was not a direct one, since many chemical and physical processes are involved, which play a central role in the development of pollutants and their geographical distribution.

The emissions analysis pointed out the following: (i) regions of *Alentejo* and *Algarve* had reduced values in different pollutants in comparison with the other regions, with some decreases of emissions in those regions; (ii) the metropolitan area of Lisbon presented the most problematic region in relation to the emissions of all the pollutants under study, with few improvements and high emissions in relation to all the other regions; and (iii) only in the country central region, there was an increase in carbon dioxide emissions.

The analysis of air quality showed that: (iv) regarding to ozone, concentration decreased throughout the country; (v) in the case of nitrogen dioxide and particulate matter, most of the northern half of the country shows an increase in concentrations, contrary to the situation observed in the southern half that presents improvements; (vi) the analysis of sulfur dioxide and fine particulate matter presented concentration increases in the regions of *Alentejo* and the metropolitan area of Lisbon, while other regions show improvements with the reduction of pollutants concentrations in the atmosphere.

In terms of public health, the studied health indicators presented different trends, depending on the cause of mortality and the region in question. *Alentejo* and *Algarve* regions showed decreasing trends, with those registering fewer causes of mortality in other regions. The regions of the North, Center and metropolitan area of Lisbon had high mortality values, compared to the other regions.

Possible actions to mitigate emissions of air pollutants should focus on the metropolitan area of Lisbon, North and Central parts of the country, particularly the districts of Lisbon, *Setúbal* and Porto.

RESUMO

A necessidade de mitigação dos efeitos da poluição e redução das emissões e das consequentes concentrações de poluentes atmosféricos despoletou o desenvolvimento de várias ferramentas de monitorização dos valores de poluição atmosférica registrados, como o índice de qualidade do ar. O índice de qualidade do ar específica o estado da poluição em cada área territorial e futuras projeções de poluição. Várias estações de monitorização estão distribuídas em todo o mundo e fornecem uma análise global, nacional e regional do *status* da qualidade do ar.

Este estudo compreende uma revisão da literatura, análise crítica e caso de estudo de indicadores de qualidade do ar e seus impactos na saúde pública. O principal objetivo deste estudo foi analisar os impactos na saúde em relação aos indicadores de qualidade do ar e cenário de emissão de poluentes para Portugal. Este trabalho foi realizado entre fevereiro e setembro de 2017, com foco na literatura de 1995 a 2017, usando as bases de pesquisa da *Web of Science*, *PubMed* e *ScienceDirect*. A metodologia principal aplicada ao caso de estudo foi a análise de emissões, concentrações de poluentes, da qualidade do ar, efeitos sobre a saúde, no seu desenvolvimento e tendências entre os anos 2009 e 2015, bem como, a discussão das suas potenciais relações. Atendendo ao objetivo principal deste estudo, foi necessário considerar que uma análise direta entre emissões, concentrações poluentes da qualidade do ar e efeitos sobre a saúde, não era aplicável considerando os diversos processos químicos e físicos que estão envolvidos, e que desempenham um papel central no desenvolvimento de poluentes e na sua distribuição geográfica.

A análise das emissões destacou vários pontos: (i) Alentejo e o Algarve com valores de emissões reduzidos para quase todos os poluentes em estudo comparativamente às outras regiões; (ii) a área metropolitana de Lisboa é a região mais problemática quanto às emissões, com poucas melhorias e elevadas emissões comparativamente às outras regiões; (iii) a região central do país apresentou um incremento das emissões de dióxido de carbono.

A análise da qualidade do ar apresentou: (iv) a concentração de Ozono troposférico diminuiu em todo o país; (v) no caso dos poluentes, dióxido de nitrogénio e matéria particulada de diâmetro igual ou inferior a 10 micrómetros (PM10), grande parte do norte do país apresenta um aumento nas respetivas concentrações; situação contrária observada na metade sul do país que apresentou melhorias; (vi) na análise de dióxido de enxofre e matéria particulada de diâmetro igual ou inferior a 2.5 micrómetros (PM2.5), as regiões do Alentejo e a área metropolitana de Lisboa mostraram um aumento, enquanto outras regiões apresentaram melhorias com a redução das concentrações de poluentes na atmosfera.

Em termos de saúde pública, os indicadores de saúde estudados mostraram diferentes tendências, dependendo da causa da mortalidade e da região em questão, destacando-se as melhorias, principalmente nas regiões alentejana e algarvia. As regiões do Norte, Centro e área metropolitana de Lisboa são regiões que apresentam elevados índices de mortalidade, em comparação com as outras regiões. As possíveis ações para mitigar as emissões de poluentes atmosféricos devem ser concentradas na área metropolitana de Lisboa, nas regiões Norte e Centro, em particular nos distritos de Lisboa, Setúbal e Porto.

LIST OF PUBLICATIONS

POSTER PRESENTATION

Air Pollution and its Adverse Impacts on Human Health: FUTURAR project review
Ana Mendes, Solange Costa, Joana Ferreira, Joana Leitão, Pedro Torres, Carlos Silveira, Hélder Relvas, Myriam Lopes, Alexandra Monteiro, Peter Roebeling, Ana Isabel Miranda, João Paulo Teixeira
Young Researchers Day 2017 organized by National Health Institute Dr. Ricardo Jorge, 8th May, Lisbon, Portugal.

ARTICLES IN NATIONAL JOURNALS WITH SCIENTIFIC PEER-REVIEW

Impactes da poluição atmosférica na Saúde: Perspetivas do Projeto FUTURAR
Ana Mendes, Solange Costa, Joana Ferreira, Joana Leitão, Pedro Torres, Carlos Silveira, Hélder Relvas, Myriam Lopes, Alexandra Monteiro, Peter Roebeling, Ana Isabel Miranda, João Paulo Teixeira
Boletim Epidemiológico Observações do Instituto Nacional de Saúde Doutor Ricardo Jorge, Volume 6 – Número Especial 9, Doenças Não Transmissíveis, 2017 ISSN: 0874-2928 | ESSN: 2182-8873 – Disponível em: http://www.insa.min-saude.pt/category/informacao-e-cultura-cientifica/publicacoes/boletim-epidemiologico-observacoes/#sthash.qfmiidyr.dpuf

ARTICLES IN INTERNATIONAL JOURNALS WITH SCIENTIFIC PEER-REVIEW

To be submitted to Atmospheric Environment (IF: 3.629; Q1)
Air Pollution: a scenario approach and its Public Health Impacts
Pedro Torres, Joana Ferreira, Carlos Silveira, Solange Costa, Maria do Carmo Pereira, Hélder Relvas, Myriam Lopes, Alexandra Monteiro, Peter Roebeling, Ana Isabel Miranda, João Paulo Teixeira, Ana Mendes

LIST OF TABLES

LIST OF FIGURES

LIST OF EQUATIONS

ABBREVIATIONS

$(NH_4)_2SO_4$	------------------	Ammonium sulphate
µg/m^3	------------------	Microgram per cubic meter
µm	------------------	Micrometre
AIDS	------------------	Acquired Immune Deficiency Syndrome
APA	------------------	Associação Portuguesa do Ambiente
AQG	------------------	Advanced Quality Group
AQI	------------------	Air quality index
AQS	------------------	Air Quality Standard
ARF	------------------	Acute Respiratory Failure
atm	------------------	Atmosphere (unit of pressure)
BaP	------------------	Benzo(a)pyrene
BOD	------------------	Biological Oxygen Demand
C3a	------------------	Anaphylatoxin fragment
C_6H_6	------------------	Benzene
CAFE	------------------	Clean Air for Europe Programme
CH_4	------------------	Methane
CO	------------------	Carbon monoxide
CO_2	------------------	Carbon dioxide
COD	------------------	Chemical Oxygen Demand
COHb	------------------	Carboxyl HemogloBin
COPD	------------------	Chronic Obstructive Pulmonary Disease
DNA	------------------	Deoxyribonucleic acid
Dp	------------------	Diameter of particle
DPSEEA	------------------	Driving Force-Pressure-State-Exposure-Effect-Action
EC	------------------	European Commision
EEA	------------------	European Environment Agency
EFFIS	------------------	European Forest Fire Information System
EU	------------------	European Union
FCT	------------------	Foundation for Science and Technology
FEDER	------------------	European Regional Development Fund
FEUP	------------------	Faculty of Engineering of the University of Porto
GHG	------------------	Green House Gases
H·	------------------	Hydrogen free radical
H_2	------------------	Hydrogen
H_2O	------------------	Chemical formula of water
H_2SO_4	------------------	Sulfuric acid
HA	------------------	Hyaluronic acid
HbA	------------------	Adult hemoglobin
HbF	------------------	Fetal hemoglobin

HCl	------------------	Hydrogen chloride
Hg	------------------	Mercury
HIV	------------------	Human Immunodeficiency Virus
HNO_3	------------------	Nitric acid
hv	------------------	High Voltage
IHR	------------------	Internet Health Resources
INE	------------------	National statistical institute
INERPA	------------------	National Inventory of Atmospheric Emissions
IPCC	------------------	Intergovernmental Panel on Climate Change
IQAr	------------------	Air Quality Index
ISPUP	------------------	Institute of Public Health, University of Porto
IUPAC	------------------	International Union of Pure and Applied Chemistry
Kg/m^3	------------------	Kilogram per cubic metre
LEPABE	------------------	Laboratory for Process Engineering, Environment, Biotechnology and Energy
MF	------------------	Masculine/Feminine
Mt	------------------	Megatonne
N	------------------	Element Nitrogen
N_2	------------------	Atmospheric Nitrogen
N_2O	------------------	Nitrous oxide
NCO·	------------------	Isocyanato radical
NEC	------------------	National Emission Ceilings
NH·	------------------	Aminylene
NH_3	------------------	Ammonia
NH_4^+	------------------	Ammonium
NH_4NO_3	------------------	Ammonium nitrate
NO	------------------	Nitrogen oxide
NO_2	------------------	Nitrogen dioxide
NO_2^-	------------------	Nitrite ion
NO_3^-	------------------	Nitrate ion
NOx	------------------	Generic term for the nitrogen oxides
O	------------------	Element Oxygen
$O(^1D)$	------------------	Oxygen atom in an excited singlet state
$O(^3P)$	------------------	Atomic Oxygen
O_2	------------------	Atmospheric oxygen
O_3	------------------	Ozone
OH·	------------------	Hydroxyl radical
PGIg	------------------	Prostacyclin
pH	------------------	potential of Hydrogen
PM	------------------	Particulate Matter
PM0,1	------------------	Particulate Matter of 0,1 µm in Diameter

PM10	------------------	Particulate Matter of 10 µm in Diameter
PM2,5	------------------	Particulate Matter of 2,5 µm in Diameter
PmV	------------------	Pulmonary Microvascular Pressure
ppb	------------------	Parts per Billion
ppm	------------------	Parts per million
PRIME	------------------	Plume RIse Model Enhancements
RAINS	------------------	Regional Air Pollution INformation and Simulation
R'CHO	------------------	Aldehyde
RH	------------------	Generic volatile organic compound
RO	------------------	Alkoxyl radical
RO_2	------------------	Peroxy radical
SICO	------------------	Information System for Death Certificates
SIRIC	------------------	System of Civil Registry and Identification
SO_2	------------------	Sulfur dioxide
SO_3	------------------	Sulfur trioxide
SOx	------------------	Sulfur Oxides
$t.Km^{-2}$	------------------	Ton per square kilometer
TB	------------------	Tuberculosis
TxAg	------------------	Triacylglycerol
UFP	------------------	Ultra-Fine Particle
VOC	------------------	Volatile Organic Compound
WHO	------------------	World Health Organization
YLL	------------------	Years of life lost (percentage of total)

TABLE OF CONTENTS

PART I - INTRODUCTION

1. OVERVIEW

Air pollution has wide-ranging and harmful effects on human health and is a major issue for the global community and urban sustainability [1, 2]. Outdoor air quality plays an important role in public health and has long been a concern for citizens [3].

In order to mitigate the effects and reduce emissions and consequent concentrations of atmospheric pollutants, several tools have been developed to monitor the development of recorded atmospheric pollution values, serving as indicators of the environmental status of the areas concerned. One such indicator often used is the air quality index.

The Portuguese air quality index is an indicator that quantitatively evaluates several environmental parameters, resulting in a global classification, providing a generalized view of the situation in question. The reason air quality index differ from country to country is that the choice of parameters for this indicator varies according to the entity responsible for determining the indicator, and because of that, it is always required the list of parameters used when the air quality index is presented.

This air quality indicator specifies the state of pollution in each territorial area and future projections of pollution. Moreover, this indicator has a ratio proportional to the percentage of population in question to sense the harmful health effects that derives from the aggravation of the pollution [4]. Several monitoring stations are distributed around the world to provide a global, national and regional analysis of the existing air quality status. The air quality index is the result of a specific method of calculation, using reference values for various substances (all calculated and calibrated for measurements). This information is later treated and, using color coding, presented in multichromatic maps, also considering the health effects associated with the substances.

The resulting values are then compared with the ranges of concentration values associated with a color scale in which areas not covered by the index calculation method are shown in gray as shown in Figure 1.

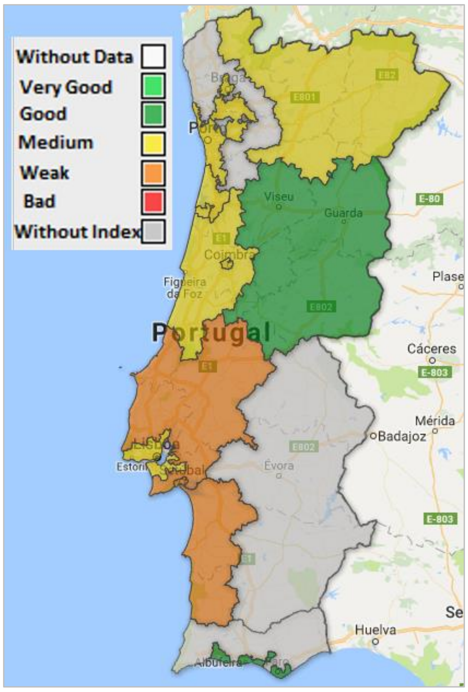


Figure 1 - Map of the air quality index in Portugal (information for August 12, 2017) Adapted [5]

In 2010, urban areas contained more than half of the world's population, and in Europe, three-quarters of the population lived in cities, as presented in Figure 2. Studies point out to a tendency to increase these values: by the year 2030 it is speculated that six decimals of the world population will live in urban environment and by the year 2050, it is estimated to a value of seven tenths [6].

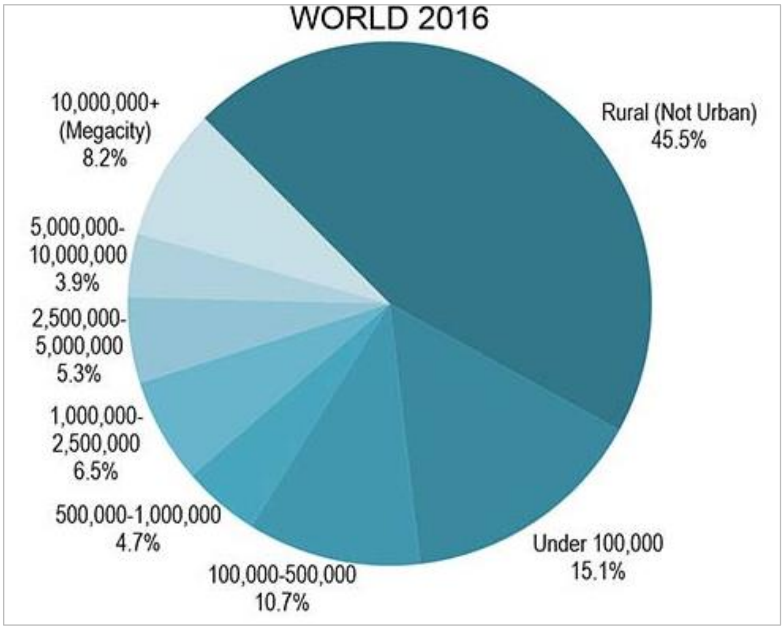


Figure 2 - World Population Distribution: Urban & Rural [7]

With the industrial advances and technological developments observed in the last decades, a crucial factor for the evaluation of society development will be the environmental evaluation of the industrial sectors and the daily life of the population. In developed countries, outdoor air pollution comes mainly from industry, automotive combustion vehicles, power generation and domestic activities [8]. From an environmental perspective, the assessment of emissions of air pollutants is crucial because of the increase in emission levels for anthropogenic air pollutants, which far exceed the values recorded in recent decades [9].

Studies have been carried out to identify pollutant sources and the physicochemical characterization of atmospheric pollutants and their harmful contributions to the population, thus allowing the identification of sources of pollutant emissions such as the burning of fossil fuels [8].

Since the beginning of the 20th century, public health problems have attracted attention, especially in urban centers, due to its greater concentration of pollutants [9]. In urban centers, industrial and vehicular emissions are of major importance but in rural areas and smaller cities, biomass burning and forest fires have a major share in the contribution to air pollution in rural centers [8].

The International Union of Pure and Applied Chemistry (IUPAC) defines air pollution as "the presence of substances in the atmosphere, resulting either from human activity or natural processes, present in sufficient concentration, for a sufficient time and under circumstances such as to interfere with comfort, health or welfare of persons or the environment". As such, a quantitative analysis of air pollutants and their respective effects on health is vital, providing all the necessary conditions for the design of strategies to reduce emissions of air pollutants.

Air pollution is a mixture of compounds that vary depending on the emitting sources, and the processes involving the emitted pollutants. There are four main types of air pollution sources: mobile sources – such as cars, buses, planes, trucks, and trains; stationary sources – such as power plants, oil refineries, industrial facilities, and factories; area sources – such as agricultural areas, cities, and wood burning fireplaces; and natural sources – such as wind-blown dust, wildfires, and volcanoes [10] (Figure 3). Generally, pollutants monitored by air quality monitoring networks are: Ozone (O_3), Lead (Pb), Carbon monoxide (CO), Sulfur oxides (SO_x), Nitrogen oxides (NO_x), Particulate matter (PM), Ammonia (NH_3) and Methane (CH_4) [8].

It is understood as primary air pollutant, all the pollutant emitted directly to the air, coming from the emitting sources [11]. Dusts, fumes and a wide range of toxic chemicals can be included in the classification of primary pollutants, being exhaust fumes from road traffic and industrial chimneys, presented as some of the sources of primary pollution [12]. This type of pollutant can directly affect the population through inhalation or skin contact, but can also serve as a precursor to secondary air pollutants [11].

For secondary pollutants, this classification is subsequently to the fact that these pollutants are not emitted from conventional sources, but instead are generated or modified from the primary pollutants. The generation of the secondary pollutants occurs by reactions of chemical compounds present in the atmosphere or by reactions with sunlight, taking photochemical reactions [12].

Pollution from human-generated and natural sources are created in one place, transported through the air, and sometimes changed by chemical reactions before being deposited (Figure 3) [13]. The effects of this pollution can be local, as well as, global since air pollution is a transboundary issue.

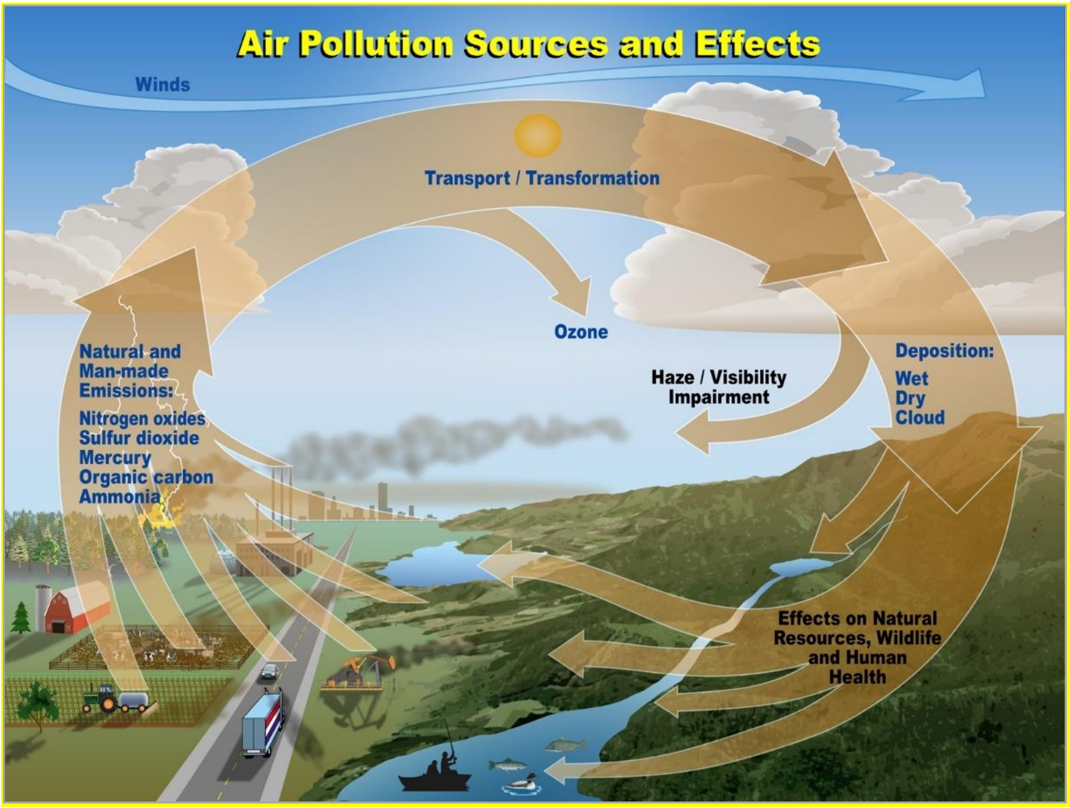


Figure 3 - Air pollution sources and effects [13]

The association between air pollution and the impact on health of people with respiratory and cardiovascular diseases has been demonstrated by epidemiological studies [14-22]. It was also proven a relationship with the increase in mortality and morbidity due to cardiovascular and respiratory disease in susceptible population [23-27]. Moreover, it have been shown that atmospheric pollutants such as O_3 and PM cause health risks in many developed countries and have had harmful effects on the population at increasingly lower concentrations [28].

Annually, reports have been published regarding emissions of air pollutants [29], covering the European panorama with national geographic scale, that is, these report cover all the member countries of the EU, individually. Different situation in this work, which addresses only Portugal but on a regional scale.

Air pollution is not limited to the political and geographical borders of countries, therefore international action is required to reduce levels of pollutant emissions worldwide. A report released by the World Health Organization (WHO) has recognized air pollution as the greatest health risk [30]. In 2005, the WHO published an update to the Air Quality Guidelines, which set the limit values for exposure to air pollutants and public health consequences. Since then several reports released by WHO, have been focusing the health and economic impacts and costs of air pollution [31-35], as well as, reviews for several outdoor and indoor air pollutants [36-39].

The need to devise emission reduction strategies at a global level is closely linked to past events that have brought large numbers of casualties and have had serious adverse effects on the population. The table 1 lists some of the major events related to outdoor air pollution throughout history.

Table 1 - Air pollution incidents throughout history [40]

Location	Air Pollution Incident	Date
Meuse Valley, Belgium **[41]**	Fog	1930
Los Angeles, America	Photochemical Smog	1940
Donora, America **[42]**	Haze	1948
London, England **[43]**	Smog	1952
Several cities, China **[44]**	Haze	Current

Air pollutants present changes in their mobility and durability when climate change is imposed in their environment. Conditioning factors such as temperature, wind speed and direction, and precipitation are some of the meteorological factors that cause changes in the implications associated with the exposure of the pollutants to the population. Weather conditions are some of the factors that divide the exposure of pollutants into two types according to their duration, short-term and long-term exposure [45].

Although there has been a decline in atmospheric pollutant concentrations in some parts of the world, recent studies [46-48] corroborate the long-term exposure of pollutants with mortality from cardiopulmonary and lung cancer [49].

Regarding short-term exposure, there is a greater severity with the direct and immediate contact of the pollutant, affecting profoundly the first immunologic barriers of the organism. Table 2 presents some of the effects associated to atmospheric pollutants considering different time intervals of exposure of the population.

Table 2 - Health effects due to exposure to pollutant [15]

Pollutant	Health effects related	
	Short-term exposure	Long-term exposure
Nitrogen Dioxides (NO_2)	Effects on lung function, especially asthmatics; Increased inflammatory reactions in the airways; Increase in hospital admissions; Increased mortality	Reduction of lung function; Increased likelihood of respiratory symptoms
Tropospheric ozone (O_3)	Adverse effects on lung function and symptoms; Inflammatory reactions of the lung; Increased use of medication; Increase in hospital admissions; Increased mortality	Reduction of lung function development
Particulate Matter (PM)	Inflammatory reactions of the lung; Respiratory symptoms; Adverse effects on the cardiovascular system; Increased use of medication; Increase in hospital admissions; Increased mortality	Increased respiratory symptoms of the "lower" airways; Reduction of lung function in children and adults; Increased COPD (Chronic Obstructive Pulmonary Disease); Reduction in life expectancy
Sulfur Dioxide (SO_2)	Increase in hospital admissions for cardiovascular diseases; Increased mortality in children due to respiratory diseases	Reduction of lung function; Increased resistance of specific airways and respiratory symptoms (Effects resulting from exposure to particulates)
Carbon Monoxide (CO)	Effects related to interference in the transport of O2 (hypoxia, neurological alterations - behavioral, among others); Increased daily mortality and hospital admissions for cardiovascular diseases	-
Volatile Organic Compounds (VOCs)	-	Reduction of lung function; Increased likelihood of respiratory symptoms

These harmful effects on the population health may be accompanied by negative impacts on biodiversity both in areas close to emission sources and in remote areas due to transport phenomena of long-range pollutants [50].

In addition to economic impacts, represented by increased medical costs, number of obits and decreases in productivity indexes for days lost, air pollution is also responsible for damage to buildings, roads, bridges and other public constructions [51].

Featuring such a wide range of adverse effects, a spreading rate of harmful effects and a widespread cause-and-effect relationship to such a diverse geographic dimension, from local to global levels, air pollution is a global challenge, requiring in the initial phase, corrective and preventive measures at the local level [52].

This work also deals with issues related to the relationship between the evolutionary trends of atmospheric pollutants and the evolution of selected health indicators to qualify public health, particularly in mainland Portugal.

In the course of this work, brief descriptions of atmospheric pollutants considered to be of greater influence in public health will be presented, some of which will be the subject of further study in this work, from emission sources to associated effects in public health.

In addition to having a special focus on these pollutants as they have the greatest environmental impact, these pollutants are also identified as some pollutants with harmful effects and more severe associated medical complications.

The cause - effect relationship found between pollutants and health indicators is best documented and quantified for PM [53, 54], as the strongest cause-and-effect relationship between pollutants [55-57].

Associated with air pollution, harmful effects on health encompass a range of steps, from harmless irritations to death. For each individual, the sensed symptoms may present diverse levels of spread and seriousness, depending on a set of endogenous and exogenous factors, from immunology and biochemical factors to climatic conditions and viral infections [58-60].

This work comprises a literature review, critical analysis and Case Study of air quality indicators and its impacts in the field of public health. The studied pollutants were chosen among those responsible for acidification, eutrophication and ground-level O_3 pollution.

2. AIMS

The main outcome of this study was to analyze the health impacts regarding the air quality indicators and pollutant emission scenario for Portugal.

A set of conclusions was produced focusing the evolution of air pollutants concentrations and emissions between 2009 and 2015 and its public health impacts. This review and Case Study sets important information for environmental and public health decision-making processes of the Portuguese representatives.

To accomplish the proposed main goal, the following specific objectives were set along the work.

i. Review and critical analysis of air quality indicators and its impacts in the field of public health;
ii. Review of national pollutant emissions survey;
iii. Review of the Portuguese air quality index;
iv. Study of the framework of emissions of atmospheric pollutants in a national perspective;
v. Review and study of the public health impacts due to air pollution considering the Portuguese scenario.

3. METHODOLOGY

3.1. LITERATURE REVIEW ANALYSIS

This review was conducted between February and September 2017, focusing on the literature from 1995 to 2017, using the Web of Science, PubMed and ScienceDirect databases. The keywords and phrases used for the search were the combinations of terms "pollution", "air", "health effects", "environmental guidelines", "heavy metals", "air quality index", "Particles", "nitrogen emissions", "ozone", "tropospheric ozone", "acid deposition", "inhalation of smoke", "methane ", "emission reduction scenarios", "emission trends", "Ammonia", "carbon monoxide", "carbon dioxide", "European Environment Agency", "sulfuric acid", "volatile organic compounds", "environmental legislation", "particulate matter AND air quality AND health effects" , "NO2 AND air quality AND health effects", "ozone AND air quality AND health effects", "ammonia AND air quality AND health effects" and "methane AND air quality AND health effects".
The survey included articles in English and Portuguese, review articles, case studies, and international and national reports. Articles older than 1995 were excluded, given that the older articles used, focus more on specific case studies of past events involving the topic in question. Other exclusion criteria focused on repeated articles and articles that did not cover the specific subject matter required.

Figure 4 presents the flowchart of the research material selection process.

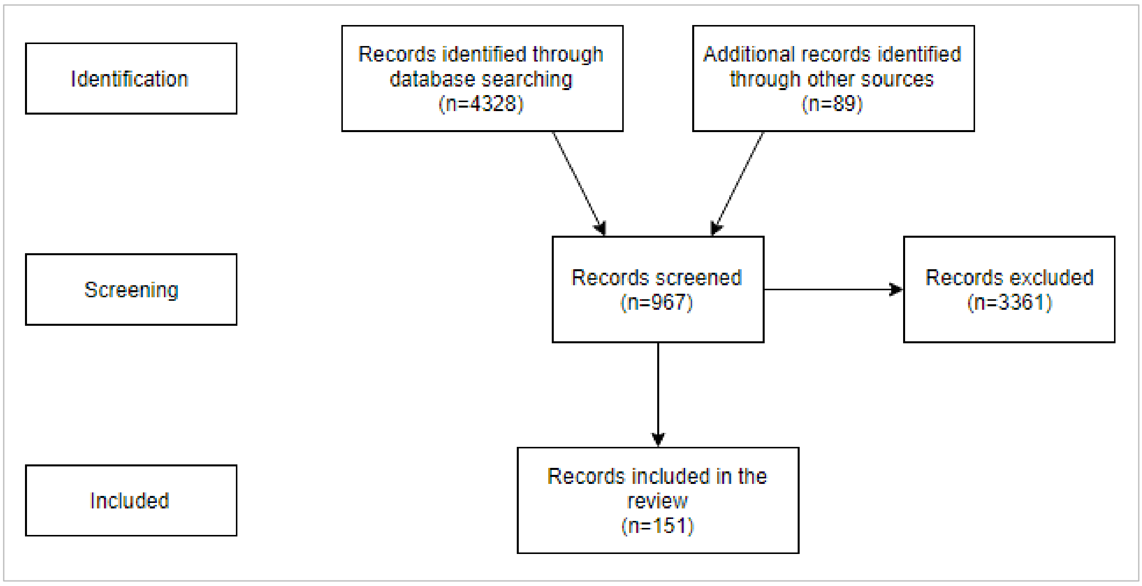


Figure 4 - Search material selection process flowchart

3.2. Case Study Data Analysis

In this paper, several criteria for selecting information were taken into consideration, with the purpose of creating a concise research and conclusions relevant to the focus of this study.

All the decision-making processes during this study were always in compliance with the selected criteria, which were divided into four stages: (i) selection of three study fronts (emissions, concentrations, and health indicators); (ii) selection of years used for the study (2009 and 2015); (iii) selection of information on emissions of air pollutants at national level; (iv) selection of information for concentrations; and selection of health indicator information.

The principal method of the Case Study was the emissions analysis, air quality pollutant concentrations and health effects expressed by indicators and their possible relationships. All the criteria related to the choice of geographic scales and time intervals took into consideration the most recent information available, as well as, selecting information in order to facilitate the qualitative analysis of the data collected, thus helping to understand the conclusions obtained in this work.

Considering the main objective of this study, it was necessary to consider that a direct analysis of the relations between these three inputs (emissions, air quality pollutant concentrations and health effects) was not a direct one, since many chemical and physical processes are involved, which play a central role in the development of pollutants and their geographical distribution. These processes are based on: transport of air pollutants, conditioned by weather conditions; chemical transformation processes, such as the generation of secondary pollutants and associated factors; and deposition, which is also associated with climatic factors.

For this study, the years 2009 and 2015 were used, not because there were significant meteorological or environmental anomalies recorded in those years, but as were the two most recent years with reports presented by the main source of information selected for this study (INERPA - National Inventory Of Emissions [61]) for the emissions part. Additionally, the years 2009 and 2015 were selected, since they are the most recent years where an aggregation of information is presented [62], of all the pollutants.

Regarding the other parts of the study, the years used were the same as those of the emissions, even though there are data referring to more recent years, since data coherence is a curricular factor for studies of this type.

For the collection of information on air pollutant emissions, the focus was on collecting information from government sources only to guarantee impartiality in data acquisition.

The National Pollutant Emission Inventory (INERPA) was the source of information for the emission of pollutants, being an instrument of analysis of air quality, whose entity responsible for its performance is the Portuguese Environmental Agency (APA).

For the analysis of the spatial distribution of pollutant emissions for the years under study, the decision was taken to aggregate the values in regions, resulting in maps of the regions emissions in mainland Portugal, similar to the maps shown in Figure 5, differentiated only on the geographic scale used In the analysis of the data. The process of choosing the geographic scale in the issue of emissions took into consideration the presentation of the data referring to the public health indicators, which are also found in areas of the country: North, Center; Alentejo; Algarve and Lisbon.

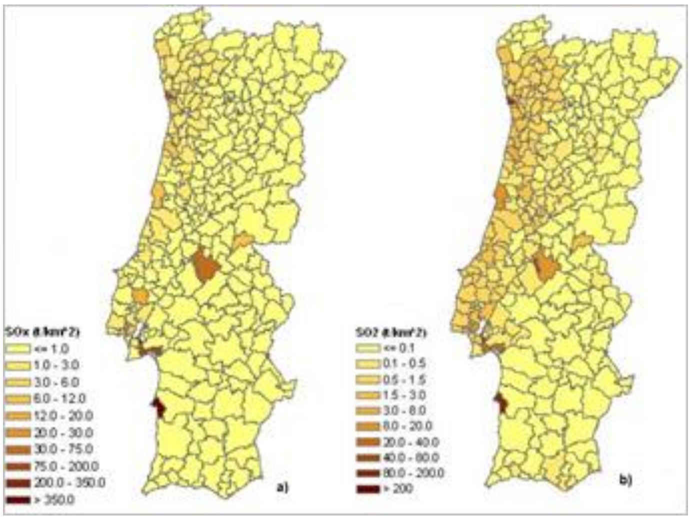


Figure 5 - Examples of maps of spatial distribution of SOx in continental Portugal for the years 2005 (a) and 2008 (b) [63, 64]

This aggregation was necessary because, for the emissions data, all values were grouped into municipalities and divided into emission sector, and for the latter, it was also necessary to aggregate the data to obtain a general perception of all the resulting emissions in each district in the two years in question.
The analysis of emissions was based on the study of emission maps, which followed certain specifications, such as the presentation of the same APA color scheme in previous year's emission reports, as well as the use of the same ranges of emission values presented in map captions.

This part of the information was obtained through the APA's Online Air Quality Database, which is a multifaceted tool that publishes information from air quality indexes to pollutant concentrations statistics at each monitoring station.
Regarding the spatial distribution, air monitoring stations do not cover the entire national territory and are not uniformly dispersed, with a greater concentration of stations in the coastal zone of

the country. The spatial divisions used in the presentation of the concentration values in the database used are referred to as Air Quality Index zones (IQAr zones), and only one monitoring station was chosen for each zone for each type station (background, traffic and industrial).

In the existence of more than one air monitoring station, in a given zone, the selection criterion fell on the population density of the area covered by the stations in question, giving priority to the station with the highest population density.

After the selection of the monitoring stations, the validation criteria of concentration values presented in the database were delineated. Values presented with null efficiencies were discarded and presented as nonexistent, as when they were presented as "???".

This third part of the information collection focuses on the main public health indicators related to medical complications in the field of respiratory and cardiac systems.

As in the previous stages, it was important to gather information from public governmental sources and, unlike information on emissions and concentrations, information was collected from 2009 to 2015, in order to more rigorously assess evolutionary trends of public health in Portugal.

The main indicators related to the respiratory system and some indicators related to cardiac complications with possible causes associated to air pollution were selected, resulting in a set of eight health indicators, for the years 2009 to 2015.
As the required information was available at various geographic levels, it was decided to select information by zones of Portugal (North, Center, Metropolitan Area of Lisbon, Alentejo and Algarve), since the remaining options were too comprehensive (national geographic level) or with own type of aggregation without any coincidence with the municipal, district or IQAr zones.

The option to graph the information in question was chosen with the purpose of facilitating the temporal analysis of the evolutionary trends of the parameters indicative of the Portuguese public health situation, until the year 2015 (the most recent year available).

PART II - PRESENT STUDY

4. LITERATURE REVIEW

4.1. PARTICULATE MATTER

As the pollutant that most affects the population, PM consists of sulfate, nitrates, sodium chloride, coal, mineral dust, water, ammonia, biological compounds, organic compounds and various metals, being a complex mixture of material in the solid state, liquid and gaseous, suspended in the air [34].

Of these particles, the smaller particles (≤10 micrometers in diameter) are the most harmful particles, because they penetrate and lodge in the lungs. Although an even smaller particle size (≤2.5 micrometers – PM2.5) is recognized which, because they are even smaller particles, cause more severe damages to the respiratory system [34].

Considering the varied size found in the particles, various effects can be recorded with different gravity, always starting from the principle that the smaller the particle, the more severe the effects, as represented in the Figure 6.

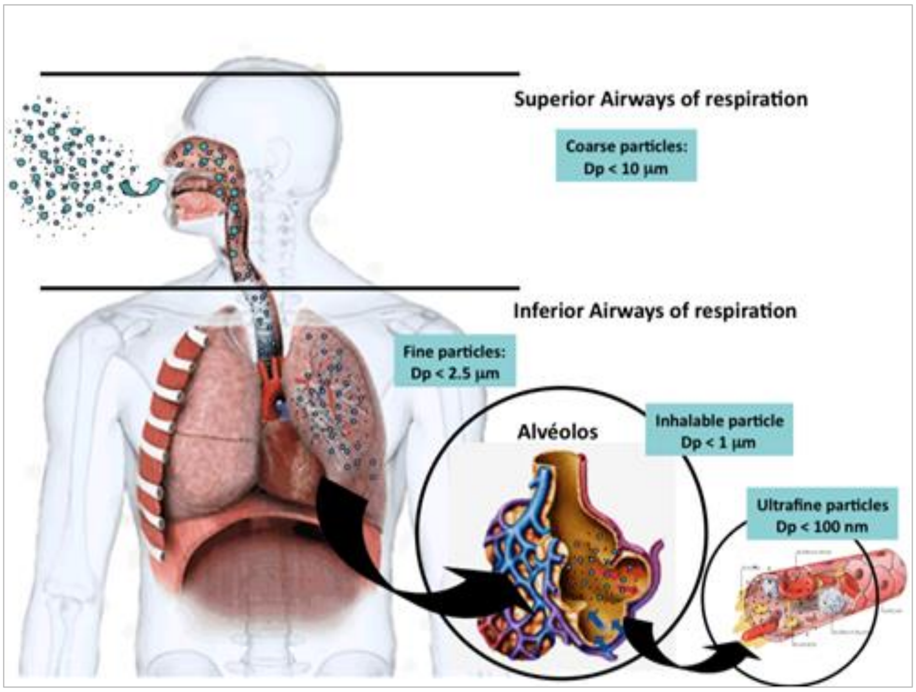


Figure 6 - Lungs penetration of difference sizes of PM: Represents the areas where PM from incomplete combustion processes is deposited in the body [65]

The organic matter that can be extracted from PM2.5 as volatile and semi-volatile organic compounds includes thousands of chemicals ranging from aromatic and alkaline compounds to polar carboxylic and aromatic acids [66].

The particle size varies and therefore different categories are defined: coarse particles, larger than 1 μm and ultrafine particles, smaller than 0.1 μm in the aerodynamic diameter, whereas the thin and ultrafine are capable of reaching the pulmonary alveoli in the upper respiratory tract, as represented in the Figure 7 [67].

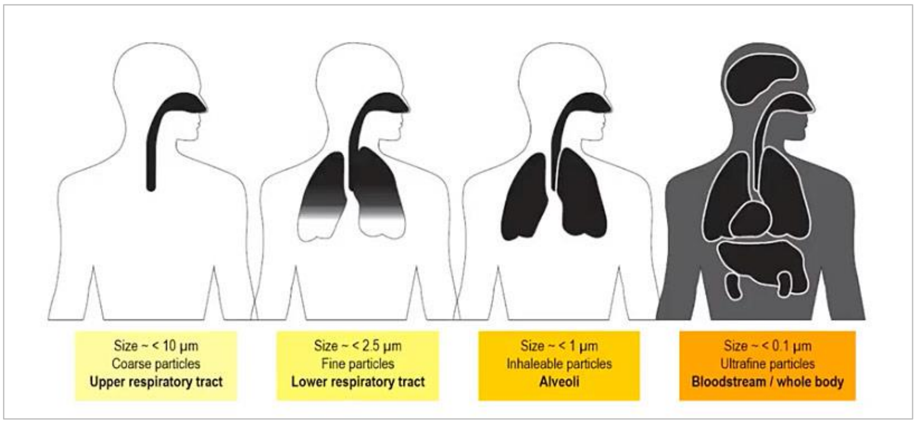


Figure 7 - Penetration of different-sized particles into the human body [68]

The airborne particles present irregularities both in the form and in their aerodynamic behavior, using terms tailored to the diameter of an ideal spherical particle, as aerodynamic diameter. The aerodynamic diameter is commonly identified as the particle size, being the sampling basis for the particles. This term is defined as the diameter of the spherical particle of density 1000 kg/m^3 and the rate of sedimentation equal to that of an irregular particle. The aerodynamic diameter alone is not sufficient to determine the shape or dimensions of a particle, as there may be particles with same aerodynamic diameter and different shapes.

Epidemiological studies have indicated that exposure to PM contributes significantly to mortality from pulmonary and cardiovascular diseases, also showing that long exposure to this pollutant results in harmful physiological effects such as altered lung function, increased blood plasma viscosity and induction of the systemic pro-inflammatory stimulus, pro-coagulant and oxidative stress.
Other consequences for human health have also been documented in several studies, reaching a wide range of effects, such as respiratory complications in children, reproductive effects, genetic damage and carcinogenic risk [66].

In accordance with the European air quality guidelines and the WHO, the values given in the Table 3 refer to the emission limit values for PM according to their aerodynamic diameter.

Table 3 - Air quality limit values for PM10 and PM2.5 as given in the EU Ambient Air Quality Directive and WHO AQGs [29]

Size Fraction	Averaging Period	EU Air Quality Directive	WHO Guidelines
PM10	1 day	50 µg/m3	50 µg/m3
	Calendar Year	40 µg/m3	20 µg/m3
PM2.5	1 day	-	25 µg/m3
	Calendar Year	25 µg/m3	20 µg/m3

The PM2.5 guideline results from the conversion of the PM10 guideline, with a ratio of 0.5 to PM2.5/PM10 in urban areas, and a ratio of 0.5 to 0.8 for rural areas [34].
Some studies indicate that PM2.5 is responsible for about 0.8 million premature deaths per year and about 6.4 million deaths globally, accounting for the annual reduction of one year of life expectancy in Europe [6].

Other studies point to the number of premature deaths to higher values such as 4 million people in the year 2012, thanks to the harmful effects of atmospheric particulate pollution, with road traffic being a major contributor to the emissions of this pollutant, particularly in urban areas [69].

Concerning the phenomenology of the PM, information was compiled in a report [29] that concludes that sulfate and organic matter are the main tributaries for the registered values of the average annual concentrations of mixed particulate material, considering that there is no mineral deposit, because if there is, this mineral dust is also a major contributor to PM10 concentrations.

Because of its complexity and variation in size, differences in exposure assessment and human dosage, several terms were used to describe PM.
Certain terms used come from the sampling methods used, resulting in "particulate matter" and "black smoke".

The site of deposition in the respiratory tract also serves to create other definitions such as "inhalable particles" relating to the respiratory tract, "thoracic particles", which are deposited in the lower respiratory tract and "respirable particles", which penetrate the region of pulmonary gas exchange, as presented in Figure 8. Other terms like PM10 come from the physiology of particles [70].

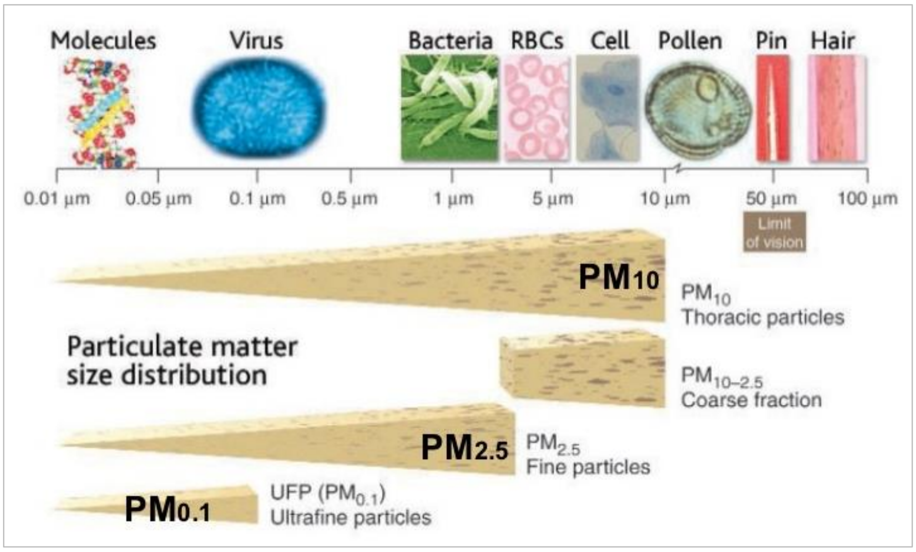


Figure 8 - PM size distribution [71]

Analyzing the trends of progression of emissions of PM in the atmosphere may be necessary to estimate the future concentrations of the particles, assuming, with some limitations, the same rate of progression recorded in the period of analysis.

With this projection of future emission concentrations, compliance with the European air quality standards established up to 2020 by the Clean Air Policy Package can be efficiently performed. When it comes to trends in the concentration of pollutants, limiting the study sample to only emissions of primary PM and precursor gases is difficult because the composition of monitored particles is not widely disseminated by the Environmental Air Quality Directive [29].

4.2. NITROGEN OXIDES

Coming from the chemical combination of nitric oxide (NO) and NO_2, NO_x are formed from combustion at high temperatures, usually from combustion processes of vehicle engines and power plants. Given that NO_2 is the main component of the oxides, this is the one with the most harmful effects on human health [72].
Nitrogen oxides can be divided into three categories depending on their formation mechanism: fuel oxides, rapid oxidation and thermal oxidation [73].

Atmospheric nitrogen fixation (NO_x thermal) and oxy-oxidation of nitrogen compounds from pulverized coal (NO_x fuel) are the main sources of NO_x production [74].

In order to form nitric oxide, the reaction of oxygen atoms coming from the dissociation of O_2 with nitrogen must occur. The necessary reactions are referred to as Zeldovich's mechanism:

$$N_2 + O\cdot \leftrightarrow NO + N \quad (1)$$

$$N\cdot + O_2 \leftrightarrow NO + O \quad (2)$$

Sensitive to temperature, stoichiometry and residence time, these reactions require a high temperature for the dissociation of oxygen and to exceed the activation energy in order to break the triple bond of the nitrogen molecule [74].
Having as main source of emission the combustion of fossil fuels, NO_x present in the combustion phases oxidizes quickly in NO_2, which in turn plays a fundamental role in the formation of tropospheric O_3, also contributing to eutrophication of water courses and acid rain [75].

During coal combustion processes, less than 5% of the total production of NO_x refers to NO_2 and nitrous acid (N_2O). This small percentage is because there is low oxygen availability and short residence times in high temperature coal flames, leading to the formation of only a small percentage of NO_2. In relation to the NO_x, there may still be formation in the initial phase of the flame, by the reaction in gas phase, presented by the following reactions:

$$O\cdot + N_2 \leftrightarrow N_2O \quad (3)$$

$$NH\cdot + NO \leftrightarrow N_2O + H\cdot \quad (4)$$

$$NCO\cdot + NO \leftrightarrow N_2O + CO \quad (5)$$

Throughout the cycle of nitrogen, several different chemical forms arise, presenting their own characteristics, behavior and effects for health and the environment. Figure 9 presents the arrangements and transformations of the various compounds present in the biochemical cycle of the nitrogen [76].

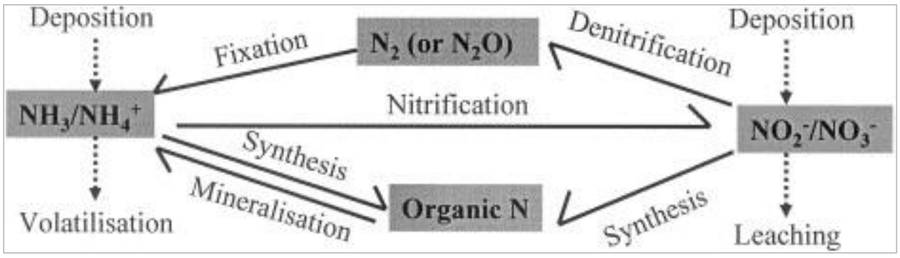


Figure 9 - Processes involved in the nitrogen cycle[76]

The largest source of NO_x emissions is combustion and coal, followed by the combustion of oil, ending with the burning of gas, which is the least emitting source of the three [74].
Guidelines from WHO and EU Air Quality Directive, dictate the concentration limits for this pollutant as shown in Table 4.

Table 4 - Air-quality standards for NO_2 (and NO_X) as set out in the EU Ambient Air Quality Directive. [29]
** - Human health limit value; ** - Alert threshold; *** - Vegetation critical level*

Averaging Period	EU Air Quality Directive	WHO AQG
1 hour	200 µg/m^3 (not to be exceeded on more than 18 hours per year)*	200 µg/m^3
	400 µg/m^3**	
Calendar Year	40 µg/m^3*	40 µg/m^3
	30 µg/m^3***	-

According to recent studies, concentrations of NO_2 emitted by road traffic do not appear to be decreasing. In some cases, there has been an increase in concentration, pointing to the use of oxidation catalysts and particulate filters used in diesel vehicles as probable cause [77].
Being involved in the formation of photochemical smog and acid rain and associated with depletion of the O_3 layer, NO_x can also affect human health more directly [73]. Nitrogen dioxide, being the most toxic oxidant, is a strong oxidant, reacting at the biomolecular level directly or by the formation of free radicals, also inducing pulmonary damage [78]. Nitric oxide reaches the population with greater severity, when the prevalence of smoking is lower and domestic consumption of gas is higher. A proportional relationship is also found between the harmful effect of nitric acid and the proportion of elderly in the population [79].

4.3. OZONE

Ozone is an effect gas that studies with a crucial role the climatic changes. Being produced in the lower troposphere through photochemical processes, it has a considerably long residence time, lasting several days in the atmospheric boundary layer and several weeks in the troposphere.
Several studies have shown that O_3, even existing in large concentrations at certain sites, may have been formed in a different location and even far away, having been carried by the wind [80].

Normally, O_3 is not emitted by natural processes or anthropogenic sources. Instead, O_3 is formed by precursors [81]. O_3 precursors are NO_x, CO and VOCs.
These precursors have sources of emitters ranging from combustion of fossil fuels and motor vehicles (NO_x and CO) to transportation, emission of organic solvents and natural emissions (VOCs) [82].

NO_x and CO are the largest emissions of fossil fuels and motor vehicles. Ozone formation processes differ depending on the available precursors and their concentrations.
In Figure 10, the O_3 formation process is shown by the combination of oxygen with atomic oxygen, previously formed by the photolysis of NO_2 (a), and by the formation of O_3 when VOC's are present, since different compounds have different O_3 formation rates [82].

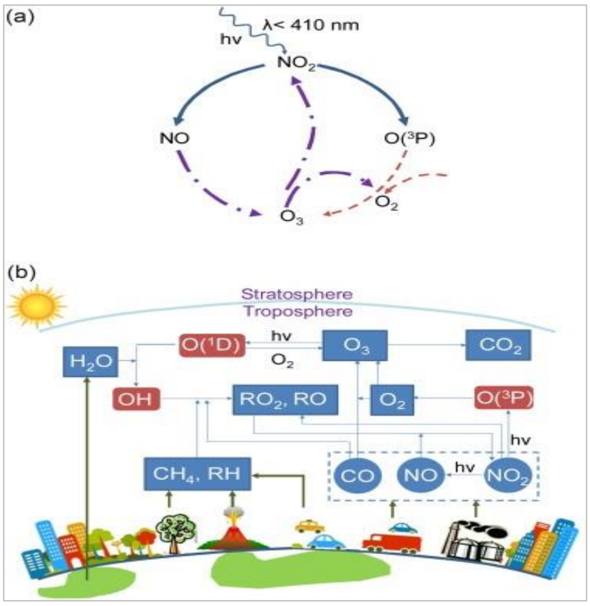


Figure 10 - Diagram of O_3 formation: (a) O_3 equilibrium without the presence of VOCs; (b) O_3 generation in the troposphere

The following equations show the relationship between the O_3 concentration and the concentrations of the precursors, given that RH represents a generic VOC [82].

$$CO \xrightarrow{NO_x} CO_2 + O_3 \tag{6}$$

$$CH_4 \xrightarrow{NO_x} CO_2 + O_3 + OH \cdot \tag{7}$$

$$RH \xrightarrow{NO_x} R'CHO + O_3 \quad (8)$$

Ozone can cause pulmonary dysfunction, induction and exacerbation of asthma, premature mortality and factors related to diabetic death [83]. Some studies [84] also reveal O_3 intervention in systemic metabolic adverse responses through the sympathetic nervous system and in systemic responses related to adverse health effects [83].

The concentrations of this gas are proportional to the concentrations of its precursors, such as NO_x, methane and nonmetallic volatile organic compounds. Table 5 presents the limit values provided by EU ambient Air Quality Directive and WHO for O_3.

Table 5 - Air-quality standards for O_3 as defined in the EU Ambient Air Quality Directive and WHO. [20] (- Information threshold; ** - Alert threshold)*

Pollutant	Averaging Period	EU Air Quality Directive	WHO Guidelines
Ozone	8-hour	120 µg/m³	100 µg/m³
	1-hour	180 µg/m³*	-
		240 µg/m³**	-

4.4. SULFUR DIOXIDE

With the growing energy needs of the population, the development of power generation plants has increased, increasing with it, all harmful pollutants emitted from generation processes, such as SO_2 [85]. As one of the most used air quality indicators, because it represents the effects of industrialization, SO_2 stands out from other pollutants when the lower atmospheric layer is approached.

In urban zoos, SO_2 comes mainly from the burning of fossil fuels (accounting for about 70% of the total emissions of this pollutant), industrial facilities (about 20%) and natural sources (about 10%) [85]. Having the worst effects on human health, SO_2 reacts with atmospheric compounds, forming small particles. Subsequently, these particles penetrate the respiratory system and cause respiratory complications, even causing cancer [85].
Exposure to high concentrations of SO_2 enhances health effects such as mortality from pulmonary and cardiovascular complications, non-accidental mortality, and cardiac hospitalization in the elderly [86].

Most studies related to the effects of SO_2 on human health fall within the mortality parameter, with few conclusions about the years of life lost (YLLs) [87].

Figure 11 graphically shows, in a shortened way, the parameters to be taken into account for the use of pollutant concentrations to determine quality of life indicators.

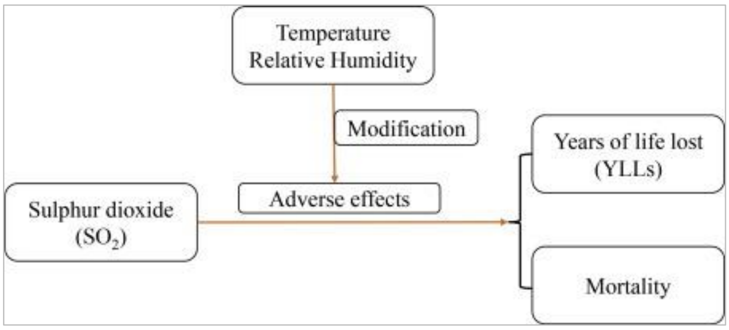


Figure 11 - Graphical representation of the conditions for the use of SO_2 as an indicator of air quality [87]

At the environmental level, SO_2 plays an important role in the formation of acid rain, which adversely affects natural systems, agriculture and building materials, as shown in the Figure 12 [88].

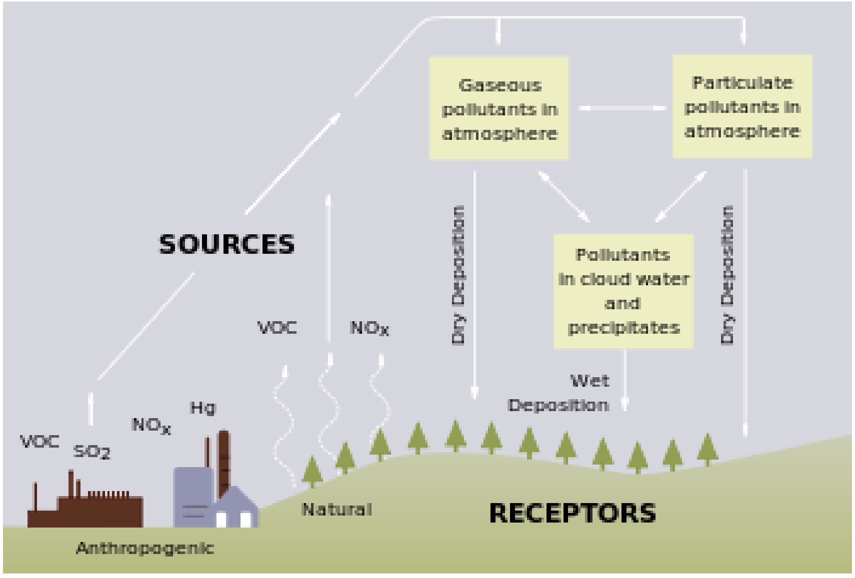


Figure 12 - Processes involved in acid deposition (source: Wikipedia) [89]

Sulfur dioxide is the primary cause of acid precipitation, The sulfate aerosol particles formed as a consequence of these emissions impair visibility and affect human health [88]. For this, the SO_2 forms a very acidic aqueous solution, explained by the equations: [85]

$$SO_2 + \frac{1}{2}O_2 \rightarrow SO_3 \quad (9)$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad (10)$$

The sulfate aerosol particles formed as a consequence of these emissions impair visibility and affect human health [88]. Studies show [90] that a percentage of people suffer abnormalities in the respiratory system after short periods of time of exposure to, thus presenting a rather high limit value, as shown in the Table 6.[34].

Table 6 - Guideline values for SO_2 emissions.
(- Human Health Limit Value; ** - Alert threshold; *** - Vegetation Critical Level)[20]*

Pollutant	Averaging Period	EU Air Quality Directive	WHO Guidelines
Sulfur Dioxide	10 minutes	-	500 µg/m^3
	1-hour	350 µg/m^3 (not exceed 24-hour exposure per year)*	-
		500**	-
	Calendar Year	20 ***	-

4.5. CARBON MONOXIDE

Carbon monoxide is a colorless, non-irritable, odorless gas from incomplete combustion and burning of fossil fuels such as coal, oil and gas [91, 92]. CO is a useful indicator of air mass transport monitoring at various scales because it presents an average tropospheric life of about two months [93].

This gas has two types of formation origin, endo and exogenous. Being the main source of CO, the endogenous source is based on oxidative degradation of heme (prosthetic group of hemoproteins such as hemoglobin and myoglobin, and are fundamental in the processes of reversible connections of oxygen and transport) [94], catalyzed by heme oxygenase. The second type of source of monoxide, an exogenous source, includes exhaust from combustion vehicles, gas heating, wood stoves, incomplete combustion and burning of fossil fuels, power stations and mining industry.

Additionally, a source of CO created by pathological conditions such as intestinal bacteriological activity, photo oxidation of organic compounds and lipid peroxidation can be considered [95].

The most common sources of poisoning by CO include oil or gas furnaces, gas or oil heating installations, kerosene heaters, combustion equipment, automotive exhaust and household fires. After inhalation, CO diffusion occurs in the blood, binding to hemoglobin, thus creating a complex called carboxyhemoglobin (COHb). This binding occurs quickly and easily because CO has a hemoglobin affinity higher than that of oxygen by about 250 times, with values higher than fetal hemoglobin (HbF) relative to adult hemoglobin (HbA) [91]. In colder climates or seasons, cases of CO poisoning are commonly associated with exposure to emissions from fire, exhaust, heating and tobacco smoke [96].

Carbon monoxide poisoning intoxicates more particularly the brain and heart, i.e. organs with a higher oxygen demand, the latter being affected after severe poisoning, resulting in myocardial infarction, cardiac arrhythmias, hypotension, cardiac arrest and death [97].

Other occurrences of CO poisoning include the inhalation of methylene chloride vapors, volatile material present in degreasers and solvents. Even without significant systemic effects, significant burns can occur. In very specific cases, and with methylene chloride ingestion, CO poisoning may occur as the liver metabolizes part of the inhaled methylene chloride to CO, storing a small part of the tissue that results in high levels of CO in the body, much higher than those recorded by direct ingestion of CO [98].

With the inhalation of CO, various harmful effects are triggered in the body, resulting in severe organ failure. The following diagram (Figure 13) highlights the main effects triggered in the respiratory system by the inhalation of fumes.

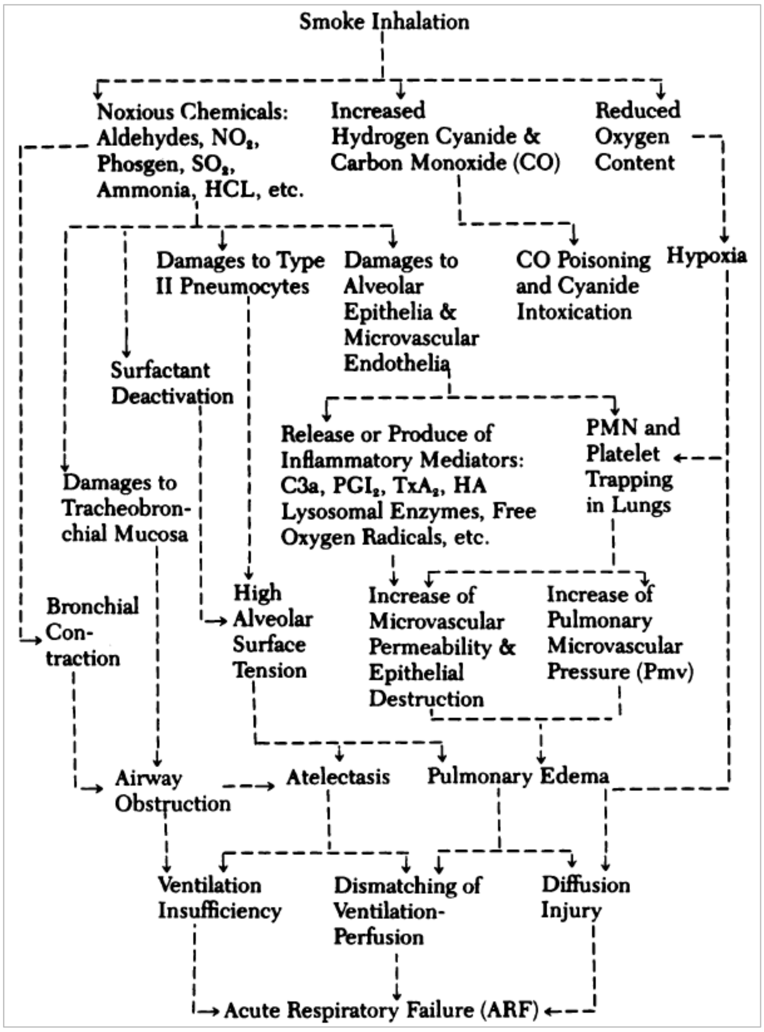


Figure 13 - Diagram of the sequence of events triggered by smoke inhalation [99]

Because CO rapidly oxidizes to carbon dioxide, existing guidelines for this pollutant focus on indoor air, since for outdoor air the attention will be drawn to carbon dioxide [75, 100, 101].

4.6. METHANE

Methane is a colorless, flammable hydrocarbon. Being the most abundant VOC in the atmosphere and presenting a very varied annual growth rate from year to year, it is a gas created by the anaerobic decomposition of organic components [102].

Featuring a tropospheric concentration of 1.8 ppm, CH_4 is the third most important greenhouse gas followed by water vapor and carbon dioxide [103].

Since 2006, with the conditions of temperature and humidity caused by a period of La Niña, tropical areas have been the main sources of methane emission from wetlands. Exploitation and energy production, landfill, animal agri-business, rice farming and biomass burning are some of the main anthropogenic sources of CH_4, Forest fires, wetlands, wildlife and geological sources are some of the main natural sources of Methane [103].
Since the 18th century, human activities involving agriculture, waste disposal and extraction of fossil fuels have greatly multiplied the emissions of CH_4, a potent greenhouse gas because of its high global warming potential and reduced atmospheric life span [104].

Given that the global warming potential of CH_4 is 25 to 28 times greater than that of CO_2 over a period of 100 years, CH_4 is an important greenhouse gas (GHG) [105, 106].

Since the beginning of the industrial revolution, mean annual concentrations of pollutants have been increasing, as in the case of atmospheric CH_4, which increased from 0.7 ppm to 1.8 ppm, reaching the highest value in the last 800,000 years [105].

Methane is also the result of microbial fermentation processes that occur in the digestion of herbivorous mammalian foods (according to some studies, about one-third of global CH_4 emissions come from enteric fermentation of ruminants) [104].

As shown in Figure 14, most of the sources emitting CH_4 are anthropogenic, with the largest contributors being the exploitation of fossil fuels and livestock.

The organic fraction of wastewater decomposes anaerobically through successive transformations of the complex into the simple, initiating the hydrolysis of complex particles to simpler polymers (e.g. proteins, carbohydrates and lipids), being hydrolyzed again, producing monomers (e.g. Amino acids, sugars) and high molecular weight fatty acids [107].

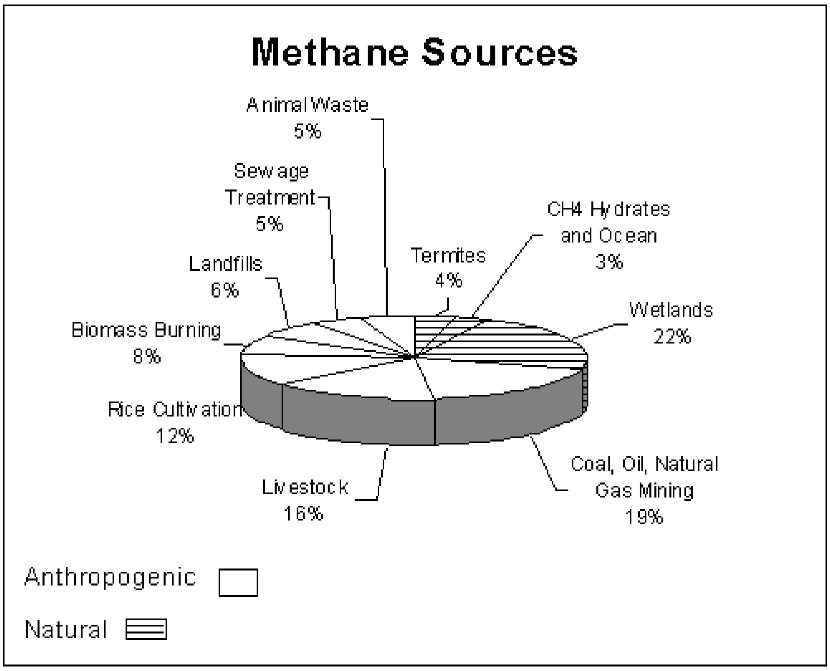


Figure 14 - Natural and Anthropogenic Sources of CH_4 [108]

Amino acids and sugars are transformed into by-products such as propionic, butyric and other volatile acids or are directly fermented into acetic acid. High molecular weight fatty acids are oxidized to by-products and hydrogen [107].

These processes are graphically reprinted in Figure 15, emphasizing the division of the process into 3 main steps.

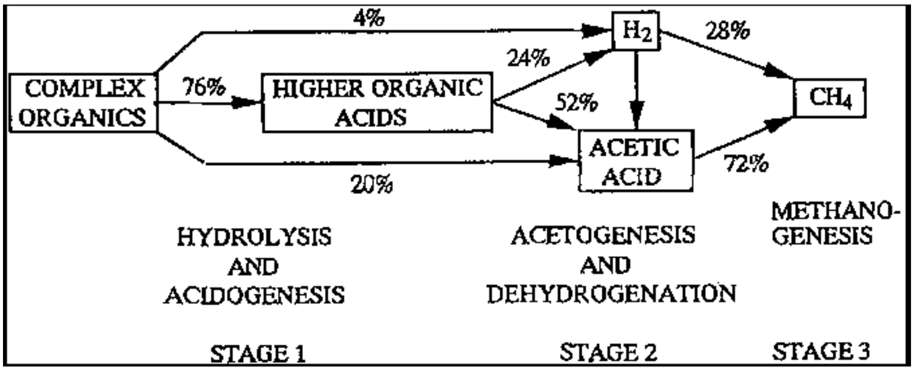


Figure 15 - Stages of methane fermentation Source: McCarty, P.L., (1982)

The production of CH_4 and CO_2 occurs through the cleavage of the acetate, and may also result from the reduction of CO_2 with hydrogen [107, 109].

In order to determine the extent of CH_4 production, the degradable organic fraction contained in wastewater is an important factor, and is generally expressed in terms of oxygen chemical demand (COD) and biochemical oxygen demand (BOD). The yield of methane has a proportional relationship with COD and BOD.

Environmental factors also play a determining role in the production of CH_4, such as temperature, wastewater treatment, pH, retention time, and competition between methanogens and sulfate reducing bacteria.

Although not a conditioning factor in the decomposition of the organic fraction, the moisture content is an essential factor in any environment.

In the case of anaerobic processes, the presence of oxygen completely inhibits CH_4 production [107].

4.7. AMMONIA

As a chemical compound consisting of a nitrogen atom and three hydrogen atoms, distributed in a tetrahedron geometry, NH_3 is the third most abundant compound in atmospheres, after nitrous oxide and atmospheric nitrogen [110].

In order to satisfy the growing and constant demand of food by the world population, NH_3 plays a fundamental role as fertilizer, estimating that in its absence in fertilizers, between 30% and 50% of the world's population would die of hunger. In 2014, even with constant demand, world production of NH_3 exceeded 140 million tons [110].

Even though the concentration of atmospheric NH_3 is highly variable and depends on the distance from the pollutant source, the typical continental concentration is between 0.1 and 10 ppb. The formation of nitrate in particles comes essentially from the reaction of NH_3 with nitric acid in the gaseous state, represented in equations 11 and 12.

$$NH_3(g) + HNO_3(g) \rightarrow NH_4NO_3(s) \quad (11)$$

Instead of nitric acid, NH_3, when reacted with sulfuric acid can also produce particles:

$$2NH_3(g) + H_2SO_4(g) \rightarrow (NH_4)_2SO_4(s) \quad (12)$$

Considering the natural cycle of nitrogen, shown in Figure 16, changes in the cycle reveal great impacts on air pollution, since at different stages, the various forms of ammonia participate in different processes of formation of other pollutants [111].

In order to compensate for the slow kinetics of the reaction in question, elevated temperatures are required. Although there is no NH_3 synthesis technology yet developed enough to perform similarly to the Haber Bosch process, the need to use other processes is crucial because of the enormous energy consumption required for this process [111].

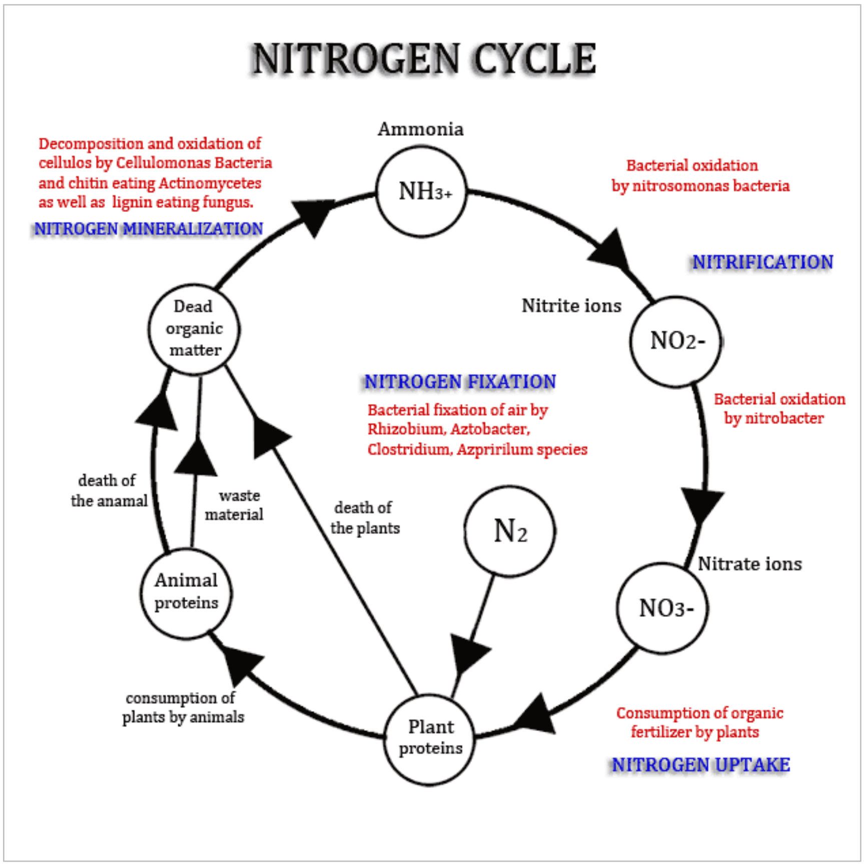


Figure 16 - Graphic representation of Nitrogen Cycle [112]

In the atmosphere, the main reactive form of nitrogen is NH_3, which comes from the application of fertilizers to the soil, animal excretion, the application of manure, the industry where the ammonia is applied and motor vehicle exhaust by-products [113].

Although there is always a high degree of uncertainty in the determination of NH_3 emissions into the atmosphere, the primary sector represents the largest NH_3 emitter [114].
The uncertainty arises from the fact that there are a limited number of studies aimed at quantifying emission factors and activity in the primary sector and that it is a very diverse sector with specific characteristics in each situation [114].

The life time in the atmosphere of this gas is limited to a few hours, since it is removed by dry and humid deposition. After deposition, nitrogen reduction products contribute to the acidification and eutrophication of both terrestrial and aquatic ecosystems, thereby reducing biodiversity [113].
At the industrial level, ammonia production is almost fully implemented through the Haber Bosch process, reacting the high purity flows of N_2 and H_2 at high temperatures (300-500 °C) and pressures (200-300 atm) as in the equation below [110].

$$3H_2 + N_2 \rightarrow 2NH_3 \quad (13)$$

4.8. AIR POLLUTION AND HEALTH EFFECTS ASSESSMENT

In the last decades, studies confirm the contribution of air pollution from the outside air contributes to morbidity and mortality, considering some effects related to short-term exposure [115]. The Global Burden of Disease study has described the worldwide impact of air pollution with as many as 3.1 million of 52.8 million all-cause and all-age deaths being attributable to ambient air pollution in the year 2010 [116]. Further, air pollution accounts for 3.1% of global disability-adjusted life years, an index that measures the time spent in states of reduced health. Air pollution is an important stimulus for the development and exacerbation of respiratory diseases, such as asthma, chronic obstructive pulmonary disease, and lung cancer, as well as, a substantial impact on cardiovascular disease. The role of statistics has always played an important role in the analysis of society, from demography, economics, politics and even public health.
The Table 7 shows the list of risk factors taken in consideration.

Table 7 - Core Health Indicators - Risk Factors List (adapted) [65]

Risk Factors	
Nutrition	**Environmental Risk Factors**
Exclusive breastfeeding rate 0-5 months of age	Population using safely managed drinking-water services
Early initiation of breastfeeding	Population using safely managed sanitation services
Incidence of low birth weight among new-borns	Population using modern fuels for cooking/heating/lighting
Children under 5 years who are stunted	Air pollution level in cities
Children under 5 years who are wasted	**Noncommunicable diseases**
Anaemia prevalence in children	Total alcohol per capita (age 15+ years) consumption
Anaemia prevalence in women of reproductive age	Tobacco use among persons aged 18+ years
Infections	Children aged under 5 years who are overweight
Condom use at last sex with high-risk partner	Overweight and obesity in adults (Also: adolescents)
Injuries	Raised blood pressure among adults
Intimate partner violence prevalence	Raised blood glucose/diabetes among adults
	Salt intake
	Insufficient physical activity in adults (Also: adolescents)

In 2013, the International Agency for Research on Cancer classified outdoor air pollution as carcinogenic to humans [32].

The contributory causal relationship of outdoor air pollution to morbidity and mortality rates is evidenced by epidemiological studies, finding consistent associations between air pollution and various health effects such as; respiratory symptoms, reduced lung function, chronic bronchitis, and mortality [115].

Regarding public health, there has always been a concern to evaluate the human well-being and health of the population, using statistical parameters that, with a posterior analysis, making possible the identification of possible anomalies and problems and the design of corrective and preventive strategies regarding well-being and public health of the population. In order to establish goals and strategies for improvement in the field of public health, the justified determination of health indicators becomes a crucial step for future analysis, as well as risk factors, that the WHO take in consideration for decision-making studies.

In an overall analysis, the health indicators can be divided into two main groups: mortality and morbidity. In a regional perspective, many of the indicators used to evaluate a country are not used due to the inability to analyze with this degree of specificity, remaining only more general indicators.

In the field of mortality, there are several indicators with sub-indicators, such as: Life expectancy; Causes of mortality; Maternal and infant mortality, with the sub-indicators like: infant, neonatal, perinatal and maternal mortality; and potential years of life lost. In the morbidity part, statistics allow us to determine indicators such as: Perceived health status, by age and gender, and by socio-economic status; Low birthweight, decayed-missing-filled teeth at age 12; Communicable diseases, like AIDS, incidence of pertussis, measles and hepatitis B; cancer; Injuries in road traffic accidents; and absence from work due to illness (self-reported and compensated) [101].

Global health indicators can be divided into several groups, considering the nature of each indicator, as presented in Table 8.

Table 8 - 100 Core Health Indicators - Health Status (adapted)[65]

Health Status	
Mortality by age and sex	**Fertility**
Life expectancy at birth	Adolescent fertility rate
Adult mortality rate between 15 and 60 years of age	Total Fertility rate
Under-five mortality rate	**Morbidity**
Infant mortality rate	New cases of vaccine-preventable diseases
Neonatal mortality rate	New cases of IHR-notifiable diseases and other notifiable diseases
Stillbirth rate	HIV incidence rate
Mortality by cause	HIV prevalence rate
Maternal mortality ratio	Hepatitis B surface infections (STIs)incidence rate
TB mortality rate	TB incidence rate
AIDS-related mortality rate	TB prevalence rate
Mortality between 30 and 70 years of age from cardiovascular diseases, cancer, diabetes or chronic respiratory diseases	Malaria parasite prevalence among children aged 6-59 months
Suicide rate	Malaria incidence rate
Mortality rate from road traffic injuries	Cancer incidence by type of cancer
Malaria mortality rate	

Several studies [117-124] have refer harmful effects associated with exposure to high concentrations of airborne pollutants, but similar effects are observed at lower concentrations but longer exposure. Airway irritation, followed by bronchoconstriction and dyspnea, are usually associated with exposure to high concentrations of SO_2, NO_x and some heavy metals such as arsenic, nickel and vanadium. PM may also penetrate the alveolar epithelium and O_3 may also potentiate lung inflammation or aggravate preexisting problems.[67] Figure 17 sum up the main heath impacts of air pollution.

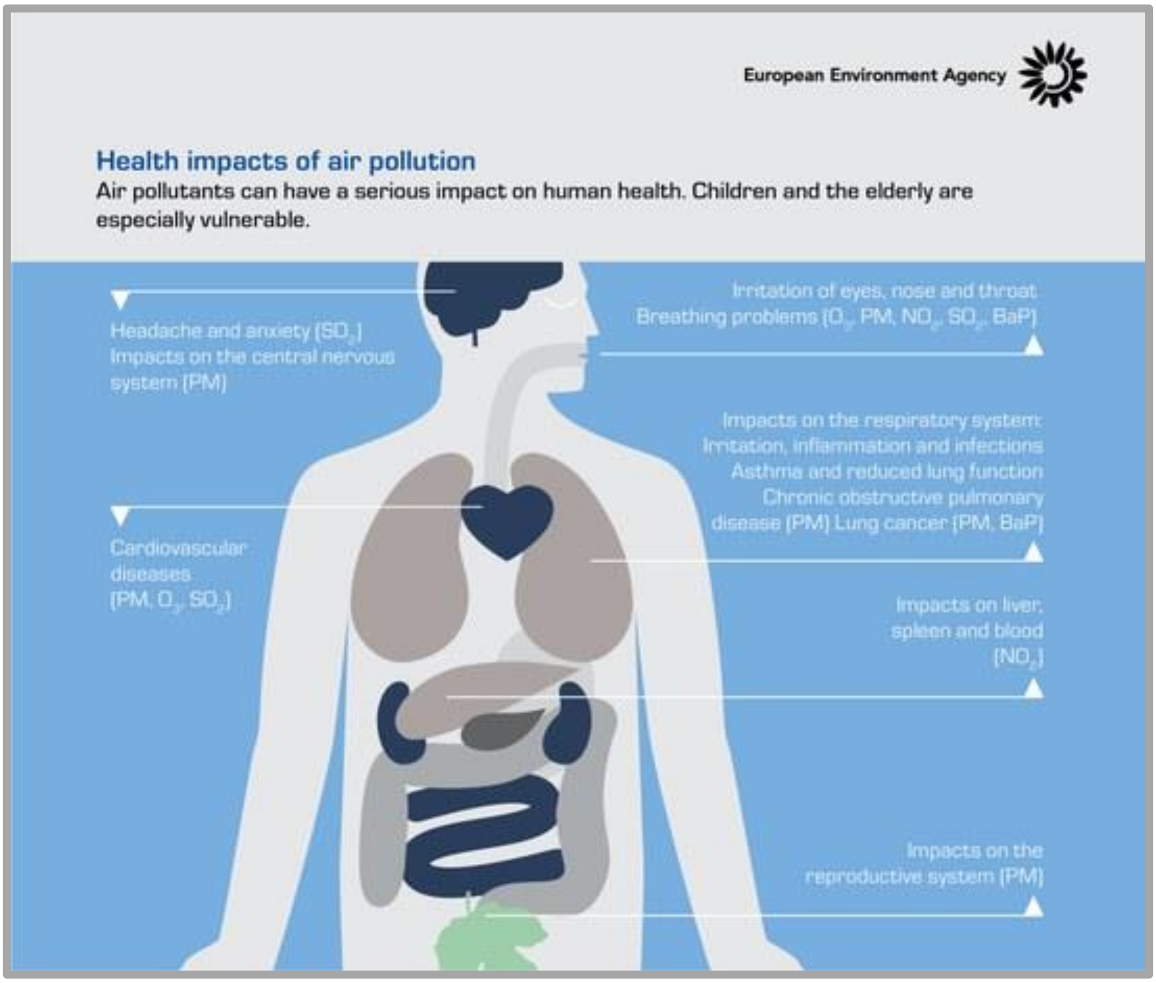


Figure 17 - Health impacts of air pollution. Source: European Environment Agency, 2017 [125]

By reducing its oxygen transfer ability, CO binds to hemoglobin by modifying its composition. With reduced oxygen availability, several organs face problems, especially oxygen-demanding organs such as the brain and heart. Thus, with oxygen deficiency, symptoms such as reduced intercourse, slow reflexes, and confusion may appear.
Particles can induce systemic and pulmonary inflammation. By encouraging lung irritations and changes in the blood coagulation level, air pollutants can block blood vessels, leading to cardiac implications such as angina and myocardial infractions.[67]

Heavy metals, such as lead, mercury and arsenic, and dioxins, are primarily responsible for harmful effects on the nervous system. After exposure to heavy metals, a relationship was noted between neurotoxicity and neuropathies, which led to a cluster of symptoms such as memory disorders, sleep disturbances, anger, fatigue, hand tremors, blurred vision and slurred speech.

Some of the main effects associated with heavy metal poisoning can be seen in Table 9.

Table 9 - Symptoms from Heavy Metals Poisoning [126]

Element	Acute Exposure (usually a day or less)	Chronic Exposure (months or years)
Lead	Encephalopathy (Brain Dysfunction) Nausea Vomiting	Anaemia Encephalopathy Foot drop/wrist drop (palsy) Nephropathy (kidney disease)
Cadmium	Pneumonitis (lung inflammation)	Lung cancer Osteomalacia (softening of bones) Proteinuria (excess protein in urine; possible kidney damage)
Mercury	Vomiting Diarrhea Fever	Stomatitis (inflammation of gums) Nausea, tremor Nephrotic syndrome (kidney disorder) Neurasthenia (neurotic disorder) Paralgesia (metallic taste) Pink Disease (pain and pink discoloration of hands and feet)
Arsenic	Nausea, vomiting, diarrhea Encephalopathy Multi-organ effects Arrhythmia Painful neuropathy	Hypopigmentation/Hyperkeratosis Cancer Diabetes

These heavy metals can also induce serious deleterious effects in the excretory system, particularly in the kidneys, causing initial tubular dysfunctions. These dysfunctions may be confirmed by an increase in the indices of excretion of low molecular weight proteins, accompanied by the decrease in the glomerular filtration rate.[67]

Environmental assessments play a crucial role in combating environmental problems and sustainable development both locally and globally, and studies are selected to collect selected knowledge for the purpose of useful publication for the delineation of strategies and decision-making [127].

Through statistical data, various assessments can be made according to the required focus, such as health impact assessments, which help to develop development policies in the area, taking into account the positive and negative implications for public health [128].

In the conjecture of sustainable development, several impact assessments have included in their study, environmental impacts, parameters and indicators related to quality of life associated with air pollution. These measures are due to the knowledge of the effects of environmental changes, caused directly and indirectly on the health of the people, and to the fact that the entities that make the decisions, have objectives that cover a set of themes, not only public health but also quality of Life and development sustainability.

Cost-benefit and multicriteria analyzes are presented as valid tools for evaluating health and environment outcomes, but are less used by health care decision makers because of their complexity and the need for a large amount of information [129].

The Driving Force-Pressure-State-Exposure-Effect-Action (DPSEEA), shown in Figure 18, is considered one of the most suitable one for developing environmental health indicators for climate change and health. The framework looks at the triggering relationships between driving forces, environmental pressures, exposure and effect [130].

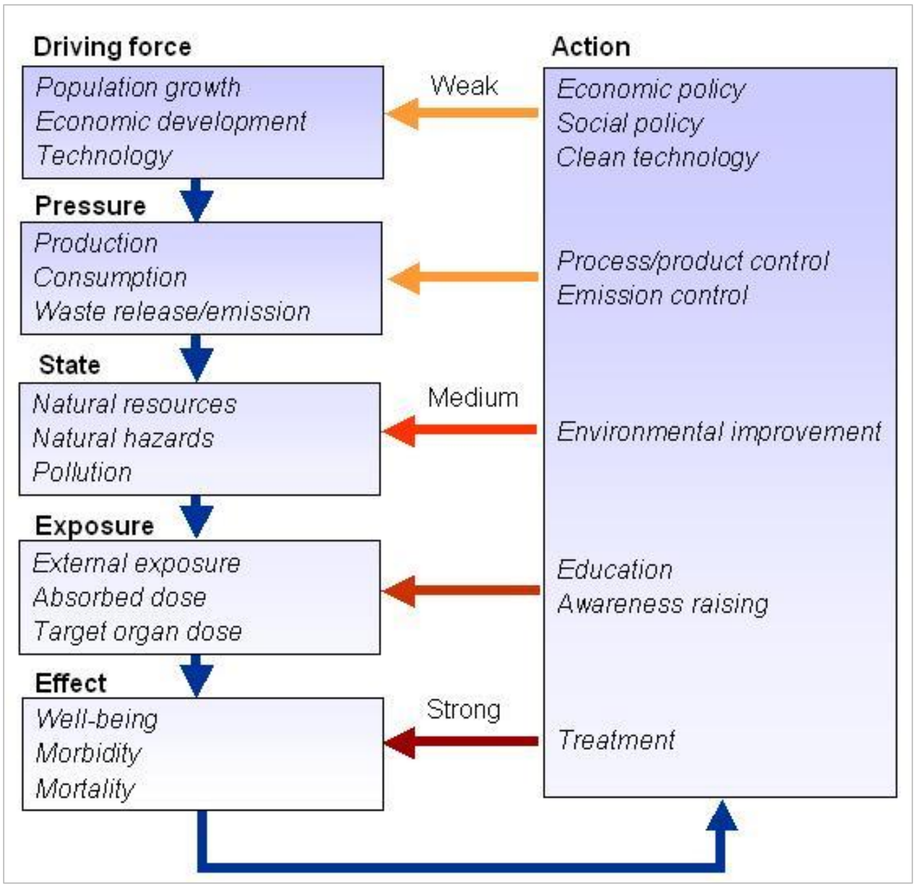


Figure 18 - Framework for the development and identification of environmental health indicators [131]

4.9. EMISSIONS REDUCTION SCENARIOS

With the aim of reducing global warming and consumption of fossil fuels and improving air quality, the implementation of mitigating policies and measures becomes a demanding issue for European climate and energy policies [132] (Figure 19).

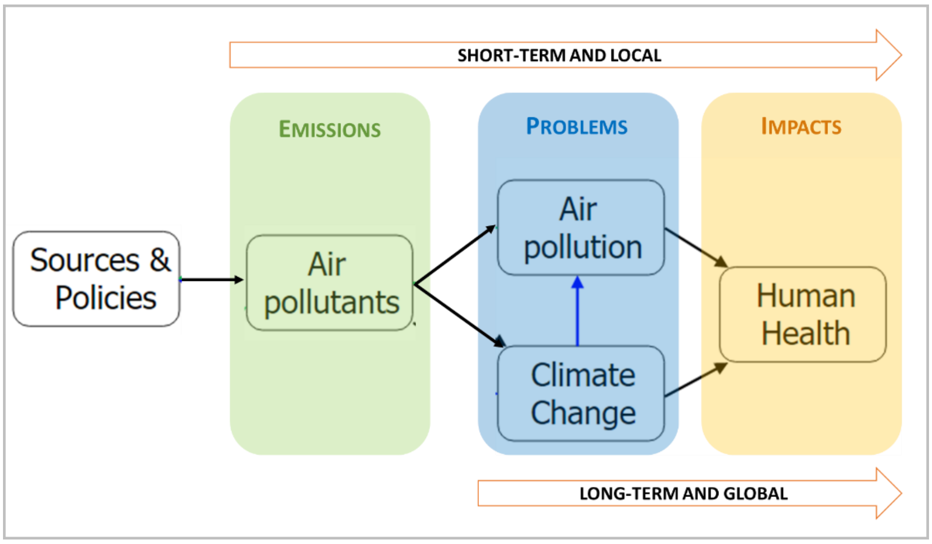


Figure 19 - Co-benefits of mitigating air pollutants emissions for future air quality and human health. (adapted from West et al. NCC 2013) [133]

Given that around 25% of total emissions of greenhouse gas emissions come from road transport, the focus of European policies is on the use of motor vehicles, and a number of measures have now been implemented at European level and various outlined strategies [132].

In the early 1990s, several long-term emission reduction scenarios were created (as shown in the Figure 20) by the Intergovernmental Plan on Climate Change (IPCC), these scenarios being heavily used to analyze impacts and options for mitigating climate change [134].

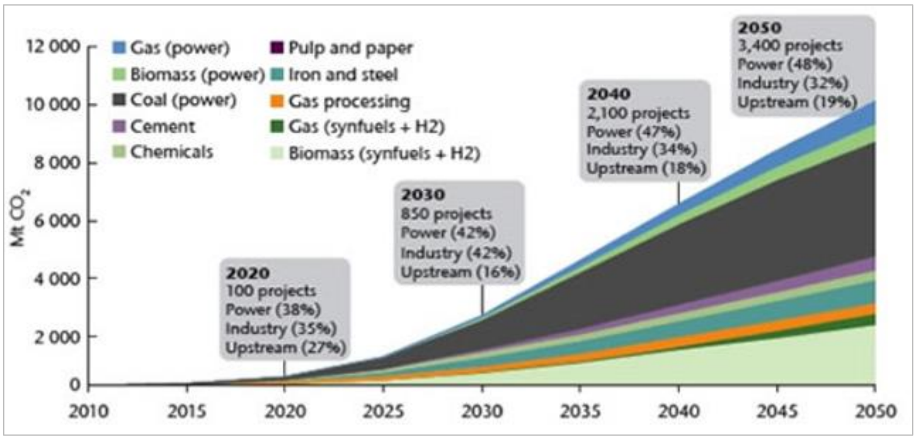


Figure 20 - Example of a global emissions reduction scenarios [135]

In 1996, the IPCC Plenary decided on a new set of scenarios that came to encompass issues such as carbon intensity and energy supply, sulfur emissions and disagreement between developed and developing countries. This decision was made due to an assessment of scenarios in 1995, focusing on the understanding of driving forces of emissions and methodologies [134].

Considering scenarios as visualizations of alternative situations of the future on some limitations and conditions, they are an important tool to analyze the influence of the driving forces on the future results of associated emissions and uncertainties.

Analyzing climate change, the scenarios also imply climate modeling, impact assessment and mitigation [134].

Thus, a range of scenarios has been developed to analyze the range of driving forces and emissions to reach the current understanding of the associated uncertainties. Any scenario encompasses subjective components and its analysis is open to varied interpretations [134].

Normally, all scenarios have the premise that in the future, the gross world product is greater than 10 to 26 times per 100 years assuming then that the scenarios in the future will be richer than at present [134].

In 2001 the "Clean Air for Europe" program was set up to develop policies supporting the development of the Thematic Strategy on Air Pollution in the Sixth Environmental Action Program, which was used by the European Parliament.
This program, through the use of scenarios and projections, laid the foundations for an integrated policy for the protection of human health and the environment against the negative effects of atmospheric pollution, taking into account restrictive economic issues, concentrating mainly on O_3 and PM [136].

In the cost-effectiveness analysis of the policies presented in the legislative review on air quality, the Clean Air for Europe program prepares a range of policy analysis tools, aggregated by validated database scientists.
In order to represent the situation of all Member States and economic sectors, several models are used, such as: integrated RAINS assessment model for air pollution and greenhouse gases [137]; PRIMES models for energy sectors in the European Union [138]; the TREMOVE model dedicated to transport [139]; and cost-benefit analysis of CAFE [140, 141].

The evaluation tools applied in the search for efficient metering packages aim to bring the situation of Europe closer to its environmental objectives [141].

4.10. LEGISLATION

4.10.1. EUROPEAN OVERVIEW

Since the 1970s, European policy concerns have focused on air pollution issues, focusing on developing and implementing appropriate instruments and measures to improve air quality. Within this theme, they focused on controlling emissions from mobile sources, optimizing fuels and promoting and integrating previously defined environmental obligations for the transportation and energy sectors [142].

The WHO has established the Air Quality Guidelines explaining basic information on pollutants and their harmful effects on health as well as information on pollutants collected by the EEA [142]. In December 2013, the European Commission adopted several air protection policies, including: the Clean Air for Europe program [143], bringing new air quality targets by 2030; National emission Ceilings guidelines for the main pollutants; and a project for a new directive to reduce emissions of pollutants from medium-sized combustion plants [142].

By the end of 2016, the new NEC directive entered into force, with a transposition planned until mid-2018. By the end of March 2019, 2020 and 2030, all EU Member States should draw up a National Program of Atmospheric Pollution Control [144].
In order to ensure compliance with the reduction measures for 2020 and 2030, EU Member States have to comply with the National Air Pollution Control Program and is one of the main governance measures implemented [145].

The legal provisions to be taken into account by the guidelines require Member States to draw up, adopt and implement the program [145]. These guidelines include:

- assess the susceptibility of national emission sources to changes in air quality at national level and in neighboring Member States;
- Address the need to reduce atmospheric pollution emissions in order to achieve the objectives outlined;
- Prioritize measures to reduce black carbon emissions when evaluating PM2.5 reduction;
- Ensure the consistency between plans and programs established under European or national legislation;
- Introduce compulsory and optional measures;
- Addressing the developed policy framework (political priorities and sharing of responsibilities);
- Selection of the available range of policies considered and their adoption to meet reduction targets (including how to ensure consistency with other policy issues);

- Reduction trends projected for the period 2020 to 2030 [145].

The implementation of the measures and guidelines can be described by several elements, such as:

- Zones and agglomerates, declared by the Member States, covering the whole territory, representing basic areas of evaluation and management [146].

- Assessment of air quality, which accentuates information on compliance with environmental standards and clarification of efforts to reduce air pollution, and is only possible through the monitoring, modeling and objective estimation of air quality (it complies with established evaluative requirements and additional assessments such as the allocation of sources, with special attention to points with high pollution) [146].

- Managing air pollution by reducing harmful effects of air pollution on health and the environment by creating the need for action at Community level (such as European regulatory implications on new vehicles). In some areas, it is crucial to devise ancillary measures for European legislative compliance, with the creation of programs and packages of pollution abatement and control measures [146].

- Public sharing of information not only on environmental requirements, but also on the assessment of concentrations of pollutants, but also on information on emission reduction programs and action plans. When in an emergency situation, certain specific actions are taken to alert and inform the public of the health hazards and the recommended response actions to mitigate the exposure [146].

4.10.2. PORTUGUESE OVERVIEW

In coordination with the Regional Coordination and Development Commissions (both within the territory of mainland Portugal and in the Regional Directorates for the Environment of the Autonomous Regions), institutional bodies such as the Ministry of Environment, Spatial Planning and Regional Development have been studying More and more on the subject of air quality [147].

All the information on this subject has been constantly reviewed in order to show all scientific advances and new empirical information, such as an experience gained in the Member States, ambient air quality and a cleaner air in Europe [147].

All the legal provisions of Directive 96/62/EC of September 27th, 2000/69/EC of November 16th and 2002/3/EC of February 12th (concerning pollutants: SO_2, NO_2, NOx, PM10, Pb, C_6H_6, CO and O_3), and Council Decision 97/101/EC of January 27th, are all incorporated in Directive 2008/50/EC of May 21st, which was subsequently transposed into the national legal order by Decree-Law

No.102/2010 of September 23rd, which also includes the fourth daughter Directive 2004/107/EC of December 15th concerning arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons [147].

This decree-law establishes the goals related to air quality in compliance with the norms, guidelines and programs of the WHO, with the objective of preserving the ambient air quality when presented a good value of index and improving it when it is not the situation [147].

Depending on their competencies, several actors are responsible for making decisions and implementing measures whenever air quality objectives are not met. In these decisions and measures, short-term action plans and air quality plans are integrated [147].

On air pollution, more precisely on the reduction of mortality and morbidity rates caused by exposure to pollutants, additional objectives of continuous improvement are adopted in the intervention plan directed to PM2.5 present in ambient air [147].

Portugal is equipped with complex monitoring stations and measurement networks with the aim of assessing air quality, mostly managed by regional coordination and development committees, whose concerns accentuate in the absence of harmful interactions with other networks and stations related to other Facilities and measurement procedures [148].

In Portugal there are several entities for the control, evaluation and monitoring of indicators and parameters related to air quality, although each of these entities plays different roles and have different but convergent objectives [148].

The APA, being a national authority, focuses on: ensuring and coordinating all the necessary procedures for the implementation of Decree-Law no. 102/2010 of September 23rd, in cooperation with several entities in the process of management and evaluation of ambient air quality in Portugal; Ascertain methodologies for assessing air quality; Ensure compliance with mandatory quality assessment opinions in Portugal in the monitoring networks; Provide and guide the exchange of information with the European Commission related to data dissemination, air quality management and evaluation; Provide information transmitted to the European Commission, as well as information related to the entities responsible for implementing Decree-Law no. 102/2010 of September 23rd; Interact with Member States and with the European Commission for the purpose of implementing the aforementioned decree; And serve as national reference laboratory [148].

The Portuguese environment agency, as the national reference laboratory, also plays a key role in air quality, with the following objectives: ratify measurement systems, specifically laboratory infrastructures, methods and equipment; Publish guidelines that ensure high accuracy measurements; and to systematize nationally the quality assurance programs developed by the European Commission at Community level.

The Regional Development Coordination and Development Commissions, taking into account their territorial jurisdiction, have the responsibility of: managing and accessing ambient air quality; Ensure the accuracy and accuracy of pollutant collection and measurement; And ensure not only the sharing of environmental quality information, but also to report to the authorities on exceedance situations and to alert local authorities [148].

4.11. SITUATION IN PORTUGAL

In Portugal, with respect to air quality, the evaluation is strongly compatible with the legislation provided by the European community, and all the data and results of the studies are published in online databases. These databases make use of a simple and intuitive system, using an air quality index (AQI) [149].

The air quality index, implemented in 2001 and using the area unit as the denominator, uses the average of the in situ values of the pollutants under study, including at least the following pollutants: inhalable PM (such as PM10), O_3 and NO_2 [149].

The poor results presented by the air quality index are mainly due to PM and O_3 [149].

In the places where air quality values were presented below normal or acceptable, from the quality index, an analysis was performed to verify the evolutionary trends expected for Portugal, resulting in a graph as shown in Figure 22.

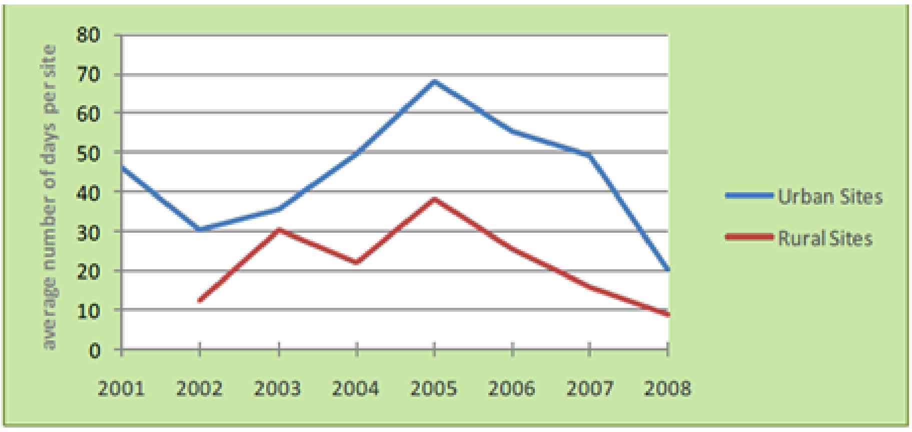


Figure 21 - Number of days on which air quality is low or bad (2001-2008) [149]

With the constant development of technologies for monitoring and controlling gaseous emissions of air pollutants into ambient air, more accurate and frequent statistical analysis has been possible, allowing an overview of the national panorama over the years, as seen in Figure 23.

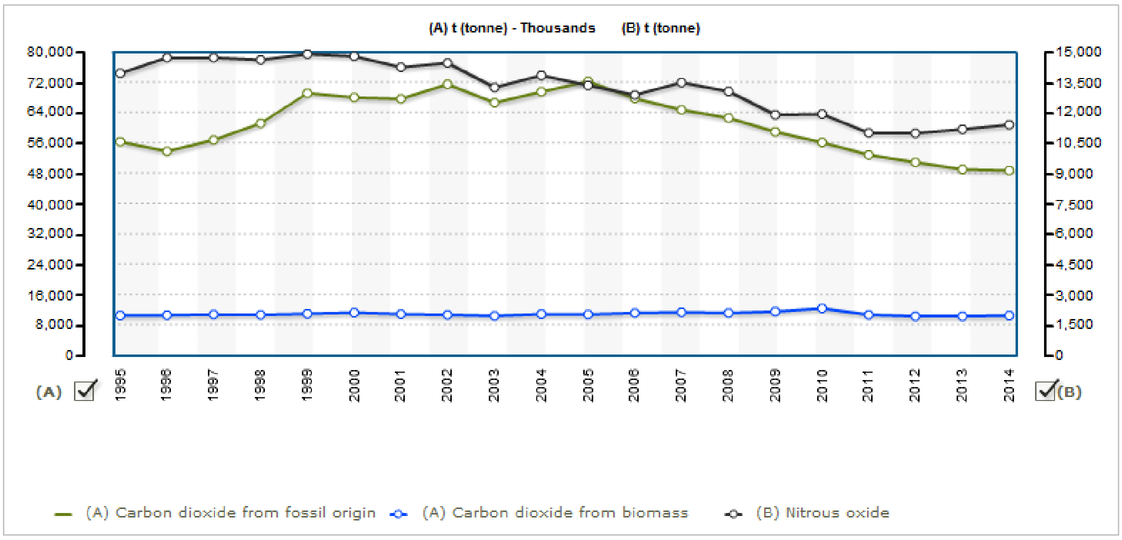


Figure 22 - Schematization of the evolutionary trend of gaseous emissions in Portugal [150]

In general, Portugal has shown a tendency to reduce emissions of atmospheric pollutants, since gradually decreasing emission values have been recorded. Several studies have been devoted to the temporal evolution of the harmful effects resulting from the emission of certain pollutants, as shown in Figure 24, where the progression of acidification indices in the country is explicit.

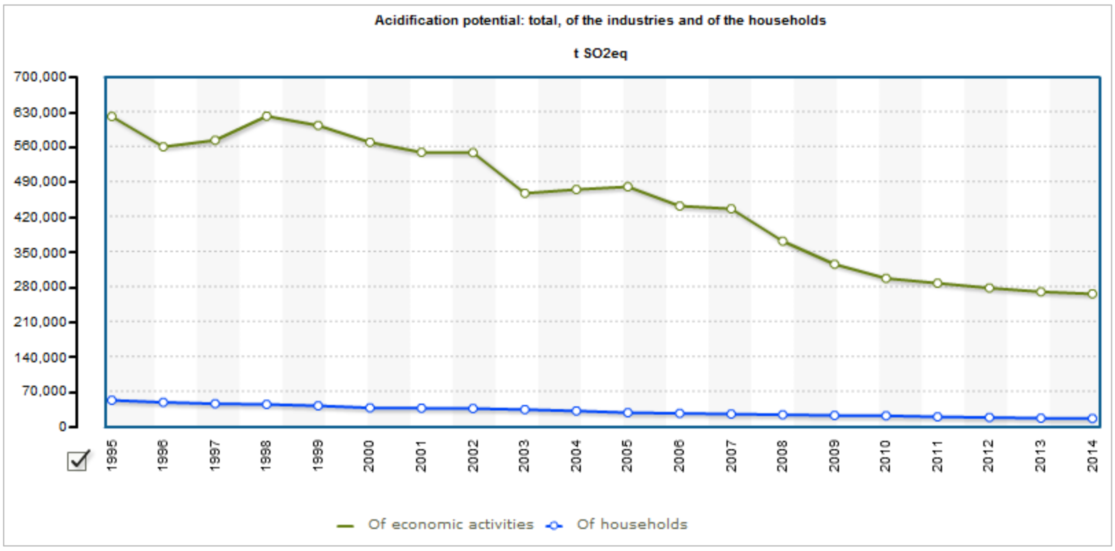


Figure 23 - Evolutionary progression of acidification potential in Portugal [151]

The analysis not only made it possible to quantify the annual values of the gaseous emissions but also allowed to relate the pollutant data to other data from different sectors or sectors, like the economic sector.

The next image (Figure 24) represents all the combined efforts of the monitoring and control entities of all outdoor air quality indicators, added in a graph that indicates roughly the state of all areas of Portugal in relation to air quality indices.

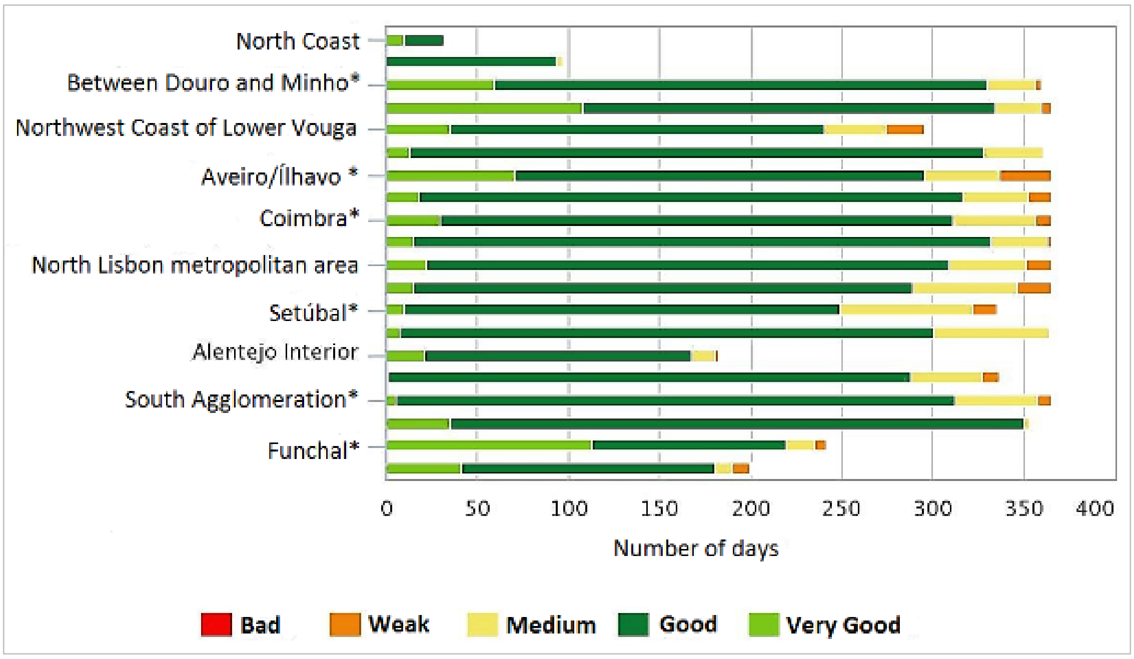


*Figure 24 - Air quality index for Portugal for 2015 (Adapted) *- Agglomeration*

5. CASE STUDY: RESULTS AND DISCUSSION

5.1. EMISSION OUTLOOK

The inventory and analysis of atmospheric pollutant emissions play a fundamental role in the identification of air pollutant emission sources, the quantification of emissions and the analysis of environmental consequences for the consequent delineation of corrective and preventive measures related to air quality [63]. Since the 80’s, the inventory of emissions along with other estimates was made at national level (e.g. the National Energy Plan estimates). These inventories, however, were poor in information, presenting limitations on the range of pollutants and human activities under study. Later, the set of variables of the study was expanded, involving more sources emitting and new activities, including, among other sources, use of solvents, farming and livestock, and urban and industrial waste [63]. The current methodology applied to the inventories is constantly updated with the objectives established by the international entities responsible for the atmospheric emission inventory at world level [63].

Our results present values and maps of geographic distributions of atmospheric pollutant emissions for the years 2009 and 2015, highlighting the trend of each pollutant in this year range.

5.1.1. METHANE

The landfill sector presents the highest percentage of methane (CH_4) emissions in Portugal, followed by the wastewater sector and the livestock sector [63].

Although there was an increase of about 2.8% in CH_4 emissions between 2008 and 2009, there was a decrease in the statistics recorded between 2003 and 2009 of around 4.4%.

Figure 25 map represents the spatial distributions of the CH_4 pollutant emissions to 2009 and 2015. Few differences are accounted for, and between the two years of study, there is an improvement in the CH_4 emissions in the North region, while in the other regions CH_4 emission levels were maintained, presenting in the overall of each region a constancy in emissions evolution.

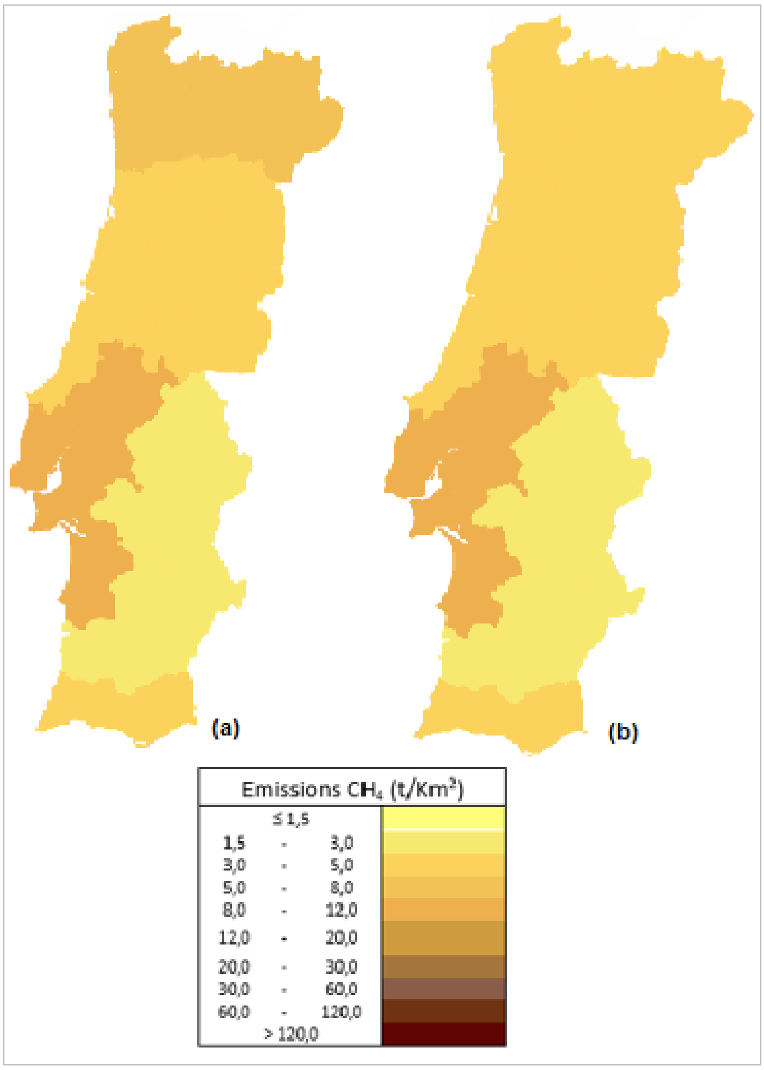


Figure 25 - Maps of spatial distribution of CH4 emissions in continental Portugal for the years 2009 (a) and 2015 (b)

5.1.2. AMMONIA

Depending on the levels of intensity of livestock and agricultural activity, ammonia (NH_3) emission levels vary both in geographic distribution and in emission values. Presence of point sources of emission associated to the sector of the industry, can lead to the existence of locations with high emission values. There was a downward trend in NH_3 emissions between 1990 and 2009 of around 22.7% [152].

Figure 26 explore the evolutionary trends of NH_3 emissions from 2009 to 2015. In relation to the NH_3 maps, there is a constancy in the NH_3 emission values evolution in the areas of the Center and Lisbon. In the North and *Alentejo* areas, there is an improvement in emissions, contrary to the trend observed in the Algarve region, which NH_3 emissions have increased over a period of 6 years.

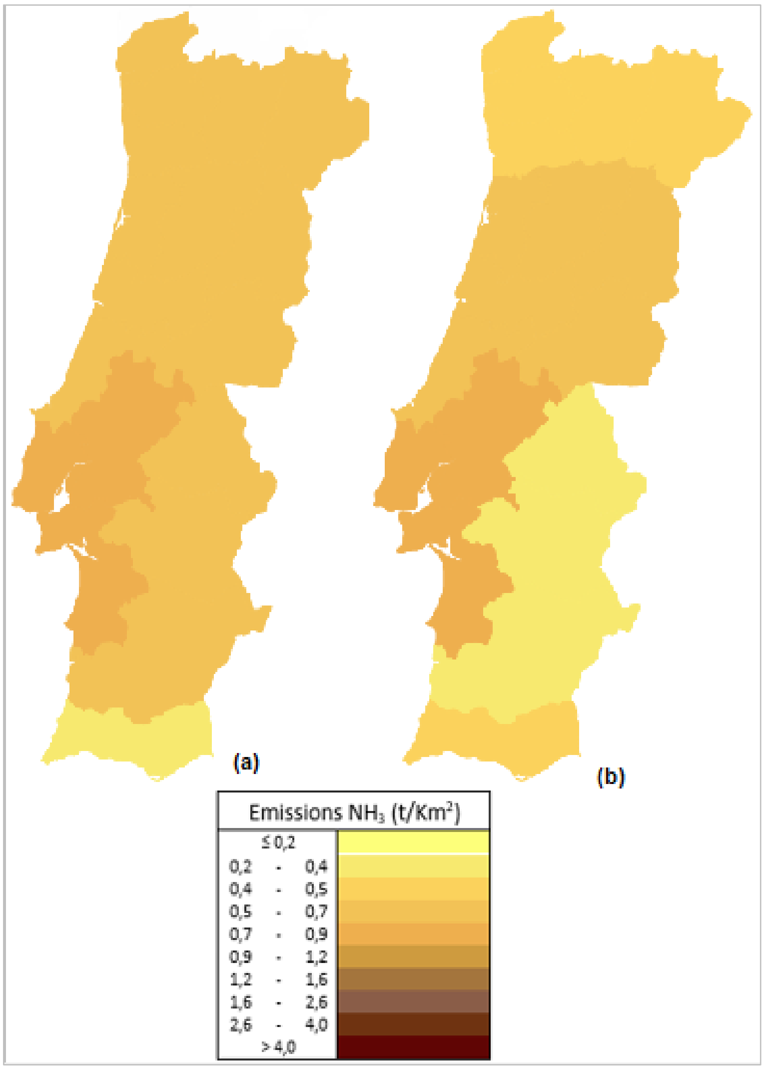


Figure 26 - Maps of spatial distribution of Ammonia emissions (NH3) in continental Portugal for the years 2009 (a) and 2015 (b)

5.1.3. Nitrogen Dioxide

Road traffic in urban centers is the main source of nitrogen dioxide (NO_2) with higher pollutant values in areas associated with industrial combustion, energy production and urban centers. Even though there was a decrease in NO_2 emissions between 2005 and 2009, the NO_2 variation between 1990 and 2009 shows an increase of 1.5% [152]. From 2009 to 2015, the emission variations of the pollutant can be observed in the emission maps of Figure 3.

Figure 27 shows an improvement in the emissions of NO_2 in the *Centro Interior*. The remaining national territory does not show any type of changes, comparing the trends between these two years.

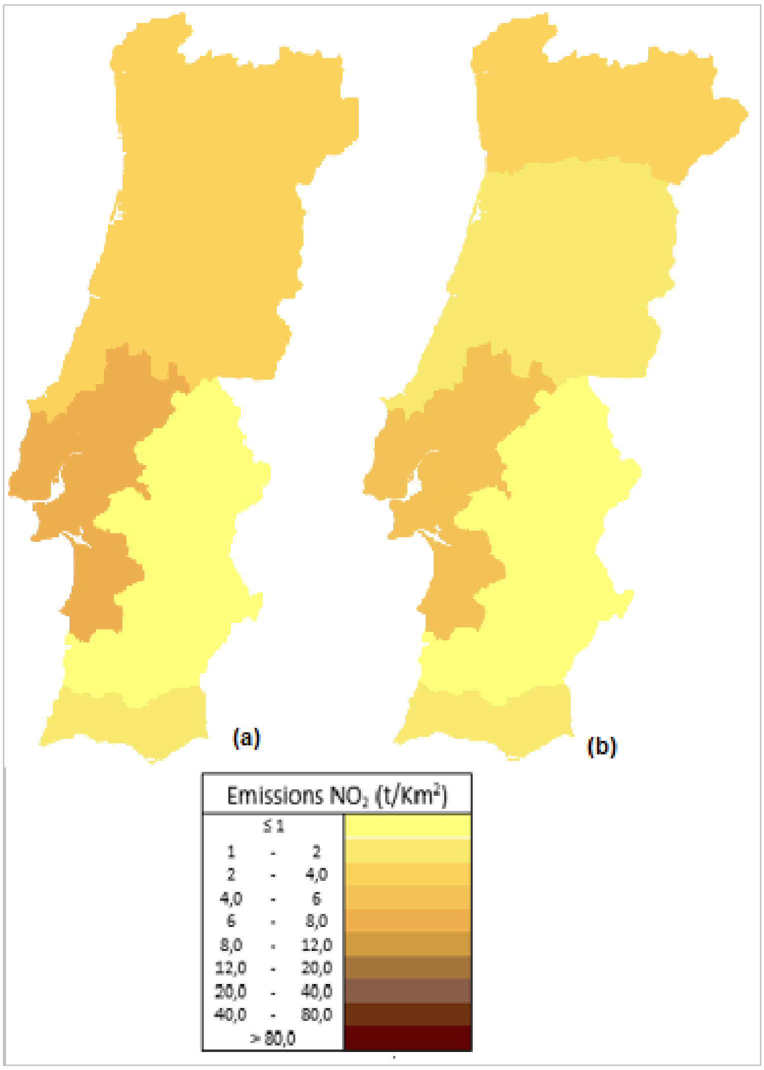


Figure 27 - Maps of spatial distribution of NO2 emissions in continental Portugal for the years 2009 (a) and 2015 (b)

5.1.4. SULFUR DIOXIDE

Considering that the main sources of sulfur dioxide (SO_2) emissions are from the industrial and energy production sectors, all locations where the main industries are located, have higher emission values compared to other regions of the territory. From 1990 to 2009, there was a significant reduction in SO_2 emissions of around 74% in the mainland [152].

Figure 28 shows the geographical dispersion of SO_2 emissions for the years 2009 and 2015. The emissions of SO_2 present some changes between the years 2009 and 2015, such as: the improvement in the regions of Algarve, North and Lisbon, and the absence of variation in emissions in the Central and *Alentejo* regions. The improvement observed in the Northern region is remarkable because it is a significant decrease in the emission values of SO_2 in the national territory.

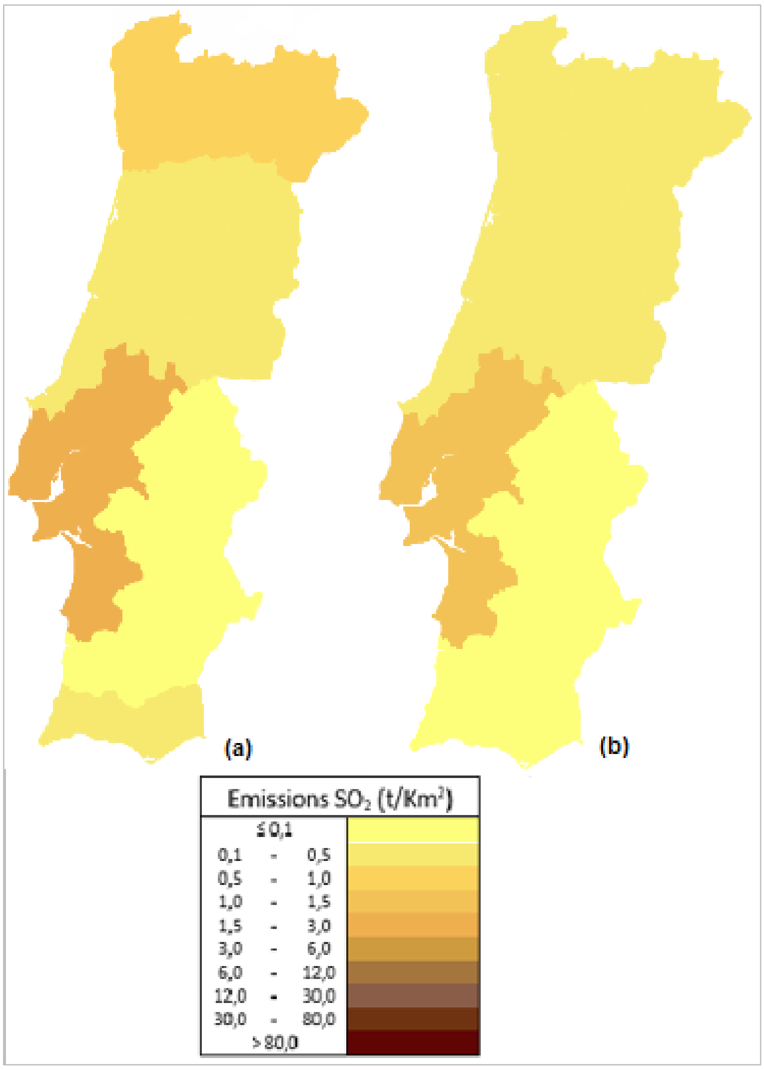


Figure 28 - Maps of spatial distribution of Sulfur Oxides emissions (SO2) in continental Portugal for the years 2009 (a) and 2015 (b)

5.1.5. CARBON DIOXIDE

Between 1990 and 2009, there was an increase of 26% of carbon dioxide (CO_2) emissions, estimating about 74.6 million tons. Focusing mostly on areas with high population density, CO_2 emissions are associated with urban centers (such as the country's coastal strip and metropolitan areas), industrial zones and energy production.

Figure 29 presents the geographic distribution of CO_2 emission trends. The case study of CO_2 also reveals several changes in the overall picture of emissions in national territory. The North and *Alentejo* regions show higher emissions in 2015 compared to 2009, while the Central and Lisbon regions are in the same range of emissions both in 2009 and 2015. The Algarve region was the only area of the country that shows improvements between these two years, revealing smaller emission levels of CO_2 emissions.

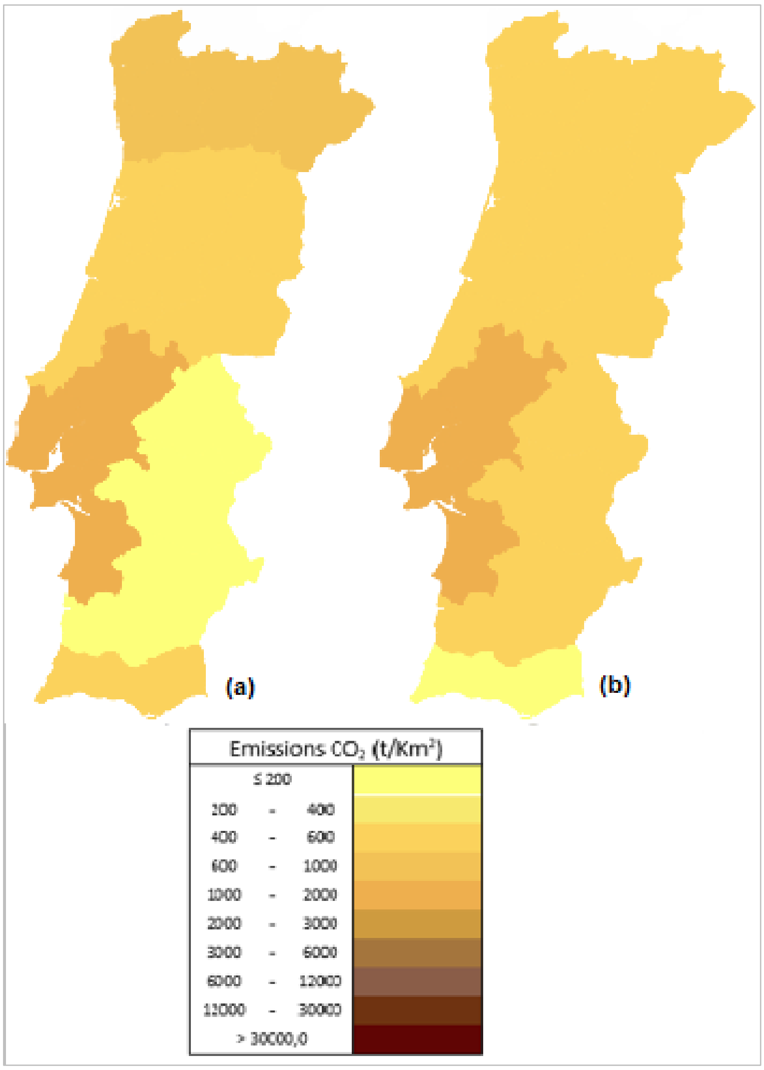


Figure 29 - Maps of spatial distribution of Carbon Dioxide emissions (CO2) in continental Portugal for the years 2009 (a) and 2015 (b)

5.1.6. Particulate Matter (PM10)

Similar to the situation observed in other pollutants, high emissions levels of particulate matter ≤ 10 micrometers (PM10) were recorded in places where the pollutants are located around a specific area, and these responsible sources account for around 33% of total PM10 emissions. The main activities that contribute to the emissions of this pollutant are distributed throughout the Portuguese territory, namely: (i) pulp production (*Viana do Castelo, Aveiro, Figueira da Foz, Vila Velha de Rodão* and *Setúbal* municipalities); (ii) steel industry (*Maia* and *Seixal* municipalities); (iii) chemical industry (*Estarreja, Vila Franca de Xira*, *Barreiro, Setúbal* and *Sines* municipalities) [153].

The PM10 emissions presented some changes over the years, with a 7.6% increase between 2003 and 2008, an important and considerable decrease from 17% from 2008 to 2009.

Figure 30, shows the trend of PM10 emissions, comparing both PM10 emission maps for 2009 and 2015. There is an improvement in emission values for almost the entire territory, with only the central region showing no variation between these years. All other regions show a reduction in emissions, leading to an overall improvement in PM10 emissions.

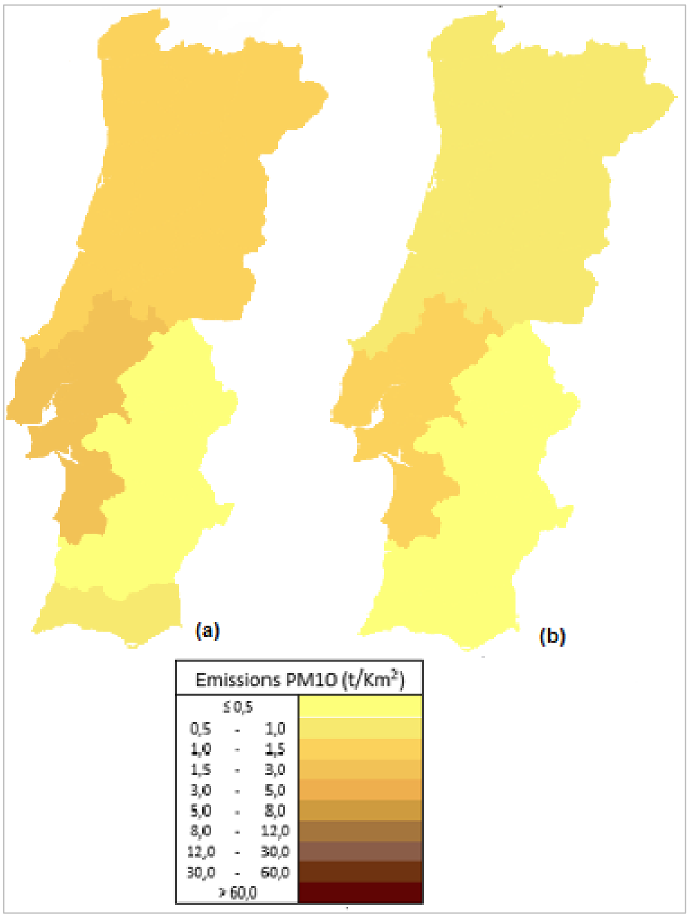


Figure 30 - Maps of spatial distribution of Particulate Matter emissions (PM10) in continental Portugal for the years 2009 (a) and 2015 (b)

5.1.7. PARTICULATE MATTER (PM2.5)

The complex mixture that is the particulate material, comes from several emitting sources, highlighting the road traffic in the urban centers and the industrial processes, these being the great emitting sources of PM2.5 [154]. The INERPA presents for the first time in 2015, results of particulate matter ≤ 2.5 micrometers (PM2.5) pollutant, an indicator of air quality for assessing the quality of air and health related quality of life. Since there is no analysis of previous years regarding emission levels, the analysis of this pollutant, presented in Figure 31 was calculated only for 2015. In the absence of an evolutionary emission trend between 2009 and 2015, it was performed a comparative analysis with PM10, expecting possible similarities in emission sources at the regional and national level.

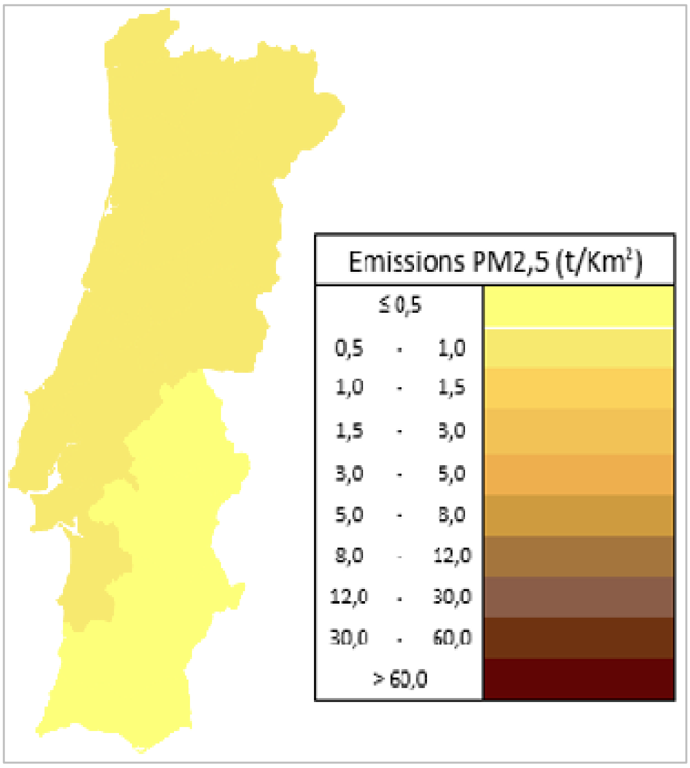


Figure 31 - Map of spatial distribution of Particulate Matter emissions (PM2.5) in continental Portugal for the year 2015

Regarding PM2.5 emissions the northern half of the country presents higher emissions than the southern half, presenting a homogeneous level within the northern and southern regions themselves, but a worst-case scenario for the northern region. This phenomenon may be related to a higher population density, a more developed industry sector and higher traffic flow than the southern inner territory regions.

Since PM2.5 belongs to the fraction of PM10 [155], several studies present the hypothesis of a linear relationship between the concentrations of these atmospheric pollutants, performing a linear regression analysis for the estimation of concentrations of PM2.5 concentrations of PM10 [156, 157].

One of the tools used to determine a linear relationship of pollution between pollution levels and climatic conditions is the Pearson correlation coefficient, which also involves temporal and spatial variability in its variables [158, 159].

It should be noted that the relationship between climatic conditions and PM is not a simple correlation, involving several factors associated, among other fields, with geographic location and season [156].

Even though there is a large fraction of PM2.5 in PM10 constitution, most of the cities with atmospheric pollution from transportation and other sources associated with combustion often neglect sources of non-combustion, such as dust suspension, the coarser fraction of the particle composition. As such, it becomes necessary to measure and analyze either PM10 or PM2.5 [160].

The comparative analysis of the spatial distributions of PM10 and PM2.5 emissions presents some conclusions, such as: the similarity of areas with higher atmospheric pollutant emissions, with the regions of Lisbon, Center and North presenting higher values than remaining regions for both pollutants; and the existence of higher values of PM10 in the whole country, compared to PM2.5, this large discrepancy in values is due to the emission values (and even concentration values) of PM10 encompassing the values referring to PM2.5.

Even if the PM2.5 emission values are included in the values associated with PM10, the PM2.5 analysis becomes crucial for the geographic identification of the critical sites of this pollutant, with more harmful effects on human health, compared to PM10.

5.1.8. EMISSIONS OVERVIEW

Summarizing the emissions analysis of the years 2009 and 2015, for the various pollutants, several points can be highlighted: (i) the regions of the *Alentejo* and *Algarve* have reduced values in the analysis of the different pollutants compared to the other regions, with some decreases emissions in these regions; (ii) the Lisbon metropolitan area presents the most problematic region with regard to the emissions of all the studied pollutants, showing scarce improvements between 2009 and 2015 and high pollutant emissions compared to all other regions; (iii) the North region presented, in some pollutants, values at an intermediate level, and other pollutants as particles presented reduced values, compared to the other regions; (iv) only in the *Centro Interior* region, between 2009 and 2015, there was an increase in CO_2 emissions.
Tables XX and XX show the variations of air pollutant emissions for the years under study, by region, highlighting the percentages of variations for each region and for mainland Portugal.

Table 10 - Summary table of the variations between emissions of atmospheric pollutants (NOx, SOx, PM10, CH4) for the years 2009 and 2015

Pollutants	**NOx**			**SOx**			**PM10**			**CH_4**		
Years/Variation	**2009**	**2015**	**Δ**	**2009**	**2015**	**Δ**	**2009**	**2015**	**Δ**	**2009**	**2015**	**Δ**
	(t/Km^2)	**(t/Km^2)**	**(%)**	**(t/Km^2)**	**(t/Km^2)**	**(%)**	**(t/Km^2)**	**(t/Km2)**	**(%)**	**(t/Km^2)**	**(t/Km^2)**	**(%)**
Algarve	1,974	1,306	-33,8	0,117	0,058	-103,4	0,567	0,393	-30,8	3,648	3,203	-13,9
Centro	2,785	1,836	-34,1	0,463	0,472	1,9	1,391	0,858	-38,3	4,172	3,744	-11,4
Norte	2,841	2,068	-27,2	0,705	0,262	-169,3	1,085	0,757	-30,2	6,890	5,243	-31,4
Lisboa	6,664	4,758	-28,6	2,473	1,331	-85,8	2,578	1,138	-55,9	10,347	8,922	-16,0
Alentejo	0,408	0,376	-7,8	0,025	0,014	-86,7	0,197	0,130	-33,7	1,523	2,755	44,7
Portugal			-26,3			-88,7			-37,8			-5,6

Table 11 - Summary table of the variations between emissions of atmospheric pollutants (CO2, NH3, PM2.5) for the years 2009 and 2015

Pollutants	**CO_2**			**NH_3**			**PM2,5**
Years/Variation	**2009**	**2015**	**Δ**	**2009**	**2015**	**Δ**	**2015**
	(t/Km2)	**(t/Km2)**	**(%)**	**(t/Km2)**	**(t/Km2)**	**(%)**	**(t/Km2)**
Algarve	317,648	260,254	-22,1	0,307	0,239	-28,5	0,276
Centro	403,777	415,274	2,8	0,501	0,619	19,0	0,694
Norte	621,638	505,331	-23,0	0,543	0,486	-11,6	0,570
Lisboa	1934,885	1922,466	-0,6	0,740	0,806	8,1	0,961
Alentejo	56,520	51,548	-9,6	0,565	0,402	-40,7	0,072
Portugal			-10,5			-10,7	

5.2. Air Quality Concentrations Overview

Air quality index and evaluation is performed through monitoring stations distributed throughout the national territory, performing specific functions and having distinct objectives between them. In addition to industrial and traffic monitoring stations, the most common type of station is the background station, working with urban, suburban and rural day-to-day data.

The spatial distribution of the background stations considered in this analysis are presented in Figure 32. As shown most stations are in the littoral part of the national territory, with some stations responsible for monitoring the inner country regions. The monitoring stations distribution represents the population density distribution focused more in urban centers, followed by the amount of pollutants generated on those areas.

Figure 32 - Spatial distribution of background monitoring stations in urban environment, used in this analysis

Table 10 displays the identification of the reported monitoring stations, describing the IQAr zones, county, and station designation.

Table 11 presents the annual concentrations of ozone and nitrogen dioxide for the years 2009 and 2015. Table 11 analysis presented an improvement in ozone concentrations between the years 2009 and 2015, almost throughout the national territory. Nevertheless, this development was not accompanied by the same trends regarding NO2 concentrations. Although some improvement, particularly in the Central region, throughout the rest of the territory, there was an increase in NO2 concentrations, thus worsening air quality. Some values of these variations had an increase of NO2 concentrations of more than 200% between 2009 and 2015, at the northernmost station of the country. In opposite, the same station (Paços de Ferreira) presented an improvement of about 30% of ozone concentrations.

Table 12 - Identifying information of the background monitoring stations in urban environment, used in this analysis (original denomination)

ZONE IQAR	COUNTY	STATION
Norte Litoral	*Viana do Castelo*	*Minho-Lima*
Entre Douro e Minho	*Paços de Ferreira*	*Paços de Ferreira*
Norte Interior	*Vila Real*	*Douro Norte*
Porto Litoral	*Porto*	*Sobreiras-Lordelo do Ouro*
Centro Interior	*Fundão*	*Fundão*
Aveiro/Ílhavo	*Ilhavo*	*Ilhavo*
Centro Litoral	*Leiria*	*Ervedeira*
Coimbra	*Coimbra*	*Instituto Geofisico de Coimbra*
Oeste, Vale do Tejo e Península de Setúbal	*Lourinhã*	*Lourinhã*
Lisbon Metropolitan Area - Norte	*Lisbon*	*Olivais*
Lisbon Metropolitan Area - Sul	*Almada*	*Laranjeiro*
Setúbal	*Setúbal*	*Arcos*
Alentejo Litoral	*Santiago do Cacém*	*Monte Velho*
Alentejo Interior	*Alandroal*	*Terena*
Algarve	*Alcoutim*	*Cerro*
Aglomeração Sul	*Faro*	*Joaquim Magalhães*

Table 13 - Concentrations of pollutants (O_3; NO_2), in the years 2009 and 2015, for the different monitoring stations selected (source: QualAr [38])

STATIONS (ORIGINAL DENOMINATION)	**2009**	**2015**	**Improvement**	**2009**	**2015**	**Improvement**
	O_3 (µg/m^3)	**O_3 (µg/m^3)**	**(%)**	**NO_2 (µg/m^3)**	**NO_2 (µg/m^3)**	**(%)**
Minho-Lima	60,9	54,4	10,7	4,1	12,6	-207,3
Paços de Ferreira	43,5	29,6	32,0	18,5	17,2	7,0
Douro Norte	90,6	74,8	17,4	3,4	4,3	-26,5
Sobreiras-Lordelo do Ouro	50,4	39,9	20,8	20,6	25,3	-22,8
Fundão	67,8	65,0	4,1	5,4	6,9	-27,8
Ilhavo	53,0	40,8	23,0	8,6	13,5	-57,0
Ervedeira	62,7	53,6	14,5	5,9	5,9	0,0
Instituto Geofisico de Coimbra	54,8	51,7	5,7	15,3	14,7	3,9
Lourinhã	68,4	62,7	8,3	6,4	5,3	17,2
Olivais	48,9	49,5	-1,2	34,1	29,2	14,4
Laranjeiro	55,4	54,8	1,1	30,3	26,6	12,2
Arcos	63,7	59,7	6,3	19,0	14,2	25,3
Monte Velho	63,9	70,4	-10,2	5,5	5,9	-7,3
Terena	46,1	42,3	8,2	6,6	3,5	47,0
Cerro		79,1			2,8	
Joaquim Magalhães		62,9			11,2	

Some limitations should be noted that, even after selecting the pollutants with the highest percentage of complete data, there are still some gaps in the presented values. However, for the qualitative nature of this analysis, the absence of these values had a reduced effect on the possible conclusions and discussions of these results.

Table 12 shows the concentrations of SO_2 and PM10, for the years under study, for the selected monitoring stations.
In the case of SO_2, several values are not presented due to a reduced percentage of time records for the years in question, which would make any presentation of annual values inaccurate.
With exception to Lisbon metropolitan area and *Alentejo Litoral* region, SO_2 values between the two years showed improvements in other regions (stations), from about 8% to almost 100%. In cases where air quality deteriorated for SO_2, there was an increase in concentration values of almost 70% for Lisbon and more than 30% for the *Alentejo Litoral* region.

When analyzing the variations of PM10 concentrations throughout all the monitoring stations under study, it is confirmed that, excluding 5 cases of increase of concentrations, all the other stations registered improvements between the two years.
In the southernmost regions of the country there are only improvements in air quality related to PM10. Also, in the northern half of Portugal there were some improvements, however there were a significant number of concentrations increasing between 40% and 60%.
Table 4 presents the concentration values addressing fine particles (PM2.5).
These results (Table 13) provided a clear picture that the southern part of the country had worsened its air quality for PM2.5 between 2009 and 2015. While in the northern half of Portugal, there were improvements along the selected stations in that region.

In some of the abovementioned pollutants, the lack of data in some monitoring stations is mainly due to the fact that there is not a sufficient number of hourly values for a year so that an annual average can be made, either on a daily basis or hourly basis, ensuring the accuracy of the data presented. Other reason for the lack of data is the reorganization of the pollutants under study in each station. With the population density fluctuation, several stations change pollutants to analyze, when their demand is no longer justified.

Regarding the analysis of air quality, given the concentrations of several pollutants between the years of study, it can be noted that: (i) in relation to ozone, concentration decreases were registered throughout the country; (ii) in the case of NO_2 and PM10, most of the northern half of the country shows an increase in pollutant concentrations of the, a situation that is contrary to the South half that presents improvements in air quality; in the analysis of SO_2 and fine particles (PM2.5), the regions of *Alentejo* and the Lisbon metropolitan area show increases in concentrations between 2009 and 2015, while the other regions show improvements with decreasing concentrations of pollutants existing in the atmosphere.

Table 14 - Concentrations of pollutants (SO_2; PM10), in the years 2009 and 2015, for the different monitoring stations selected (source: QualAr [38])

STATIONS (ORIGINAL DENOMINATION)	2009	2015	Improvement	2009	2015	Improvement
	SO_2 (µg/m³)	SO_2 (µg/m³)	(%)	PM10 (µg/m³)	PM10 (µg/m³)	(%)
Minho-Lima	1,1			15,9	14,5	8,8
Paços de Ferreira	1,9			13,5	21,7	-60,7
Douro Norte	5,6	3,6	35,7	20,4	18,1	11,3
Sobreiras-Lordelo do Ouro	5,4			24,8	17,5	29,4
Fundão	1,8	0,3	83,3	12,5	13,8	-10,4
Ilhavo	3,4	0,1	97,1	20,8	26,7	-28,4
Ervedeira	4,5	2,1	53,3	15,6	22,0	-41,0
Instituto Geofisico de Coimbra	1,3	1,0	23,1	20,1	20,6	-2,5
Lourinhã	1,6			18,9	15,3	19,0
Olivais	0,6	1,0	-66,7	26,2	20,2	22,9
Laranjeiro	1,0			30,5	22,0	27,9
Arcos				26,6	23,4	12,0
Monte Velho	4,2	5,5	-31,0	23,5	22,1	6,0
Terena	3,4	3,1	8,8	23,4	19,7	15,8
Cerro		3,5			17,5	
Joaquim Magalhães		4,7			21,6	

Table 15 - Concentrations of pollutants (PM2.5), in the years 2009 and 2015, for the different monitoring stations selected (source: QualAr [38])

STATIONS (ORIGINAL DENOMINATION)	2009	2015	Improvement
	PM2.5 (µg/m³)	PM2.5 (µg/m³)	(%)
Minho-Lima	10,2		
Paços de Ferreira	8,2	8,2	0,0
Douro Norte	5,0	4,5	10,0
Sobreiras-Lordelo do Ouro	8,9	5,1	42,7
Fundão	7,1	5,6	21,1
Ilhavo			
Ervedeira	6,8	7,4	-8,8
Instituto Geofisico de Coimbra			
Lourinhã	8,7	7,4	14,9
Olivais	11,3	11,3	
Laranjeiro	11,9	13,5	-13,4
Arcos			
Monte Velho	11,0	12,8	-16,4
Terena	9,6	12,3	-28,1
Cerro		6,7	
Joaquim Magalhães		11,9	

5.3. PUBLIC HEALTH INDICATORS

The analysis of public health indicators used in this study focuses on data on mortality from specific causes of death. These data were developed by the National Institute of Statistics (INE), using data from the Integrated System of Civil Registry and Identification (SIRIC) [161] and the Information System for Death Certificates (SICO) [162, 163].

The increase in life expectancy and the compression of deaths at more advanced ages translates the age standard of mortality, associated to the progressive transformation of the causes of morbidity and mortality. Given the causes of mortality, undefined causes are presented as a problem in mortality analyzes, representing about 10% of the total number of deaths [164].

Mortality and morbidity rates associated with diseases of the circulatory system have a very significant impact because, if these medical implications did not exist, life expectancy would increase by about 5 ½ years for men and about 4 years for women [164].

Since the beginning of the years 2000 to 2009, there has been a decrease in the number of deaths caused by diseases of the circulatory system, of around 18.5%, even though there was a 3.7% increase between 2006 and 2007, as Figure 33. Regarding the deaths caused by medical complications related to the respiratory system, there were several fluctuations in the annual values between 2003 and 2009. As can be seen in Figure 34, from 2003 to 2004, there was a decrease in the values of 9.1%. Registered only a further decrease between 2006 and 2007 of 4.8%. Between 2003 and 2009, in an overview, it registered an increase of about 27.6%, highlighting the sharp increase between 2004 and 2005 of 30.3%.

Figure 35 and 36 show the evolutionary trends number of deaths associated to causes indirectly or directly related to emissions of atmospheric pollutants and consequent inhalation or contact. The information is divided by associated death and according to geographic distribution, grouping in regions of Portuguese national territory.

The data related to the number of deaths associated with malignant neoplasm of the respiratory tract indicate a constant increase between 2009 and 2015, highlighting the similarity of the variations from year to year in all regions. Figure 35 presents the shape of the resulting line in the graphic equal between different regions, even though it is presented in different levels of values. Another point to emphasize is the clear division between the northern and southern regions, and the values, although they follow the same trend, are clearly higher for the southernmost regions of the country compared to the northern regions.

Blood circulation and bidirectional transport of nutrients and gases between cells, are functions of the circulatory system. Given that this vital system involves blood vessels where gas and nutrient exchange occurs, any factor affecting the integrity of the tubing can impair the health of the circulatory system [165]. Figure 36 represents the evolutionary tendency between the years of study, individualizing the different zones of the national territory.

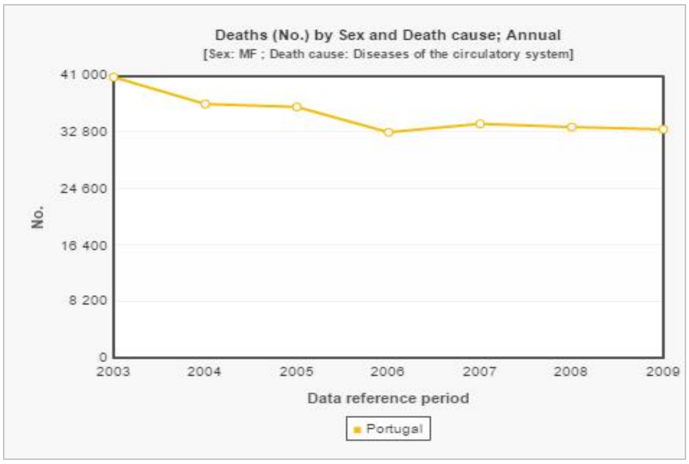


Figure 33 - Graphical representation of the evolution of the number of deaths from circulatory system diseases from the year 2003 to 2009, in Portugal (INE) [55]

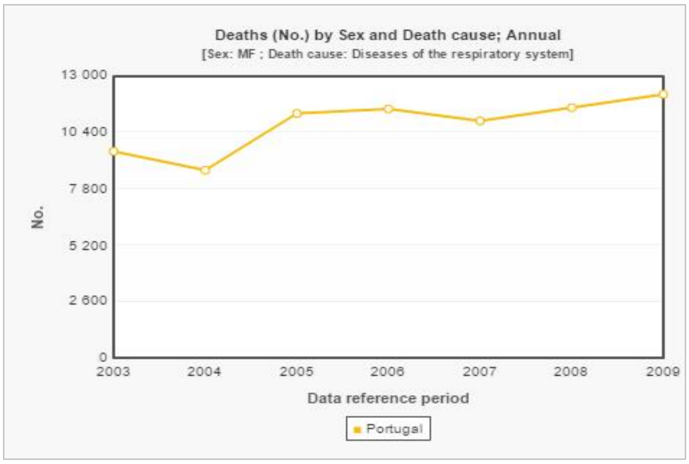


Figure 34 - Graphical representation of the evolution of the number of deaths from respiratory system diseases from the year 2003 to 2009, in Portugal (INE) [56]

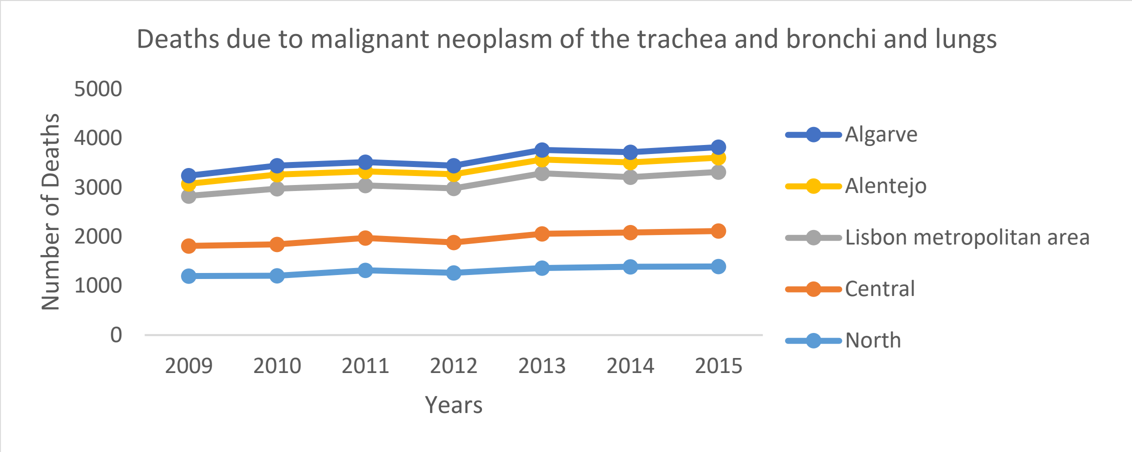


Figure 35 - Graphical representation of the evolution of the number of deaths due to malignant neoplasm of the trachea and bronchi and lungs from the year 2009 to 2015, in the different regions of continental Portugal

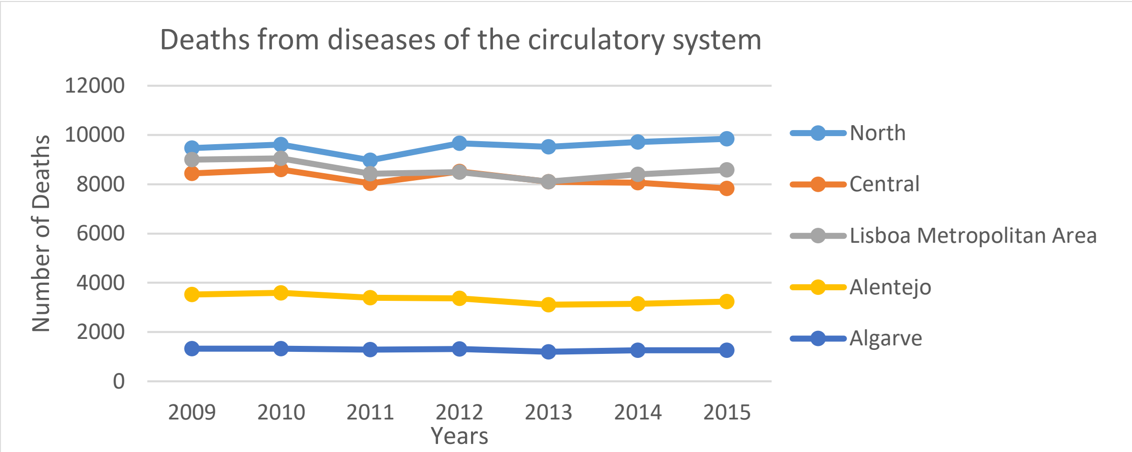


Figure 36 - Graphical representation of the evolution of the number of deaths from diseases of the circulatory system from the year 2009 to 2015, in the different regions of continental Portugal

In contrast, death cause due to malignant respiratory tract neoplasia, North and Center presented the lower values compared to other regions. This specific death cause was associated with complications of the circulatory system and pointed out the regions of Algarve and *Alentejo* with the lowest number of deaths, thus separating the southern regions of the country, with lower values, from the more northern east regions presenting highest values.

When approached for analysis, the values referring to the number of deaths due to diseases of the circulatory system, shows evolutionary trends similar across the country, given the clear inequality of values of each region.

One point to note is that although the lines shown on Figure 36 do not indicate abrupt variations, they all show small differences in behavior over time, such as the stabilization of the Algarve line; the small but steady decrease in the figures for the Central zone from 2012 to 2015, something that is not observed in the other regions; and the decreasing number of deaths in the Lisbon metropolitan area and the *Alentejo* region between 2010 and 2013.

Ischemia refers to deficiencies found in the blood supply in the body, being one of the most common causes of death in Western countries. This blood supply is due to the blockage of coronary arteries, responsible for the supply of blood to the heart muscle.
Figure 37 shows the graphical representation of the number of deaths among the years of study, referring to this type of medical complications [166].
Contrary to the cases previously seen, the evolution of the number of deaths by ischemic heart disease, is highlighted by the lines resulting from the aggregation of the values by region to be separated from each other. Taking in consideration the analysis of the specific case of the Lisbon metropolitan area, which presented the highest number of deaths in all the years observed, the graph presented in Figure 37 shows a trend of increasing numbers of deaths in each year, starting from the southern regions towards the northern regions of Portugal.

In addition to the high values registered for Lisbon, another highlight in the analysis is the sudden increase in the number of deaths associated with ischemic heart disease in the North between the years 2013 and 2014.

In the world, respiratory-related diseases, although presenting similar characteristics between so, present different clinical results.

Due to the high prevalence of this type of disease and common pathophysiological pathways, certain patients have similar symptoms of different medical complications [167].

In this sense, Figure 38 presents the evolutionary trends between 2009 and 2015 for the different regions of the national territory.
With a similar situation to the case of the temporal evolution of the number of deaths due to ischemic diseases of the heart, the analysis of the development of the variations of the numbers of deaths related to diseases of the respiratory tract reveals a uniform behavior of the evolution of the values over the years under study.
Although there are clear differences in values presented each year across the different regions of the country, the behavior of the different rows resulting in Figure 38, show practically equal variation gradients from year to year.
Therefore, certain notes can be taken regarding the general evolutionary trends of the regions. First of all, it should be distinguished that there is a clear peak in the number of deaths in the year 2012, having peaked after a growing trend of deaths throughout the country.
Following the peak of 2012, there was a decrease in deaths in all regions for about 2 years, immediately followed by a sharp increase in the North, Center and Lisbon regions, and a small increase in the *Alentejo* and Algarve regions.

Chronic obstructive pulmonary disease, asthma and pulmonary hypertension are some of the most common types of chronic respiratory disease. The most common causes of this disease, besides smoking and inhalation of chemicals, are risk factors such as atmospheric pollution and dust respiratory infections [168].

The information presented in Figure 39 represents the evolution of the numbers of deaths related to chronic lower airway diseases between the years 2009 and 2015.

When discussing the case of the evolution of the number of deaths by chronic lower airway diseases, similarities are found with the remaining analyzes of the causes of death previously presented.

The spacing between values across the different regions is one of the situations similar to previous cases, placing the southernmost regions of the country (*Alentejo* and Algarve) at a lower level, while the North occupies the highest level of deaths.

Another situation observed in the chart above is the evolution pattern of the number of deaths. With this pattern, similar across regions, it was possible to identify a peak of values in 2012, subsequently unrolling a U-pattern. This point that does not comply with the behavior of the trend lines of the other regions is in the Lisbon metropolitan area between the years 2013 and 2015. This behavior describes, between the year 2013 and 2014, an increase in death values associated with chronic lower airway diseases, followed by a stagnation of values (contrary trend to the remaining regions that presented continuous decreases in values between the peaks of 2012 until 2014).

In addition to aggravating the medical condition associated with asthma, air pollutants can also trigger the disease itself [169-171], and this atmospheric pollutants associated with road-related emissions have higher concentrations in urban centers [172].

In adults, air pollution influences the peripheral blood epigenome [173-180], and the epigenome is described as a complete description of potentially hereditary changes throughout the genome. At the cellular level, the epigenome is composed of genetic determinants, lineage and environment [172, 181].

Pollution associated with pollutant emissions may also be responsible for modifications in the process of DNA methylation (being an essential process for the development of mammary embryos, being also related to some human pathologies [182]) [172, 183].

These changes also encompass genes of innate immunity [174, 178] and asthma [177], both in cases of long-term exposure [175, 177] and in cases of short-term exposure [174, 176, 179]. The analysis of asthma as a cause of death results in Figure 16 with an evolutionary behavior over the years completely different from the other charts associated with the causes of deaths previously identified.

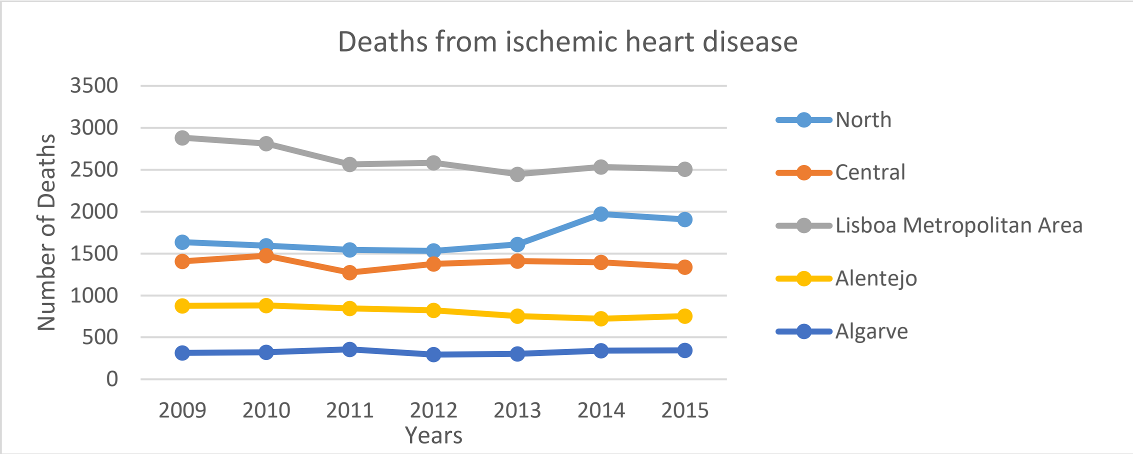


Figure 37 - Graphical representation of the evolution of the number of deaths from ischemic heart disease from the year 2009 to 2015, in the different regions of continental Portugal

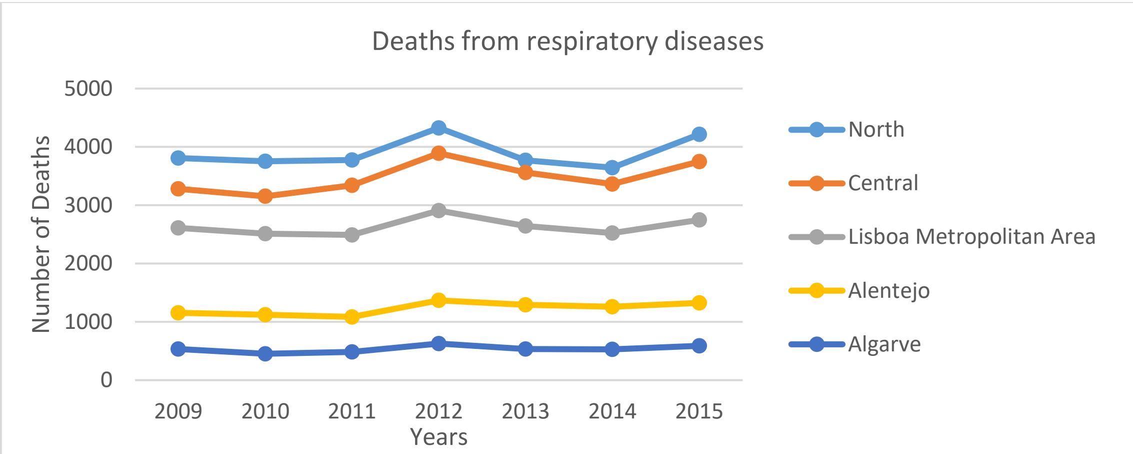


Figure 38 - Graphical representation of the evolution of the number of deaths from respiratory diseases from the year 2009 to 2015, in the different regions of continental Portugal

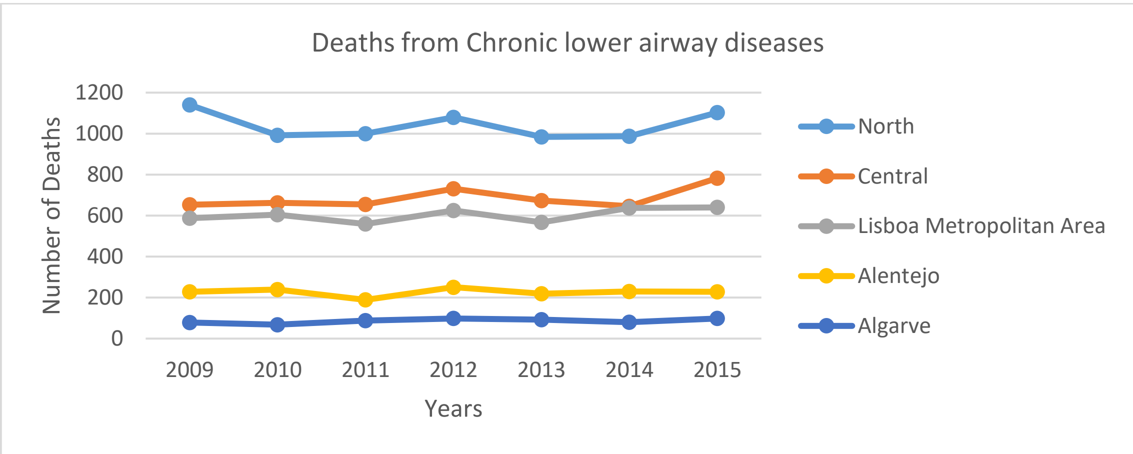


Figure 39 - Graphical representation of the evolution of the number of deaths from chronic lower airway diseases from the year 2009 to 2015, in the different regions of continental Portugal

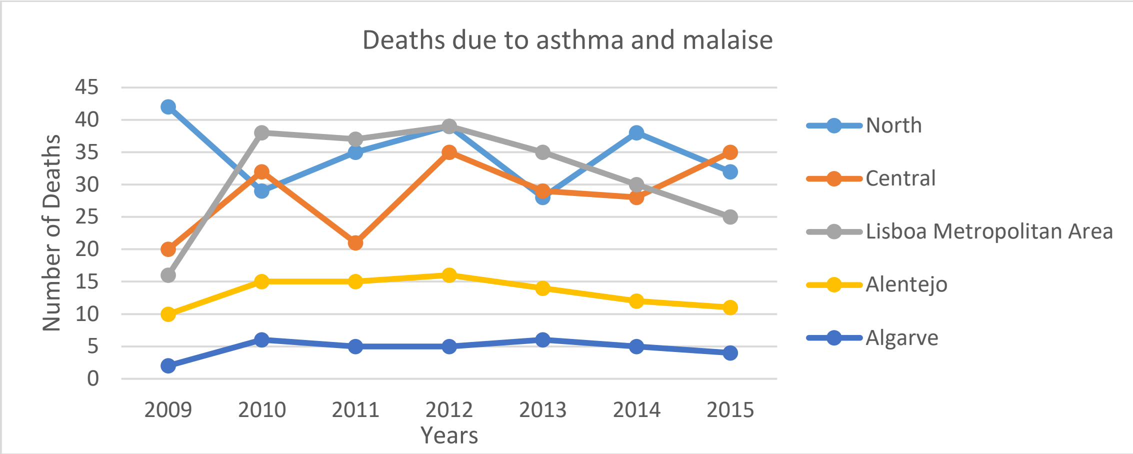


Figure 40 - Graphical representation of the evolution of the number of deaths due to asthma and malaise from the year 2009 to 2015, in the different regions of continental Portugal

Although the regions of *Alentejo* and Algarve continue to present ranges of values and behavior patterns similar to previous analyzes, the remaining regions show large variations in values each year and present a high variability between increase and decrease of values from year to year, hindering the direct definition of a behavior of the evolution of the number of deaths.

All regions have a peak in values between 2012 and 2013, excluding the North regions, where a peak is identified in 2009. Although there is a high increase in figures from 2009 to 2010 and a slight increase between 2011 and 2012, the region of Lisbon, after reaching its peak, shows a steady downward trend.

The Center region is the only one that reaches 2015 with an increasing tendency of values, considering that between 2009 and 2012 it had presented increasing and decreasing sharp variations, with respect to number of deaths values.

5.3.1. PUBLIC HEALTH OVERVIEW

For a better understanding of the evolution of health indicators over the years, information was gathered on the number of deaths associated with diseases of the respiratory system and circulatory system, with the purpose of presenting, without overlapping information, quantitative information on the variations recorded over the years under study.

In tables XX and XX, information related to the variations in the number of deaths associated with respiratory diseases and the circulatory system are presented.

Table 16 - Summary table of the variations in the number of deaths associated with respiratory diseases between 2009 and 2015 for the regions of mainland Portugal

No. of deaths caused by diseases of the respiratory system

Regions	2009	Δ	2010	Δ	2011	Δ	2012	Δ	2013	Δ	2014	Δ	2015
Norte	3809	-1,5%	3753	+0,5%	3773	+14,7%	4326	-12,9%	3770	-3,3%	3644	+15,7%	4216
Centro	3282	-4,0%	3152	+6,0%	3341	+16,5%	3891	-8,5%	3560	-5,6%	3361	+11,4%	3745
A.M. Lisboa	2612	-3,8%	2512	-0,8%	2491	+16,7%	2908	-9,1%	2643	-4,6%	2521	+9,0%	2747
Alentejo	1153	-2,6%	1123	-3,4%	1085	+26,1%	1368	-5,5%	1293	-2,6%	1260	+5,0%	1323
Algarve	536	-15,5%	453	+6,6%	483	+30,0%	628	-14,6%	536	-0,9%	531	+11,1%	590
Norte							+11%						
Centro							+14%						
A.M. Lisboa	↘						+5%						↗
Alentejo							+15%						
Algarve							+10%						

Table 17 - Summary table of the variations in the number of deaths associated with circulatory diseases between 2009 and 2015 for the regions of mainland Portugal

No. of deaths caused by diseases of the circulatory system

Regions	2009	Δ	2010	Δ	2011	Δ	2012	Δ	2013	Δ	2014	Δ	2015
Norte	9470	+1,5%	9610	-6,6%	8977	+7,6%	9661	-1,4%	9527	+2,0%	9722	+1,3%	9845
Centro	8442	+1,8%	8598	-6,5%	8035	+6,0%	8517	-4,9%	8102	-0,5%	8064	-2,9%	7834
A.M. Lisboa	9004	+0,5%	9051	-6,9%	8426	+0,9%	8499	-4,6%	8109	+3,7%	8406	+2,2%	8587
Alentejo	3522	+2,0%	3593	-5,6%	3393	-0,9%	3364	-7,6%	3110	+1,0%	3142	+2,9%	3232
Algarve	1325	-0,1%	1324	-2,4%	1292	+1,8%	1315	-8,8%	1199	+4,8%	1257	+0,3%	1261
Norte							+4,0%						
Centro							-7,2%						
A.M. Lisboa	↘						-4,6%						↗
Alentejo							-8,2%						
Algarve							-4,8%						

5.4. ENVIRONMENT AND HEALTH CONSIDERATIONS

Even though it is explicit that there is no simple correlation between the three fields addressed in this study (emissions of air pollutants, air quality, public health, more specifically mortality associated with causes potentially caused by atmospheric pollution), some hypotheses were pointed out.

The geographical proximity between the critical points of high pollutants emission with points of high concentrations of pollutants can be found throughout the national territory, formulating the hypothesis of the occurrence of transport phenomena in the short range, increasing concentrations of pollutants in the adjoining areas from emission points.

Industrial areas with a high population density, such as the *Setúbal*, Lisbon and Porto, not only had high pollutants emissions, but also high concentrations of pollutants.

The impact of air pollution on public health depends fundamentally on the likelihood of the risk of exposure to the contaminant [184]. Health, economic and environmental policies are based on empirical evidence from studies by economists [185, 186], whose purpose is to determine the economic implications of the current state of the environment and health, and the changes registered over time [184]. Among the three basic components of this work, it is possible to establish a causal relationship between them, proposing that variations in the emissions of atmospheric pollutants, lead to variations in concentrations and, consequently, lead to variations in health status. Therefore, any preventive measures to improve air quality should initially focus on emissions reduction and control.

The monitoring, management and decision-making entities, should regulate even further large industries such as the pulp and paper industry, chemicals and textiles, in terms atmospheric pollutants emissions. Allocation of resources, distributing industry across the national territory, decollating the points of emission of pollutants, and active control of industrial and environmental policies, are some of the possible measures for the process of control and reduction of air pollution and subsequent health effects [184].

6. FINAL REMARKS

In this work, environmental issues were addressed, focusing in particular on the air pollutants, both primary and secondary, analyzing: emissions of these pollutants with spatial distribution by region (NUT II); concentrations of these pollutants in Portuguese territory; and analysis of the evolution of mortality associated with causes of death in which air pollution participates as one of the possible causes.
The analysis of these three components included data of two years, 2009 and 2015, highlighting a comparative qualitative analysis between the two years of study.

In the European context over a 9-year period (2002-2011), emissions of major air pollutants have decreased, thereby improving air quality throughout the European region. Some of these pollutants show reductions with some variations, since several parts of Europe face transport of intercontinental pollutants by air and also present polluting sources of different types and weights. For particulates (PM10), there was a decrease between 20% and 44% of emissions, a decrease that was accompanied by other pollutants such as: ozone (14% - 65%); nitrogen oxides (approximately 27%); sulfur oxides (about 50% reduction); and carbon monoxide, which decreased by about 35% in a maximum daily period of eight hours [187]. Although there are reductions in primary pollutant emissions, the corresponding concentrations of pollutants are not always in tune, i.e. due to the complexity of the chemical relationships between pollutants and the atmosphere, certain pollutants, such as secondary pollutants, may not decreasing trends in emissions of their precursor pollutants [187].

In the national context, Portugal also tracks European trends, with emissions reductions in the general panorama, highlighting critical points such as the Lisbon metropolitan area and surrounding sub-regions, as well as regions with relatively low emissions of atmospheric pollutants such as Alentejo and Algarve.

In the field of public health, in relation to the European panorama, mortality has declined significantly since 2000, even though it still has high values. From 2000 to 2013, the European Union showed a 21% reduction in mortality, with Estonia's share in this reduction, with a 30% reduction in mortality [187]. According to the latest report produced by the OECD, about one third of the number of deaths in men and about 24% of the number of deaths in women. The main causes of mortality are cancer, lung cancer, colon and prostate cancer, for men, and cancer of the breast, colon and lungs, for women [187].

With the aging of the population, values related to respiratory diseases tend to increase, consequently increasing the values related to mortality associated with respiratory diseases [187].

Globally, about 3% of cancer deaths and about 5% of lung cancer deaths are attributed to particles (PM).
In the European context, figures of around 2% and 3% are presented for the causes of deaths reported, respectively [188].
In 2010, worldwide, 3.1 million deaths and about 3.1% of disability-adjusted life years were recorded, indicating the cause as exposure to fine particles (PM2.5) [188, 189].

In the case of Portugal, different trends are pointed out, depending on the cause of mortality and the region in question. In general, decreasing trends in mortality associated with the causes under study are presented mainly in the Alentejo and Algarve regions, these trends being also registered in fewer causes of mortality in other regions.
The regions of the North, Center and Metropolitan Area of Lisbon are regions with high mortality values, compared to the other regions.

With this qualitative analysis, of national focus, in the field of emissions, concentrations and air quality, and health indicators, it was possible to identify: critical areas that present high concentrations of atmospheric pollutants; areas with high pollutant emissions; and to the most affected areas in public health regarding mortality associated with causes that may involve air pollution issues. Possible future actions or intensification of current actions to mitigate emissions of atmospheric pollutants, should focus on the Lisbon, North and Center regions, in particular the districts of Lisbon, Setubal and Porto, which are some of the districts with greater population density and intensity of the manufacturing sector.

7. FURTHER DEVELOPMENTS

This study intended to perform a critical evaluation review of air quality indicators and its impacts in the field of public health. Nonetheless, are presented below a few topics of interest to be developed in the future, which will answer certain questions raised throughout this work:

i. Study of the relationship between transport processes, chemical transformation of pollutants and their deposition, with climatic conditions;

ii. In-depth study of the health symptoms, acute and chronic diseases associated with air pollution;

iii. Aggregation and revision of emission values of air pollutants of all years since the beginning of the implementation of mitigating measures of pollutant emissions;

vi. Cross-sectional analysis from the baseline pollutant emissions up to 2030 emissions reduction and its health contributions.

8. References

1. Valente, J., et al., *Individual exposure to air pollutants in a portuguese urban industrialized area.* J Toxicol Environ Health A, 2014. **77**(14-16): p. 888-99.
2. Costa, S., et al., *Integrating Health on Air Quality Assessment-Review Report on Health Risks of Two Major European Outdoor Air Pollutants: PM and NO2.* J Toxicol Environ Health B Crit Rev, 2014. **17**(6): p. 307-40.
3. Squizzato, S., et al., *Urban air quality in a mid-size city - PM2.5 composition, sources and identification of impact areas: From local to long range contributions.* Atmospheric Research, 2017. **186**: p. 51-62.
4. Gorai, A.K., et al., *An innovative approach for determination of air quality health index.* Sci Total Environ, 2015. **533**: p. 495-505.
5. AR, Q.-B.d.D.O.s.a.Q.d., *Indice de Qualidade do Ar*, in *Indices*. 2017.
6. Do Vale, I.D., A.S. Vasconcelos, and G.O. Duarte, *Inhalation of particulate matter in three different routes for the same OD pair: A case study with pedestrians in the city of Lisbon.* Journal of Transport & Health, 2015. **2**(4): p. 474-482.
7. Cox, W. *Largest Cities in the World: 2016*. 2016 [cited 2017 04/2017]; Available from: http://www.newgeography.com/content/005219-largest-cities-world-2016.
8. Braga, A.L.F., et al., *Associação entre poluição atmosférica e doenças respiratórias e cardiovasculares na cidade de Itabira, Minas Gerais, Brasil.* Cadernos de Saúde Pública, 2007. **23**: p. S570-S578.
9. GBD 2015 Mortality and Causes of Death Collaborators, *Global, regional, and national life expectancy, all-cause mortality, and cause-specific mortality for 249 causes of death, 1980-2015: a systematic analysis for the Global Burden of Disease Study 2015.* Lancet, 2016. **388**(10053): p. 1459-1544.
10. National Park Service USA *Pollutant Emissions. Available: https://www.nature.nps.gov/air/aqbasics/sources.cfm*. 2017.
11. Leaner, A. *Unit 11: Atmospheric Pollution*. The Habitalbe Planet - A Systems Approach to Environment Science 2017; Available from: https://www.learner.org/courses/envsci/unit/text.php?unit=11&secNum=0.
12. Hoyle, B. *Air Pollution - Criteria pollutants*. Science & Philosophy: Adrenoceptor (adrenoreceptor; adrenergic receptor) to Ambient 2002 [cited 2017; Available from: http://science.jrank.org/pages/154/Air-Pollution.html.
13. National Park Service USA *Air pollution sources and effects. Available: https://www.nature.nps.gov/air/studies/air_toxics/dragonfly/images/emissions-transport-deposition_annotated.jpg*. 2017.
14. Eckel, S.P., et al., *Modification of the association between ambient air pollution and lung function by frailty status among older adults in the Cardiovascular Health Study.* Am J Epidemiol, 2012. **176**(3): p. 214-23.
15. McGill, G., L.O. Oyedele, and K. McAllister, *An investigation of indoor air quality, thermal comfort and sick building syndrome symptoms in UK energy efficient homes.* Smart and Sustainable Built Environment, 2015. **4**(3): p. 329-348.
16. Garcia-Esquinas, E. and F. Rodriguez-Artalejo, *Environmental Pollutants, Limitations in Physical Functioning, and Frailty in Older Adults.* Curr Environ Health Rep, 2017. **4**(1): p. 12-20.
17. Garcia-Esquinas, E., et al., *Association of lead and cadmium exposure with frailty in US older adults.* Environ Res, 2015. **137**: p. 424-31.
18. Nunes, R.A., et al., *Particulate matter in rural and urban nursery schools in Portugal.* Environ Pollut, 2015. **202**: p. 7-16.
19. Carreiro-Martins, P., et al., *Chronic respiratory diseases and quality of life in elderly nursing home residents.* Chronic Respiratory Disease, 2016.

20. Carreiro-Martins, P., et al., *Effect of indoor air quality of day care centers in children with different predisposition for asthma.* Pediatr Allergy Immunol, 2015.
21. Carreiro-Martins, P., et al., *CO2 concentration in day care centres is related to wheezing in attending children.* European Journal of Pediatrics, 2014.
22. Mendes, A., et al., *The impact of indoor air quality and contaminants on respiratory health of older people living in long-term care residences in Porto.* Age Ageing, 2016. **45**(1): p. 136-42.
23. Bentayeb, M., et al., *Adverse respiratory effects of outdoor air pollution in the elderly.* Int J Tuberc Lung Dis, 2012. **16**(9): p. 1149-61.
24. Bentayeb, M., et al., *Indoor air pollution and respiratory health in the elderly.* J Environ Sci Health A Tox Hazard Subst Environ Eng, 2013. **48**(14): p. 1783-9.
25. Simoni, M., et al., *Adverse effects of outdoor pollution in the elderly.* J Thorac Dis, 2015. **7**(1): p. 34-45.
26. Simoni, M., et al., *Indoor air pollution and respiratory health in the elderly.* European Respiratory Journal, 2003. **21**(Supplement 40): p. 15S-20s.
27. Ancona, C., et al., *Mortality and morbidity in a population exposed to multiple sources of air pollution: A retrospective cohort study using air dispersion models.* Environmental Research, 2015. **137**: p. 467-474.
28. Krzyzanowski, M., A. Cohen, and R. Anderson, *Quantification of health effects of exposure to air pollution.* Occupational and Environmental Medicine, 2002. **59**(12): p. 791-793.
29. Agency, E.E., *Air Quality in Europe - 2016 Report*, in *Air Quality in Europe*. 2016, European Environment Agency: Copenhagen.
30. Xu, Y., W. Yang, and J. Wang, *Air quality early-warning system for cities in China.* Atmospheric Environment, 2017. **148**: p. 239-257.
31. World Health Organization, *Review of Evidence on Health Aspects of Air Pollution-REVIHAAP*. 2013: WHO, Copenhagen.
32. World Health Organization, *Press release N. 221 - IARC: Outdoor air pollution a leading environmental cause of cancer deaths*, International Agency for Research on Cancer - World Health Organization, Editor. 2013.
33. *<Health-risks-of-air-pollution-in-Europe-HRAPIE-project,-Recommendations-for-concentrationresponse-functions-for-costbenefit-analysis-of-particulate-matter,-ozone-and-nitrogen-dioxide.pdf>.*
34. World Health Organization, *Ambient (outdoor) air quality and health - Fact sheet N°313:* http://www.who.int/mediacentre/factsheets/fs313/en/. 2014.
35. World Health Organization, *Economic Cost of the Health Impact of Air Pollution in Europe: Clean Air, Health and Wealth*. 2015: WHO, Copenhagen.
36. World Health Organization, *WHO Guidelines for Indoor Air Quality: Dampness and Mould*. 2009, WHO Regional Office for Europe: Copenhagen, Denmark.
37. World Health Organization, *WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide - Global update 2005 - Summary of risk assessment*. 2006, World Health Organization,.
38. World Health Organization, *WHO Guidelines for Indoor Air Quality: Selected Pollutants, WHO Regional Office for Europe, Copenhagen, Denmark.* 2010.
39. World Health Organization, *Health effects of particulate matter*. 2013, World Health Organization - ISBN: 978 92 890 0001 7: Copenhagen, Denmark.
40. Anderson, H.R., *Air pollution and mortality: A history.* Atmospheric Environment, 2009. **43**(1): p. 142-152.
41. Nemery, B., P.H.M. Hoet, and A. Nemmar, *The Meuse Valley fog of 1930: an air pollution disaster.* The Lancet, 2001. **357**(9257): p. 704-708.
42. Kamrin, M.A., *Donora: Air Pollution Episode A2 - Wexler, Philip*, in *Encyclopedia of Toxicology (Third Edition)*. 2014, Academic Press: Oxford. p. 222-223.

43. Rodriguez, Y.R., *Great Smog of London A2 - Wexler, Philip*, in *Encyclopedia of Toxicology (Third Edition)*. 2014, Academic Press: Oxford. p. 796-797.
44. Gao, J., et al., *Haze, public health and mitigation measures in China: A review of the current evidence for further policy response.* Science of The Total Environment, 2017. **578**: p. 148-157.
45. Balbus, J.M., et al., *Implications of global climate change for the assessment and management of human health risks of chemicals in the natural environment.* Environmental Toxicology and Chemistry, 2013. **32**(1): p. 62-78.
46. Gehring, U., et al., *Long-term exposure to ambient air pollution and cardiopulmonary mortality in women.* Epidemiology, 2006. **17**(5): p. 545-51.
47. Hoek, G., et al., *Association between mortality and indicators of traffic-related air pollution in the Netherlands: a cohort study.* Lancet, 2002. **360**(9341): p. 1203-9.
48. Nafstad, P., et al., *Urban air pollution and mortality in a cohort of Norwegian men.* Environmental Health Perspectives, 2004. **112**(5): p. 610-615.
49. Beelen, R., et al., *Long-Term Effects of Traffic-Related Air Pollution on Mortality in a Dutch Cohort (NLCS-AIR Study).* Environmental Health Perspectives, 2008. **116**(2): p. 196-202.
50. Ashmore, M., *Air Pollution☆*, in *Reference Module in Life Sciences*. 2017, Elsevier.
51. Cárdenas Rodríguez, M., L. Dupont-Courtade, and W. Oueslati, *Air pollution and urban structure linkages: Evidence from European cities.* Renewable and Sustainable Energy Reviews, 2016. **53**: p. 1-9.
52. Thurston, G.D., *Outdoor Air Pollution: Sources, Atmospheric Transport, and Human Health Effects A2 - Quah, Stella R*, in *International Encyclopedia of Public Health (Second Edition)*. 2017, Academic Press: Oxford. p. 367-377.
53. Ruckerl, R., et al., *Health effects of particulate air pollution: A review of epidemiological evidence.* Inhal Toxicol, 2011. **23**(10): p. 555-92.
54. Correia, A.W., et al., *The Effect of Air Pollution Control on Life Expectancy in the United States: An Analysis of 545 US counties for the period 2000 to 2007.* Epidemiology (Cambridge, Mass.), 2013. **24**(1): p. 23-31.
55. C. Silveira, M.L., P. Roebeling, J. Ferreira, S. Costa, J. P. Teixeira, C. Borrego & A. I. Miranda, *Economic evaluation of air pollution impacts on human health: an overview of applied methodologies.* WIT Transactions on Ecology and The Environment, 2017. **198**: p. 181-192.
56. Pervin, T., U.-G. Gerdtham, and C.H. Lyttkens, *Societal costs of air pollution-related health hazards: A review of methods and results.* Cost Effectiveness and Resource Allocation, 2008. **6**(1): p. 19.
57. Eliasson, J., et al., *The Stockholm congestion – charging trial 2006: Overview of effects.* Transportation Research Part A: Policy and Practice, 2009. **43**(3): p. 240-250.
58. Aunan, K., *Exposure-response functions for health effects of air pollutants based on epidemiological findings.* Risk Anal, 1996. **16**(5): p. 693-709.
59. Euler, G.L., et al., *Chronic obstructive pulmonary disease symptom effects of long-term cumulative exposure to ambient levels of total oxidants and nitrogen dioxide in California Seventh-Day Adventist residents.* Arch Environ Health, 1988. **43**(4): p. 279-85.
60. S o/yseth, V., et al., *Relation of exposure to airway irritants in infancy to prevalence of bronchial hyper-responsiveness in schoolchildren.* The Lancet. **345**(8944): p. 217-220.
61. Ambiente, A.P.d. *Inventário Nacional de Emissões Atmosféricas (INERPA)*. 2017; Available from: https://www.apambiente.pt/index.php?ref=17&subref=150.
62. Ambiente, A.P.d. *Distribuição Espacial de Emissões (histórico)*. Instrumentos 2017 [cited 2017; Available from: https://www.apambiente.pt/index.php?ref=17&subref=150&sub2ref=1407.
63. Maciel, F.B.A.T.P.P.T.T.C.P.V.G.T.S.H., *Alocação Espacial de Emissões em 2005*. 2008, Agência Portuguesa do Ambiente: Agência Portuguesa do Ambiente.

64. Território, A.P.d.A.-M.d.A.e.d.O.d., *Emissões de Poluentes Atmosféricos por Concelho 2008:: Gases acidificantes e eutrofizantes, precursores de ozono, partículas, metais pesados e gases com efeito de estufa*, A.P.d. Ambiente, Editor. 2010, Agência Portuguesa do Ambiente: Agência Portuguesa do Ambiente.
65. Nani Guarieiro, L.L. and A.L. Nani Guarieiro, *Vehicle Emissions: What Will Change with Use of Biofuel?, Biofuels - Economy, Environment and Sustainability, Prof. Zhen Fang (Ed.), ISBN: 978-953-51-0950-1, InTech, DOI: 10.5772/52513. Available from:* http://www.intechopen.com/books/biofuels-economy-environment-and-sustainability/vehicle-emissions-what-will-change-with-use-of-biofuel-. 2013.
66. Yang, L., et al., *Biomarkers of the health outcomes associated with ambient particulate matter exposure.* Sci Total Environ, 2017. **579**: p. 1446-1459.
67. Kampa, M. and E. Castanas, *Human health effects of air pollution.* Environ Pollut, 2008. **151**(2): p. 362-7.
68. Jason, *Penetration of different-sized particles into the human body.* 2016, Camfil APC: Clean Air Matters.
69. Zheng, Y., et al., *Traffic-derived particulate matter exposure and histone H3 modification: A repeated measures study.* Environ Res, 2017. **153**: p. 112-119.
70. Putaud, J.P., et al., *A European aerosol phenomenology – 3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe.* Atmospheric Environment, 2010. **44**(10): p. 1308-1320.
71. Giovanni Invernizzi, A.R., *Conoscere per decidere: l'inquinamento da traffico veicolare di prossimità e il suo impatto sulla salute*. 2011.
72. Alcántara, V., E. Padilla, and M. Piaggio, *Nitrogen oxide emissions and productive structure in Spain: An input–output perspective.* Journal of Cleaner Production, 2017. **141**: p. 420-428.
73. Deng, L., et al., *Release of nitrogen oxides during combustion of model coals.* Fuel, 2016. **175**: p. 217-224.
74. Miller, B., *Nitrogen oxides formation and control.* 2015: p. 243-280.
75. Vaz, S.G., *Ambiente em Portugal*. 2016: Fundação Manuel Dos Santos (FFMS).
76. Lin, B.-L., et al., *Modelling a global biogeochemical nitrogen cycle in terrestrial ecosystems.* Ecological Modelling, 2000. **135**(1): p. 89-110.
77. Henschel, S., et al., *Trends of nitrogen oxides in ambient air in nine European cities between 1999 and 2010.* Atmospheric Environment, 2015. **117**: p. 234-241.
78. Meulenbelt, J., *Nitrogen and nitrogen oxides.* Medicine, 2012. **40**(3): p. 139.
79. Samoli, E., et al., *Short-term effects of nitrogen dioxide on mortality: an analysis within the APHEA project.* Eur Respir J, 2006. **27**(6): p. 1129-38.
80. Lin, S., et al., *Impact of NOx emissions reduction policy on hospitalizations for respiratory disease in New York State.* Journal of Exposure Science and Environmental Epidemiology, 2013. **23**(1): p. 73-80.
81. Li, X.B., et al., *Three-dimensional investigation of ozone pollution in the lower troposphere using an unmanned aerial vehicle platform.* Environ Pollut, 2017. **224**: p. 107-116.
82. Zhong, L., C.S. Lee, and F. Haghighat, *Indoor ozone and climate change.* Sustainable Cities and Society, 2017. **28**: p. 466-472.
83. Jerrett, M., et al., *Ambient ozone and incident diabetes: A prospective analysis in a large cohort of African American women.* Environ Int, 2017. **102**: p. 42-47.
84. Bass, V., et al., *Ozone induces glucose intolerance and systemic metabolic effects in young and aged brown Norway rats.* Toxicology and Applied Pharmacology, 2013. **273**(3): p. 551-560.
85. Hosseiniebalam, F. and O. Ghaffarpasand, *The effects of emission sources and meteorological factors on sulphur dioxide concentration of Great Isfahan, Iran.* Atmospheric Environment, 2015. **100**: p. 94-101.

86. Kanaroglou, P.S., et al., *Estimation of sulfur dioxide air pollution concentrations with a spatial autoregressive model.* Atmospheric Environment, 2013. **79**: p. 421-427.
87. Chen, F., et al., *The effects of Sulphur dioxide on acute mortality and years of life lost are modified by temperature in Chengdu, China.* Sci Total Environ, 2017. **576**: p. 775-784.
88. Smith, S.J., H. Pitcher, and T.M.L. Wigley, *Global and regional anthropogenic sulfur dioxide emissions.* Global and Planetary Change, 2001. **29**(1–2): p. 99-119.
89. Wikipedia. *Acid Rain*. 2017 [cited 2017; Available from: https://en.wikipedia.org/wiki/Acid_rain.
90. Petruzzi, S., B. Musi, and G. Bignami, *Acute and chronic sulphur dioxide (SO2) exposure: an overview of its effects on humans and laboratory animals.* Ann Ist Super Sanita, 1994. **30**(2): p. 151-6.
91. Palmer, J. and K. Von Rueden, *Carbon Monoxide Poisoning and Pregnancy: Critical Nursing Interventions.* Journal of Emergency Nursing, 2015. **41**(6): p. 479-483.
92. Aldossary, M., et al., *Carbon monoxide toxicity in Dammam, KSA: Retrospective study.* Egyptian Journal of Forensic Sciences, 2015. **5**(1): p. 36-38.
93. Yarragunta, Y., S. Srivastava, and D. Mitra, *Validation of lower tropospheric carbon monoxide inferred from MOZART model simulation over India.* Atmospheric Research, 2017. **184**: p. 35-47.
94. Immenschuh, S., et al., *Heme as a Target for Therapeutic Interventions.* Frontiers in Pharmacology, 2017. **8**: p. 15.
95. Maga, M., et al., *Influence of air pollution on exhaled carbon monoxide levels in smokers and non-smokers. A prospective cross-sectional study.* Environmental Research, 2017. **152**: p. 496-502.
96. Veen, M., *Carbon Monoxide Poisoning Caused by Water Pipe Smoking: A Case Series.* The Journal of Emergency Medicine, 2016. **51**(3): p. e41-e44.
97. Hashemzaei, M., et al., *Effects of resveratrol on carbon monoxide-induced cardiotoxicity in rats.* Environmental Toxicology and Pharmacology, 2016. **46**: p. 110-115.
98. Park, S.-H., et al., *A Study for Health Hazard Evaluation of Methylene Chloride Evaporated from the Tear Gas Mixture.* Safety and Health at Work, 2010. **1**(1): p. 98-101.
99. Wang, C.-Z., A. Li, and Z.-C. Yang, *The Pathophysiology of Carbon Monoxide Poisoning and Acute Respiratory Failure in a Sheep Model with Smoke Inhalation Injury.* Chest, 1990. **97**(3): p. 736-742.
100. Kampa, M. and E. Castanas, *Human health effects of air pollution.* Environmental Pollution, 2008. **151**(2): p. 362-367.
101. OECD, *OECD Health Statistics 2016 Definitions, Sources and Methods.* 2016.
102. Bianchi, F., et al., *Contribution of methane to aerosol carbon mass.* Atmospheric Environment, 2016. **141**: p. 41-47.
103. Van Amstel, A., *Methane. A review.* Journal of Integrative Environmental Sciences, 2012. **9**(sup1): p. 5-30.
104. Pérez-Barbería, F.J., *Scaling methane emissions in ruminants and global estimates in wild populations.* Science of The Total Environment, 2017. **579**: p. 1572-1580.
105. Yu, X., et al., *Growing season methane emissions from a permafrost peatland of northeast China: Observations using open-path eddy covariance method.* Atmospheric Environment, 2017. **153**: p. 135-149.
106. Prajapati, P. and E.A. Santos, *Measurements of methane emissions from a beef cattle feedlot using the eddy covariance technique.* Agricultural and Forest Meteorology, 2017. **232**: p. 349-358.
107. El-Fadel, M. and M. Massoud, *Methane emissions from wastewater management.* Environmental Pollution, 2001. **114**(2): p. 177-185.
108. Planets, G.I.o.C.a. *Lesson 2-1: Trends in Atmospheric Methane Concentrations*. Education: Global Methane Inventory 2010 [cited 2017; Available from: https://icp.giss.nasa.gov/education/methane/lessons/lesson2_1/charts.html.

109. Lawrence, B.A., et al., *Wetland invasion by Typha × glauca increases soil methane emissions.* Aquatic Botany, 2017. **137**: p. 80-87.
110. Shipman, M.A. and M.D. Symes, *Recent progress towards the electrosynthesis of ammonia from sustainable resources.* Catalysis Today, 2017. **286**: p. 57-68.
111. Borsari, V. and J.V.d. Assunção, *Ammonia emissions from a light-duty vehicle.* Transportation Research Part D: Transport and Environment, 2017. **51**: p. 53-61.
112. Technology, O.S. *Nitrogen, Cycling Down*. Holistic Organic Microbiology [cited 2017.
113. Hendriks, C., et al., *Ammonia emission time profiles based on manure transport data improve ammonia modelling across north western Europe.* Atmospheric Environment, 2016. **131**: p. 83-96.
114. Science, N. and T.C.A.Q.R. Subcommittee, *Atmospheric Ammonia: Sources and Fate : a Review of Ongoing Federal Research and Future Needs*. 2000: The Committee.
115. Künzli, N., et al., *Public-health impact of outdoor and traffic-related air pollution: a European assessment.* The Lancet, 2000. **356**(9232): p. 795-801.
116. Newby, D.E., et al., *Expert position paper on air pollution and cardiovascular disease.* Eur Heart J, 2014.
117. Balmes, J.R., J.M. Fine, and D. Sheppard, *Symptomatic bronchoconstriction after short-term inhalation of sulfur dioxide.* Am Rev Respir Dis, 1987. **136**(5): p. 1117-21.
118. Kagawa, J., *Evaluation of biological significance of nitrogen oxides exposure.* Tokai J Exp Clin Med, 1985. **10**(4): p. 348-53.
119. Ghio, A.J. and Y.C. Huang, *Exposure to concentrated ambient particles (CAPs): a review.* Inhal Toxicol, 2004. **16**(1): p. 53-9.
120. Uysal, N. and R.M. Schapira, *Effects of ozone on lung function and lung diseases.* Curr Opin Pulm Med, 2003. **9**(2): p. 144-50.
121. Chauhan, A.J., et al., *Exposure to nitrogen dioxide (NO2) and respiratory disease risk.* Rev Environ Health, 1998. **13**(1-2): p. 73-90.
122. Rastogi, S.K., et al., *A cross-sectional study of pulmonary function among workers exposed to multimetals in the glass bangle industry.* American Journal of Industrial Medicine, 1991. **20**(3): p. 391-399.
123. Kuo, C.Y., et al., *Accumulation of chromium and nickel metals in lung tumors from lung cancer patients in Taiwan.* J Toxicol Environ Health A, 2006. **69**(14): p. 1337-44.
124. Wegmann, M., et al., *NO2-induced airway inflammation is associated with progressive airflow limitation and development of emphysema-like lesions in C57BL/6 mice.* Experimental and Toxicologic Pathology, 2005. **56**(6): p. 341-350.
125. European Environment Agency - European Commission *Health impacts of air pollution. Infographic Prod-ID: INF-12-en Published 13 Jun 2014 Last modified 22 Apr 2016. Available:* https://www.eea.europa.eu/signals/signals-2013/infographics/health-impacts-of-air-pollution/view. 2014.
126. Mark Edwards, P.E., *Algae Secrets*. 2016: AlgaeIndustryMagazine.
127. Riousset, P., C. Flachsland, and M. Kowarsch, *Global environmental assessments: Impact mechanisms.* Environmental Science & Policy, 2017.
128. Bias, T.K. and C.G. Abildso, *Measuring policy and related effects of a health impact assessment related to connectivity.* Preventive Medicine, 2017. **95**: p. S92-S94.
129. Marsh, K., et al., *Expanding Health Technology Assessments to Include Effects on the Environment.* Value in Health, 2016. **19**(2): p. 249-254.
130. Hambling, T., P. Weinstein, and D. Slaney, *A Review of Frameworks for Developing Environmental Health Indicators for Climate Change and Health.* International Journal of Environmental Research and Public Health, 2011. **8**(7): p. 2854-2875.
131. System, I.-I.E.H.I.A. *The DPSEEA framework*. [cited 2017; Available from: http://www.integrated-assessment.eu/eu/guidebook/dpseea_framework.html.

132. Ajanovic, A. and R. Haas, *The impact of energy policies in scenarios on GHG emission reduction in passenger car mobility in the EU-15.* Renewable and Sustainable Energy Reviews, 2017. **68, Part 2**: p. 1088-1096.
133. West, J.J., et al., *Co-benefits of Global Greenhouse Gas Mitigation for Future Air Quality and Human Health.* Nature climate change, 2013. **3**(10): p. 885-889.
134. Change, I.P.o.C., *IPCC Special Report - Emissions Scenarios - Summary for Policymakers*. IPCC Special Report. 2000, the Intergovernmental Panel on Climate Change.
135. atticusdigital, *A global emissions reduction scenarios*. 2017, CCS TLM: CCS TLM.
136. Thunis, P., et al., *Analysis of model responses to emission-reduction scenarios within the CityDelta project.* Atmospheric Environment, 2007. **41**(1): p. 208-220.
137. Schöpp, W., et al., *Uncertainty analysis of emission estimates in the RAINS integrated assessment model.* Environmental Science & Policy, 2005. **8**(6): p. 601-613.
138. Capros, P., et al., *European Energy and CO 2 Emissions Trends to 2020: PRIMES model v.2.* Bulletin of Science, Technology & Society, 1999. **19**(6): p. 474-492.
139. Degraeuwe, B., et al., *Impact of passenger car NOx emissions and NO2 fractions on urban NO2 pollution – Scenario analysis for the city of Antwerp, Belgium.* Atmospheric Environment, 2016. **126**: p. 218-224.
140. Tuinstra, W., *Preparing for the European Thematic Strategy on air pollution: at the interface between science and policy.* Environmental Science & Policy, 2007. **10**(5): p. 434-444.
141. Markus Amann, I.B., Janusz Cofala, Frantisek Gyarfas, Chris Heyes, Zbigniew Klimont, Wolfgang Schöpp, Wilfried Winiwarter *Baseline Scenarios for the Clean Air for Europe (CAFE) Programme* in *Development of the Baseline and Policy Scenarios and Integrated Assessment Modelling Framework for the Clean Air for Europe (CAFE) Programme*, I.I.f.A.S.A. Schlossplatz, Editor. 2005, European Commission Directorate General for Environment, Directorate C – Environment and Health
142. Commision, E. *Air Quality - Environment*. Environment 2016 [cited 2017 05/2017]; Available from: http://ec.europa.eu/environment/air/quality/index.htm.
143. Commission, E. *The Clean Air Policy Package*. Environmenta - Air 2016 08/06/2016 [cited 2017 05/2017]; Available from: http://ec.europa.eu/environment/air/clean_air_policy.htm.
144. Commision, E. *Implementation of the National Emission Ceilings Directive*. Emissions of Air Pollutants 2017 21/03/2017 [cited 2017 08/2017]; Available from: http://ec.europa.eu/environment/air/pollutants/implem_nec_directive.htm.
145. Commission, E. *Guidance on National Air Pollution Control Programmes*. Emissions of Air Pollutants 2017 13/03/2017 [cited 2017 05/2017]; Available from: http://ec.europa.eu/environment/air/pollutants/national_control_programme.htm.
146. Commission, E. *Implementation of ambient air quality legislation* Air Quality 2016 08/06/2016 [cited 2017 05/2017]; Available from: http://ec.europa.eu/environment/air/quality/legislation/index.htm.
147. Ambiente, A.P.d. *Qualidade do Ar Ambiente*. Politicas - Ar 2017 [cited 2017 05/2017]; Available from: http://www.apambiente.pt/index.php?ref=16&subref=82&sub2ref=316.
148. Territory, M.o.E.a.o.t.O.t., *Decreto-Lei nº102/2010*, in *102*. 2010: Diário da República, 1.ª série — N.º 186 — 23 de Setembro de 2010
149. (EEA), E.E.A. *Air pollution - State and impacts (Portugal)*. Air pollution 2010 21/03/2015 [cited 2017 05/2017]; Available from: https://www.eea.europa.eu/soer/countries/pt/air-pollution-state-and-impacts-portugal.
150. Contemporâneo, P.-B.d.D.P. *Gas emissions*. 2014 [cited 2017.
151. Contemporâneo, P.-B.D.P. *Acidification potential: total, of the industries and of the households - Portugal*. 2014 [cited 2017; Available from:

http://www.pordata.pt/en/Portugal/Acidification+potential+total++of+the+industries+and+of+the+households-1073.
152. Agência Portuguesa do Ambiente - Departamento de Alterações Climáticas, A.e.R., *Emissões de Poluentes Atmosféricos por Concelho 2009:: Gases acidificantes e eutrofizantes, precursores de ozono, partículas, metais pesados e gases com efeito de estufa*. 2011: Amadora.
153. Filomena Boavida, A.T.P., Pedro Torres, Teresa Costa Pereira, Vitor Gois, Tiago Seabra, Hugo Maciel, *Alocação Espacial de Emissões em 2003*. 2008, Instituto do ambiente - Ministério do Ambiente, do Ordenamento do Território e do Desenvolvimento Regional.
154. Gao, S., et al., *Spatial statistics of atmospheric particulate matter in China.* Atmospheric Environment, 2016. **134**: p. 162-167.
155. Wang, X., et al., *Chemical composition and sources of PM10 and PM2.5 aerosols in Guangzhou, China.* Environ Monit Assess, 2006. **119**(1-3): p. 425-39.
156. Yin, Q., et al., *Estimation of daily PM2.5 concentration and its relationship with meteorological conditions in Beijing.* Journal of Environmental Sciences, 2016. **48**(Supplement C): p. 161-168.
157. Che, H., et al., *Aerosol optical properties under the condition of heavy haze over an urban site of Beijing, China.* Environ Sci Pollut Res Int, 2015. **22**(2): p. 1043-53.
158. Russo, A., et al., *Neural network forecast of daily pollution concentration using optimal meteorological data at synoptic and local scales.* Atmospheric Pollution Research, 2015. **6**(3): p. 540-549.
159. Filonchyk, M., et al., *A study of PM2.5 and PM10 concentrations in the atmosphere of large cities in Gansu Province, China, in summer period.* Journal of Earth System Science, 2016. **125**(6): p. 1175-1187.
160. Index, W.A.Q. *Why is PM2.5 often higher than PM10? Is PM10 still a relevant measure?* 2017; Available from: http://aqicn.org/faq/2013-02-02/why-is-pm25-often-higher-than-pm10/#.
161. conhecimento, R.C.d. *SIRIC - Sistema Integrado do Registo e Identificação Civil*. 2017; Available from: http://www.rcc.gov.pt/Directorio/Temas/MA/Paginas/SIRIC---Sistema-Integrado-do-Registo-e-Identifica%C3%A7%C3%A3o-Civil-.aspx.
162. Saúde, D.-G.d. *SICO - Sistema de Informação dos Certificados de Óbito*. 2017; Available from: https://www.dgs.pt/paginas-de-sistema/saude-de-a-a-z/sico-sistema-de-informacao-dos-certificados-de-obito.aspx.
163. Estatística, I.N.d., *Causas de morte 2015*. 2017, Instituto Nacional de Estatística: Instituto Nacional de Estatística. p. 188.
164. Oliveira, I.T.d., *Mortalidade: Compressão, Deslocamento e Causas de Morte (Portugal 1950-2005)*, in *Revista de Estudos Demográficos*. 2010.
165. MD-Health. *Common Circulatory System Diseases and All You Need to Know*. Heart Diseases 2017 [cited 2017; Available from: http://www.md-health.com/Circulatory-System-Diseases.html.
166. GERTLER, M.M., et al., *Ischemic Heart Disease.* Insulin, Carbohydrate, and Lipid Interrelationships, 1972. **46**(1): p. 103-111.
167. Athanazio, R., *Airway disease: similarities and differences between asthma, COPD and bronchiectasis.* Clinics, 2012. **67**(11): p. 1335-1343.
168. Kokic, M. *Chronic respiratory diseases*. 2017 [cited 2017; Available from: http://www.who.int/respiratory/en/.
169. Guarnieri, M. and J.R. Balmes, *Outdoor air pollution and asthma.* The Lancet, 2014. **383**(9928): p. 1581-1592.
170. Mann, J.K., et al., *Short-term effects of air pollution on wheeze in asthmatic children in Fresno, California.* Environ Health Perspect, 2010. **118**(10): p. 1497-502.
171. Meng, Y.Y., et al., *Outdoor air pollution and uncontrolled asthma in the San Joaquin Valley, California.* J Epidemiol Community Health, 2010. **64**(2): p. 142-7.

172. Yang, I.V., C.A. Lozupone, and D.A. Schwartz, *The environment, epigenome, and asthma.* Journal of Allergy and Clinical Immunology, 2017. **140**(1): p. 14-23.
173. Baccarelli, A., et al., *Rapid DNA methylation changes after exposure to traffic particles.* Am J Respir Crit Care Med, 2009. **179**(7): p. 572-8.
174. Bellavia, A., et al., *DNA hypomethylation, ambient particulate matter, and increased blood pressure: findings from controlled human exposure experiments.* J Am Heart Assoc, 2013. **2**(3): p. e000212.
175. Bind, M.A., et al., *Air pollution and gene-specific methylation in the Normative Aging Study: association, effect modification, and mediation analysis.* Epigenetics, 2014. **9**(3): p. 448-58.
176. Guo, L., et al., *Effects of short-term exposure to inhalable particulate matter on DNA methylation of tandem repeats.* Environ Mol Mutagen, 2014. **55**(4): p. 322-35.
177. Sofer, T., et al., *Exposure to airborne particulate matter is associated with methylation pattern in the asthma pathway.* Epigenomics, 2013. **5**(2): p. 147-54.
178. De Prins, S., et al., *Influence of ambient air pollution on global DNA methylation in healthy adults: A seasonal follow-up.* Environment International, 2013. **59**: p. 418-424.
179. Bind, M.A., et al., *Air pollution and markers of coagulation, inflammation, and endothelial function: associations and epigene-environment interactions in an elderly cohort.* Epidemiology, 2012. **23**(2): p. 332-40.
180. Chen, P., et al., *Differential methylation of genes in individuals exposed to maternal diabetes in utero.* Diabetologia, 2017. **60**(4): p. 645-655.
181. Matthews, D.R., et al., *Homeostasis model assessment: insulin resistance and beta-cell function from fasting plasma glucose and insulin concentrations in man.* Diabetologia, 1985. **28**(7): p. 412-9.
182. Dodge, J.E., et al., *De novo methylation of MMLV provirus in embryonic stem cells: CpG versus non-CpG methylation.* Gene, 2002. **289**(1–2): p. 41-48.
183. Bernstein, B.E., A. Meissner, and E.S. Lander, *The Mammalian Epigenome.* Cell, 2007. **128**(4): p. 669-681.
184. Lu, Z.-N., et al., *The dynamic relationship between environmental pollution, economic development and public health: Evidence from China.* Journal of Cleaner Production, 2017. **166**(Supplement C): p. 134-147.
185. Pautrel, X., *Reconsidering the Impact of the Environment on Long-run Growth when Pollution Influences Health and Agents have a Finite-lifetime.* Environmental and Resource Economics, 2008. **40**(1): p. 37-52.
186. Katrakilidis, C., I. Kyritsis, and V. Patsika, *The dynamic linkages between economic growth, environmental quality and health in Greece.* Applied Economics Letters, 2016. **23**(3): p. 217-221.
187. Guerreiro, C.B.B., V. Foltescu, and F. de Leeuw, *Air quality status and trends in Europe.* Atmospheric Environment, 2014. **98**(Supplement C): p. 376-384.
188. Mirabelli, M.C., et al., *Outdoor PM2.5, Ambient Air Temperature, and Asthma Symptoms in the Past 14 Days among Adults with Active Asthma.* Environ Health Perspect, 2016. **124**(12): p. 1882-1890.
189. OECD/EU, *Health at a Glance: Europe 2016*. OECD Publishing.

APPENDIX: PUBLICATIONS

POSTER PRESENTATION

Instituto Nacional de Saúde Doutor Ricardo Jorge

REPÚBLICA PORTUGUESA SAÚDE

SNS SERVIÇO NACIONAL DE SAÚDE

FUTURAR

Air quality in Portugal in 2030 – a policy support

supported by PTDC/AAG-MAA/2569/2014, POCI-01-0145-FEDER-016752

FCT Fundação para a Ciência e a Tecnologia
MINISTÉRIO DA CIÊNCIA, TECNOLOGIA E ENSINO SUPERIOR

COMPETE 2020 PORTUGAL 2020

AIR POLLUTION AND ITS ADVERSE IMPACTS ON HUMAN HEALTH: FUTURAR PROJECT REVIEW

Ana Mendes[1,2], Solange Costa[1,2], Joana Ferreira[3], Joana Leitão[3], Pedro Torres[4], Carlos Silveira[3], Hélder Relvas[3], Myriam Lopes[3], Alexandra Monteiro[3], Peter Roebeling[3], Ana Isabel Miranda[3], João Paulo Teixeira[1,2]

[1] Departamento de Saúde Ambiental, Instituto Nacional de Saúde Doutor Ricardo Jorge (INSA), Portugal
[2] EPIUnit – Instituto de Saúde Pública, Universidade do Porto (ISPUP), Portugal
[3] CESAM e Departamento de Ambiente e Ordenamento, Universidade de Aveiro, Portugal
[4] Faculdade de Engenharia da Universidade do Porto, Portugal

BACKGROUND

Air pollution harms human health and the environment. Anthropogenic activities are responsible for the emission of gaseous and particulate pollutants that modify atmospheric composition. Many of these pollutants can cause severe health problems (Figure 1) and negative impact on ecosystems. According to the latest report of the European Environment Agency on air quality (2016), air pollution implications are mainly due to high levels of particulate matter (PM) and ozone (O_3) in the atmosphere, responsible for the degradation of air quality at the regional/local scale, as well as, inducing acute and chronic health effects.

The **FUTURAR project** (HTTP://FUTURAR.WEB.UA.PT/EN/PROJECT) is focused on the assessment of environmental and health impacts associated with air pollutant emission reductions for 2030 imposed by the new National Emission Ceilings Directive (NECD). This Directive sets national reduction commitments for five pollutants, sulphur dioxide, nitrogen oxides, volatile organic compounds, ammonia and fine particulate matter ($PM_{2.5}$), responsible for acidification, eutrophication and ground-level ozone pollution which leads to significant negative impacts on human health and the environment.

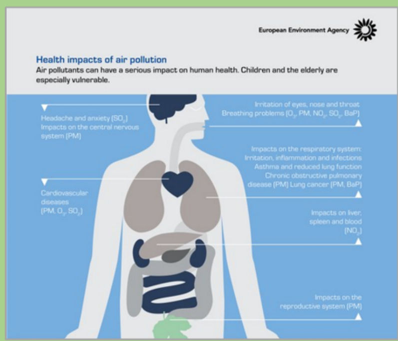


Figure 1. Health impacts of air pollution. *Source: European Environment Agency, 2017*

AIM

THE MAIN GOAL OF **FUTURAR** PROJECT IS TO INVESTIGATE THE AIR QUALITY IMPACTS, COSTS AND BENEFITS OF EMISSION REDUCTION PROJECTIONS FOR 2030.

METHODOLOGY

This project addresses policy-oriented research gaps, namely country-specific exposure-response functions for most important morbidity endpoints, together with a cost-benefit analysis (Figure 2). By this approach the reference exposure-response functions suitable for Portugal will be used to estimate health impacts (Table 1) for each emission reduction scenario and produce reference maps for each selected health indicator.

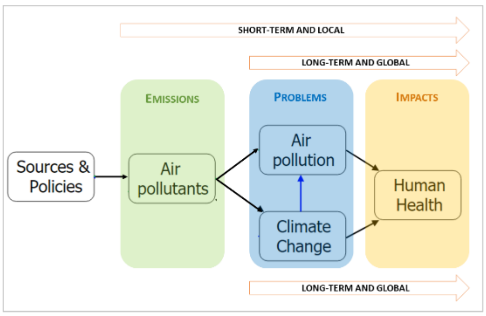


Figure 2.
Co-benefits of mitigating air pollutants emissions for future air quality and human health. (*adapted from West et al. NCC 2013*)

Table 1. OECD Health Status Indicators: Mortality (Portugal)

	Year	Number of total deaths	Deaths per 100 000 population (standardised rates)	Years lost, /100 000 population, aged 0-69 years old
Malignant neoplasms of trachea, bronchus, lung	2010	3658	29,1	..
	2011	3711	29,2	..
	2012	3675	28,5	..
	2013	4010	30,7	..
	2014	3937	29,9	..
Acute myocardial infarction	2010	4976	39,1	..
	2011	4627	35,3	..
	2012	4614	34,4	..
	2013	4568	33,4	..
	2014	4619	34,2	..
Diseases of the respiratory system	2010	11792	93,1	..
	2011	11930	90,4	..
	2012	13908	102,1	..
	2013	12627	90,7	..
	2014	12164	88	..
Malignant neoplasms of trachea, bronchus, lung	2010	..	..	168,9
	2011	..	..	176,2
	2012	..	..	171,9
	2013	..	..	182,1
	2014	..	..	163,3
Acute myocardial infarction	2010	..	..	112,5
	2011	..	..	109,7
	2012	..	..	96,5
	2013	..	..	94
	2014	..	..	121,1
Diseases of the respiratory system	2010	..	..	120,9
	2011	..	..	122,6
	2012	..	..	98,3
	2013	..	..	106,6
	2014	..	..	110,2

Source OECD.Stat (http://stats.oecd.org/index.aspx?DatasetCode=HEALTH_STAT) May 2017

OUTPUTS

THIS RESEARCH ADDRESSES PUBLIC HEALTH POLICY STRATEGIES TO BE TAKEN AT NATIONAL AND REGIONAL LEVEL BY COMPETENT AUTHORITIES TO CONTROL THE EMISSIONS OF AIR POLLUTANTS USING INTEGRATED ASSESSMENT MODELLING TOOLS FOR POLICY SUPPORT OF THESE CLEAN AIR STRATEGIES.

ARTICLES IN NATIONAL JOURNALS WITH SCIENTIFIC PEER-REVIEW

Instituto Nacional de Saúde Doutor Ricardo Jorge, IP

2017 número especial 9 2ª série

Doenças Não Transmissíveis

Lisboa_INSA, IP | Nº especial _ Doenças Não Transmissíveis | ISSN: 2183-8873

Observações_ Boletim Epidemiológico

sumário_

_Editorial

Instituto_Nacional de Saúde Doutor Ricardo Jorge

_Impactos da poluição atmosférica na saúde: perspetivas do projeto FUTURAR

Health impacts related to air pollution: perspectives of the FUTURAR project

Ana Mendes[1,2], Solange Costa[1,2], Joana Ferreira[3], Joana Leitão[3], Pedro Torres[4], Carlos Silveira[3], Hélder Relvas[3], Myriam Lopes[3], Alexandra Monteiro[3], Peter Roebeling[3], Ana Isabel Miranda[3], João Paulo Teixeira[1,2]

a.sofia.mendes@insa.min-saude.pt

(1) Departamento de Saúde Ambiental, Instituto Nacional de Saúde Doutor Ricardo Jorge, Porto, Portugal
(2) Unidade de Investigação em Epidemiologia. Instituto de Saúde Pública, Universidade do Porto, Porto, Portugal
(3) Centro de Estudos do Ambiente e do Mar; Departamento de Ambiente e Ordenamento, Universidade de Aveiro, Aveiro, Portugal
(4) Faculdade de Engenharia, Universidade do Porto, Porto, Portugal

_Resumo

A poluição atmosférica é um importante fator para o desenvolvimento e exacerbação de doenças respiratórias, como a asma, a doença pulmonar obstrutiva crónica e o cancro do pulmão, bem como, um impacto substancial na doença cardiovascular. Além disso, a poluição atmosférica responde por 3,1% dos anos de vida ajustados pela incapacidade global (tempo gasto em estados de saúde reduzida). Apesar da redução de emissões de poluentes atmosféricos e melhoria da qualidade do ar verificadas na Europa e em Portugal na última década, os valores limite legislados para as concentrações de ozono (O_3) e matéria particulada (PM) continuam a ser excedidos. Nesse contexto, o projeto FUTURAR - Qualidade do Ar em Portugal em 2030 - apoio à decisão política, pretende investigar os impactos, custos e benefícios das projeções de redução de emissões para 2030, usando modelos numéricos para estimar a distribuição espacial dos efeitos no ambiente e na saúde em Portugal.

_*Abstract*

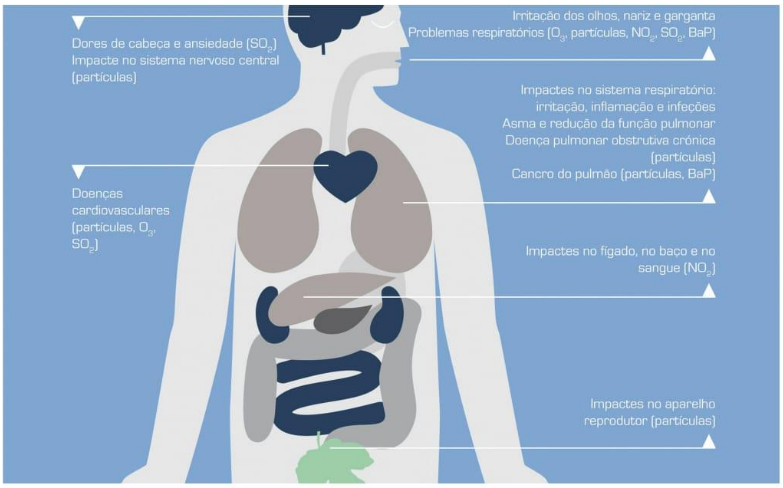
Air pollution is an important stimulus for the development and exacerbation of respiratory diseases, such as asthma, chronic obstructive pulmonary disease, and lung cancer, as well as, a substantial impact on cardiovascular disease. Furthermore, air pollution accounts for 3.1% of global disability-adjusted life years (time spent in states of reduced health). Despite the atmospheric emissions decrease and the air quality improvement in Europe and Portugal over the last decade, EU standards for ozone (O_3) and particulate matter (PM) concentrations are still exceeded. In this context, the main goal of FUTURAR project is to investigate the air quality impacts, costs and benefits of emission reduction projections for 2030, using modelling tools to estimate the spatial distribution of environment and health impacts over Portugal.

_Introdução

O impacto mundial da poluição do ar é de 3,1 milhões de mortes no ano de 2010, em 52,8 milhões referentes a todas as causas e todas as idades, segundo o estudo da *Global Burden of Disease* (1). Em 2013, a Agência Internacional de Investigação em Cancro (IARC) classificou a poluição do ar como cancerígena para os seres humanos (2). Nesse sentido, a poluição atmosférica é um importante fator para o desenvolvimento e exacerbação de doenças respiratórias, como asma, doença pulmonar obstrutiva crónica e cancro do pulmão, bem como, um impacto substancial na doença cardiovascular (3) (figura 1).

Apesar da redução de emissões e melhoria da qualidade do ar verificadas na Europa e em Portugal na última década, os valores limite legislados para as concentrações de ozono (O_3) e matéria particulada (PM) continuam a ser excedidos. Neste sentido, a Estratégia Temática para a Poluição Atmosférica (ETPA) revista em dezembro de 2013, incluía uma proposta de diretiva para a revisão dos Tetos de Emissão Nacionais (TEN) de NOx, SO_2, COV e NH_3 e um teto adicional para $PM_{1.5}$ a atingir em 2030, que foi publicada em dezembro de 2016 (Diretiva (UE) 2016/2284) (4). A avaliação de impacto da ETPA, elaborada pelo IIASA (*International Institute for Applied Systems Analysis*), demonstra a magnitude dos benefícios económicos resultantes da melhoria da saúde obtida na aplicação de um cenário MTFR (considerando a redução máxima tecnicamente possível) para os anos 2025 e 2030 relativamente ao cenário CLE (relativo às reduções decorrentes da legislação atual) (5, 6). De acordo com a referida avaliação, a legislação atual em conjunto com medidas adicionais levará a um aumento de 52%

Figura 1: Impacto da poluição atmosférica na saúde.



Fonte: *European Environment Agency*, 2017 (9)

na esperança média de vida na Europa em 2030 relativamente a 2005 e uma diminuição do número de mortes prematuras devido à exposição ao O_3 em 34%. A redução da deposição irá proteger a biodiversidade e as áreas florestais contra a acidificação. Segundo os resultados atualizados (7, 8), a opção custo-eficaz das medidas de redução de emissões para a melhoria da qualidade do ar (correspondente a 70% dos benefícios do MTFR) prevê para Portugal um decréscimo das emissões de SO_2 em 83% relativamente a 2005 (8). As emissões de NOx e PM deverão sofrer uma redução de 61 e 68% respetivamente, o NH_3 19% e os COV 44%. No entanto, estas estimativas são obtidas por país com base em emissões totais nacionais, sem informação ao nível da variabilidade espacial e alocação geográfica dos impactos na qualidade do ar e na saúde.

Neste contexto, o projeto FUTURAR – Qualidade do Ar em Portugal em 2030 – apoio à decisão política (http://futurar.web.ua.pt/en/project), pretende investigar os impactos, custos e benefícios das projeções de redução de emissões para 2030, usando modelos numéricos para estimar a distribuição espacial dos efeitos no ambiente e na saúde em Portugal.

_Objetivo

Este artigo pretende descrever a importância do projeto FUTURAR no âmbito da saúde ambiental, numa perspetiva de apoio à tomada de decisão na definição de estratégias públicas relativas ao controlo da qualidade do ar e avaliação de impactos da poluição atmosférica no ambiente e na saúde em Portugal.

_Projeto, indicadores base e impactos na saúde

O projeto FUTURAR detém uma abordagem inovadora de apoio à decisão política em Portugal, com o objetivo de obter os seguintes instrumentos e respostas de investigação:

i) cenários de emissões desagregados mais robustos com especial destaque para uma melhor caracterização espacial das emissões de NH_3 provenientes da agricultura, com base nas emissões atuais e projeções dos cenários TEN por setor de atividade e poluente, e em dados e projeções de energia, população, uso do solo;

ii) estimativas da qualidade do ar futura com elevada resolução temporal e espacial para os anos base e alvo dos TEN (2020, 2025, 2030) considerando alteração da meteorologia e modelos de qualidade do ar *online* e *offline* e um conhecimento aprofundado dos processos de formação e remoção de O_3 e PM;

iii) mapeamento dos indicadores de qualidade do ar que traduzem o impacto dos cenários de redução de emissões no ambiente e na saúde usando as relações concentração-resposta mais adequadas a Portugal;

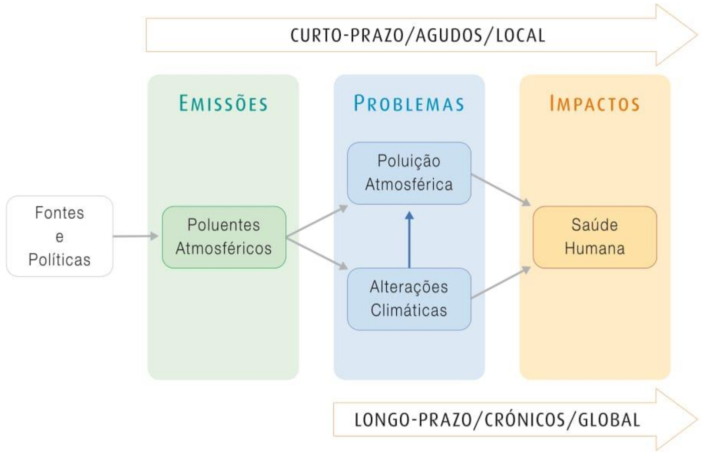
iv) mapeamento do custo-benefício (figura 2) associado a cada um dos cenários TEN, considerando as funções concentração-resposta, globalmente aceites e específicas para cada país, para a maioria das causas de morbilidade e mortalidade (tabela 1).

Tabela 1: OECD Indicadores do estado de saúde: mortalidade (Portugal).

	Ano	Número de mortes totais	Mortes / 100 000 habitantes (taxas padronizadas)	Anos perdidos / 100 000 habitantes com idade entre 0 a 69 anos
Neoplasias malignas da traqueia, brônquios e pulmão	2010	3658	29,1	..
	2011	3711	29,2	..
	2012	3675	28,5	..
	2013	4010	30,7	..
	2014	3937	29,9	..
Enfarte agudo do miocárdio	2010	4976	39,1	..
	2011	4627	35,3	..
	2012	4614	34,4	..
	2013	4568	33,4	..
	2014	4619	34,2	..
Doenças do sistema respiratório	2010	11792	93,1	..
	2011	11930	90,4	..
	2012	13908	102,1	..
	2013	12627	90,7	..
	2014	12164	88	..
Neoplasias malignas da traqueia, brônquios e pulmão	2010	..	..	168,9
	2011	..	..	176,2
	2012	..	..	171,9
	2013	..	..	182,1
	2014	..	..	163,3
Enfarte agudo do miocárdio	2010	..	..	112,5
	2011	..	..	109,7
	2012	..	..	96,5
	2013	..	..	94,0
	2014	..	..	121,1
Doenças do sistema respiratório	2010	..	..	120,9
	2011	..	..	122,6
	2012	..	..	98,3
	2013	..	..	106,6
	2014	..	..	110,2

Fonte: OECD.Stat, maio 2017 (10).

Figura 2: Cobenefícios da mitigação das emissões de poluentes atmosféricos para a futura qualidade do ar e saúde humana.



Adaptado de: West JJ, *et al.*, 2013 (10).

A revisão de literatura sobre os impactos na saúde, realizada no âmbito do projeto MAPLIA - *Moving from Air Pollution to Local Integrated Assessment* (http://projeto-maplia.web.ua.pt/) (12), continua a ser desenvolvida e alargada para adquirir uma compreensão importante e completa sobre o estado da arte (estudos coorte e de caso-controle) dos efeitos na saúde e métricas relacionadas à exposição humana a O_3, PM e seus precursores abordadas pela diretiva TEN. Está em curso a identificação das funções de exposição-resposta, com foco na morbidade, mortalidade e indicadores de saúde relevantes, usando a distribuição espacial das concentrações de poluentes do ar para cada cenário, considerando a variabilidade geográfica em Portugal. Estas funções de exposição-resposta serão usadas para estimar o impacto de saúde de cada cenário de redução de emissões e produzir mapas para cada indicador de saúde selecionado.

Este projeto auxiliará as entidades competentes (ex. Agência Portuguesa do Ambiente, APA) na definição de estratégias políticas ao nível nacional/regional para o controlo das emissões. Apoiará também a implementação das novas políticas de qualidade do ar em Portugal e contribuirá para o programa nacinal de controlo da poluição atmosférica (PNCPA) (um requisito da nova diretiva TEN), através da elaboração de recomendações e orientações à decisão política. Além disso, os resultados do FUTURAR permitirão ajustar e corrigir as estimativas efetuadas pelo IIASA para Portugal relativamente à redução de emissões e impactos no ambiente e na saúde.

Financiamento:

Projeto financiado pelo FEDER-COMPETE e pela Fundação para a Ciência e Tecnologia (POCI-01-0145-FEDER-016752), (PTDC/AAG-MAA/2569/2014).

Referências bibliográficas:

(1) Newby DE, Mannucci PM, Tell GS, et al.; ESC Heart Failure Association.Expert position paper on air pollution and cardiovascular disease. Eur Heart J. 2015;36(2):83-93b.Epub 2014 Dec 9.

(2) International Agency for Research on Cancer, World Health Organization. IARC: Outdoor air pollution a leading environmental cause of cancer deaths. IARC Press release n. 221, 17 October 2013. www.iarc.fr/en/media-centre/iarcnews/pdf/pr221_E.pdf

(3) European Environment Agency. Air quality in Europe: report 2016. Copenhagen: EEA, 2016(EEA Report no. 28/2016). www.eea.europa.eu/publications/air-quality-in-europe-2016

(4) Comissão Europeia. Diretiva (UE) 2016/2284 do Parlamento Europeu e do Conselho, de 14 de dezembro de 2016, relativa à redução das emissões nacionais de certos poluentes atmosféricos, que altera a Diretiva 2003/35/CE e revoga a Diretiva 2001/81/CE. http://data.europa.eu/eli/dir/2016/2284/oj

(5) Comissão Europeia. Proposta de Diretiva do Parlamento Europeu e do Conselho, de 18 de dezembro de 2013, relativa à redução das emissões nacionais de determinados poluentes atmosféricos e que altera a Diretiva 2003/35/CE /* COM/2013/0920 final - 2013/0443 (COD) */ http://eur-lex.europa.eu/legal-content/PT/TXT/?uri=CELEX:52013PC0920&qid=1499856718543

(6) Holland M. Cost-benefit analysis of final policy scenarios for the EU Clean Air Package (Version 2. Corresponding to IIASA TSAP Report #11, Version 2a), Edinburgh, Ecometrics Research and Consulting UK, 2014. http://ec.europa.eu/environment/air/pdf/TSAP%20CBA.pdf

(7) Amann M (ed). The final policy scenarios of the EU Clean Air Policy Package (TSAP Report #11 Version 1.1a). Laxenburg, Austria: International Institute for Applied Systems Analysis, 2014. http://ec.europa.eu/environment/air/pdf/TSAP.pdf

(8) Amann M (ed). Adjusted historic emission data, projections, and optimized emission reduction targets for 2030: a comparison with COM data 2013 - Part B: results for Member States (TSAP Report #16B Version 1.1). Laxenburg, Austria: International Institute for Applied Systems Analysis, 2015. http://ec.europa.eu/environment/air/pdf/review/TSAP_16b.pdf

(9) Comissão Europeia. Agência Europeia do Ambiente. Sinais – Viver num clima em mudança, 2013, Infografia - Impacte da poluição atmosférica na saúde (2013-05-06). Copenhaga, AEA, 2013. www.eea.europa.eu/pt/sinais-da-aea/sinais-2013/infografia/impacte-da-poluicao-atmosferica-na-saude-2

(10) West JJ, Smith SJ, Silva RA, et al. Co-benefits of Global Greenhouse Gas Mitigation for Future Air Quality and Human Health. Nat Clim Chang. 2013;3(10):885-89. www.ncbi.nlm.nih.gov/pmc/articles/PMC4051351/

(11) Organisation for Economic Co-operation and Development. OCDE.Stat [Em linha]. [consult. 20/6/2017]. http://stats.oecd.org/index.aspx?DatasetCode=HEALTH_STAT)

(12) Costa S, Ferreira J, Silveira C, et al. Integrating health on air quality assessment: review report on health risks of two major European outdoor air pollutants: PM and NO_2. J Toxicol Environ Health B Crit Rev. 2014;17(6):307-40.