Universidade de Aveiro Departamento de Ambiente e Ordenamento
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Regional and urban air quality modelling under a climate change scenario

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Regional and urban air quality modelling under a climate change scenario

Tese apresentada à Universidade de Aveiro para cumprimento dos requisitos necessários à obtenção do grau de Doutor em Ciências e Engenharia do Ambiente, realizada sob a orientação científica do Doutor Carlos Borrego, Professor Catedrático do Departamento de Ambiente e Ordenamento da Universidade de Aveiro e sob coorientação científica da Doutora Anabela Carvalho, Técnica Superior do Departamento de Ambiente e Ordenamento da mesma Universidade, e do Doutor Stefano Galmarini, Investigador Sénior do Joint Research Center (JRC).

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"It always seems impossible until it's done."

Nelson Mandela

o júri

presidente

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palavras-chave

Qualidade do ar, emissões de poluentes atmosféricos, alterações climáticas, cenários de emissão, modelação numérica

resumo

A interação entre as alterações climáticas e a qualidade do ar é neste momento um assunto emergente em termos de implementação de políticas e de investigação. As alterações climáticas causarão mudanças no clima, o que irá afetar a concentração e dispersão dos poluentes atmosféricos. Assim, o principal objetivo deste trabalho é avaliar os impactos das alterações climáticas na qualidade do ar em 2050 em Portugal e na área urbana do Porto. Numa primeira fase, realizou-se uma caracterização da qualidade do ar em Portugal para o período de 2002 a 2012, na qual se identificou que os poluentes NO_2, PM10 e O_3 são os mais críticos em termos de qualidade do ar. Adicionalmente, foi analisada a influência da meteorologia na qualidade do ar para esses três poluentes nas duas maiores áreas urbanas nacionais (Porto e Lisboa), sendo que o O_3 é estatisticamente dependente da temperatura na maioria das suas componentes. Após o entendimento dos problemas de qualidade do ar nacionais e da influência da meteorologia nos mesmos, o sistema de modelos WRF-CAMx foi testado e todos os seus dados de entrada foram preparados. As emissões nacionais atuais foram estimadas com maior detalhe de desagregação para melhorar as simulações de qualidade do ar; o modelo de emissões, EmiPro-RCP, foi desenvolvido para estimar as emissões de 2050 tendo em conta os cenários de emissão RCPs. O sistema de modelos WRF-CAMx foi testado e avaliado para Portugal e para a área urbana do Porto, verificando-se que é uma ferramenta adequada para realizar as simulações de qualidade do ar em cenário climático. Realizaram-se simulações regionais com o modelo CAMx versão 6.0, para dois períodos: histórico e futuro (2045-2050), de forma a simular os impactos do clima futuro e das futuras emissões antropogénicas na qualidade do ar para a região de estudo. O cenário climático, bem como as emissões, foram projetadas tendo como base o cenário RCP8.5. Os resultados provenientes das simulações demonstram que, se as emissões antropogénicas se mantiverem constantes em 2050, as concentrações de NO_2, PM10 e O_3 irão aumentar em Portugal. Quando, aos efeitos das alterações climáticas se juntaram as futuras emissões antropogénicas, verifica-se que as concentrações médias anuais de NO_2 irão diminuir e as concentrações médias anuais de PM10 aumentam em Portugal e diminuem na área urbana do Porto. Os resultados de O_3 estão relacionados com as variações de concentração dos seus precursores, verificando-se as maiores reduções nas áreas urbanas e os aumentos nas áreas suburbanas. Toda a análise realizada aos dados das simulações para a área urbana do Porto indica que, no caso de PM10 e O_3, irá existir um aumento de ocorrência de valores extremos de concentração, ultrapassando os valores legislados de cada poluente.

Este estudo constitui uma ferramenta científica inovadora que pode ser relevante para uma futura e cuidada gestão da qualidade do ar, de forma a mitigar os impactes das alterações climáticas na qualidade do ar.

keywords

Air quality, atmospheric pollutant emissions, climate change, emission scenarios, numerical modelling.

abstract

The better understanding of the interactions between climate change and air quality is an emerging priority for research and policy. Climate change will bring changes in the climate system, which will affect the concentration and dispersion of air pollutants. The main objective of the current study is to assess the impacts of climate change on air quality in 2050 over Portugal and Porto urban area. First, an evaluation and characterization of the air quality over mainland Portugal was performed for the period between 2002 and 2012. The results show that NO_2, PM10 and O_3 are the critical pollutants in Portugal. Also, the influence of meteorology on O_3, NO_2 and PM10 levels was investigate in the national main urban areas (Porto and Lisboa) and was verified that O_3 has a statistically significant relationship with temperature in most of the components. The results also indicate that emission control strategies are primary regulators for NO_2 and PM10 levels. After, understanding the national air quality problems and the influence that meteorology had in the historical air quality levels, the air quality modelling system WRF-CAMx was tested and the required inputs for the simulations were prepared to fulfil the main goal of this work. For the required air quality modelling inputs, an Emission Projections under RCP scenarios (EmiPro-RCP) model was developed to assist the estimation of future emission inventories for GHG and common air pollutants. Also, the current emissions were estimated for Portugal with a higher detailed disaggregation to improve the performance of the air quality simulations. The air quality modelling system WRF/CAMx was tested and evaluated over Portugal and Porto urban area and the results point out that is an adequate tool for the analysis of air quality under climate change. For this purpose, regional simulations of air quality during historical period and future (2045-2050) were conducted with CAMx version 6.0 to evaluate the impacts of simulated future climate and anthropogenic emission projections on air quality over the study area. The climate and the emission projections were produced under the RCP8.5 scenario. The results from the simulations point out, that if the anthropogenic emissions keep the same in 2050, the concentrations of NO_2, PM10 and O_3 will increase in Portugal. When, besides the climate change effects, is consider the projected anthropogenic emissions the annual mean concentrations of NO_2 decrease significantly in Portugal and Porto urban area, and on the contrary the annual mean PM10 concentrations increases in Portugal and decrease in Porto urban area. The O_3 results are mainly caused by the reduction of ozone precursors, getting the higher reductions in urban areas and increases in the surrounding areas. All the analysis performed for both simulations for Porto urban area support that, for PM10 and O_3, there will be an increase in the occurrence of extreme values, surpassing the annual legislated parameters and having more daily exceedances.

This study constitutes an innovative scientific tool to help in future air quality management in order to mitigate future climate change impacts on air quality.

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Glossary

AQP	Air Quality Plans
AT	Alert Threshold
BIAS	Systematic error
C_6H_6	Benzene
CAMx	Comprehensive Air quality Model with extensions
CH_4	Methane
CLE	Current Legislation scenario
CLRTAP	Convention on Long-range Transboundary Air Pollution and the EU National Emission Ceilings Directive
CMAQ	Community Multi-scale Air Quality Model
CMIP5	Coupled Model Intercomparison Project Phase 5
CO	carbon monoxide
CO_2	carbon dioxide
EEA	European Environmental Agency
EmiPro-RCP	Emission Projections under RCP scenarios model
EP	Execution Programme
EU	European Union
FD	Framework Directive
GHG	Greenhouse Gases
H2SO4	Sulphuric acid
IIASA	International Institute for Applied Systems Analysis
INERPA	Portuguese national emission inventory
IPCC	International Panel of Climate Change
IS92	IPCC scenarios 1992
IT	Information Threshold
KZ	Kolmogorov-Zurbenko filter
LTO	Long-Term Objective
LV	Limit Value
MFR	Maximum Feasible Reduction scenario
MT	Margin of Tolerance
MS	Member State
N_2O	Nitrous oxide
NH_3	Ammonia
NIR	National Inventory Report
NMVOC	Non-Methane Volatile Organic Compounds
NO	Nitrogen monoxide

NO_2	Nitrogen dioxide
O_3	Ozone
Pb	Lead
PEA	Portuguese Environmental Agency
PM	Particulate Matter
PM10	Particulate Matter with a 50% efficiency cut-off at 10 mm aerodynamic diameter
PM2.5	Particulate Matter with a 50% efficiency cut-off at 2.5 mm aerodynamic diameter
r	Correlation coefficient
CCDR	Regionals Coordination and Development Commissions
RCP	Representative Concentration Pathway
RMSE	Root Mean Square Error
SNAP	Standardized Nomenclature for Air Pollutants
SRES	Special Report on Emission Scenarios
TAPM	The Air Pollution Model
TREM	Transport Emission Model for Line Sources
TV	Target Value
UNFCC	United Nations Framework Convention on Climate Change
VOCs	Volatile Organic Compounds
WRF	Weather Research Forecast model

Chapter 1

1 Introduction

1.1 Overview

According to the World Health Organization (WHO, 2006), "Clean air is considered to be a basic requirement of human health and well-being. However, air pollution continues to pose a significant threat to health worldwide". The WHO assessed that more than 2 million premature deaths each year can be attributed to the effects of urban air pollution (WHO, 2006).

Along with air pollutants emissions, air quality is strongly dependent on weather conditions, and is therefore expected to be affected by long-term changes in weather statistics, i.e., by the changing of climate. Climate change will bring changes in the magnitude and frequency of all key components and natural cycles of the climate system (IPCC, 2013b). These changes will affect the concentration and dispersion of air pollutants, and consequently the magnitude and frequency of the air pollution episodes. Besides that, changes in future weather patterns will affect future human activities, which will lead, in turn, to an impact over the air pollutant emissions. In this sense, a better understanding of the effects of climate change on regional and urban air quality is an emerging priority for research and policy.

1.1.1 Climate change: concept, evidences and impacts

In the last International Panel for Climate Change (IPCC) 2013 report (AR5) (IPCC, 2013a), climate change is defined as a change in the state of the climate that can be identified (e.g., by using statistical tests) through changes in the mean and/or the variability of its properties that persists for an extended period, typically decades or longer. Climate change may be due to natural internal processes or external forcings such as the modulations of the solar cycles, volcanic eruptions, and persistent anthropogenic changes in the composition of the atmosphere or in land use. This concept differs from the one in the United Nations Framework Convention on Climate Change (UNFCCC), where climate change refers to a change of climate that is attributed directly or indirectly to human activity which alters the composition of the global atmosphere adding up to the natural climate variability observed over comparable time periods. The UNFCCC thus makes a distinction between climate change attributable to human activities, on altering the atmospheric composition, and climate variability attributable to natural causes (IPCC, 2013a).

There are many indicators of climate change which include physical responses such as changes in the following variables: surface temperature, atmospheric water vapour, precipitation, severe events, glaciers, ocean and land ice, and sea level. Global mean surface air temperatures over land and oceans have increased over the last 100 years. Temperature measurements in the oceans show a continuing increase in their heat content. Analyses based on measurements of the Earth's radiative budget suggest a small positive energy imbalance that serves to increase the global heat content of the Earth system. Observations from satellites and in situ measurements show a trend of significant reductions in the mass balance of most land ice masses and in Arctic sea ice. The oceans' uptake of carbon dioxide (CO_2) is having a significant effect on the chemistry of sea water. Paleoclimatic reconstructions have helped place ongoing climate change in the perspective of natural climate variability (Cubasch et al., 2013).

Figure 1.1 illustrates the influence of the anthropogenic forcings in the increase of global average temperature and in the ocean heat content, confirming that the increase of GHG gases by anthropogenic processes contribute for the global warming.

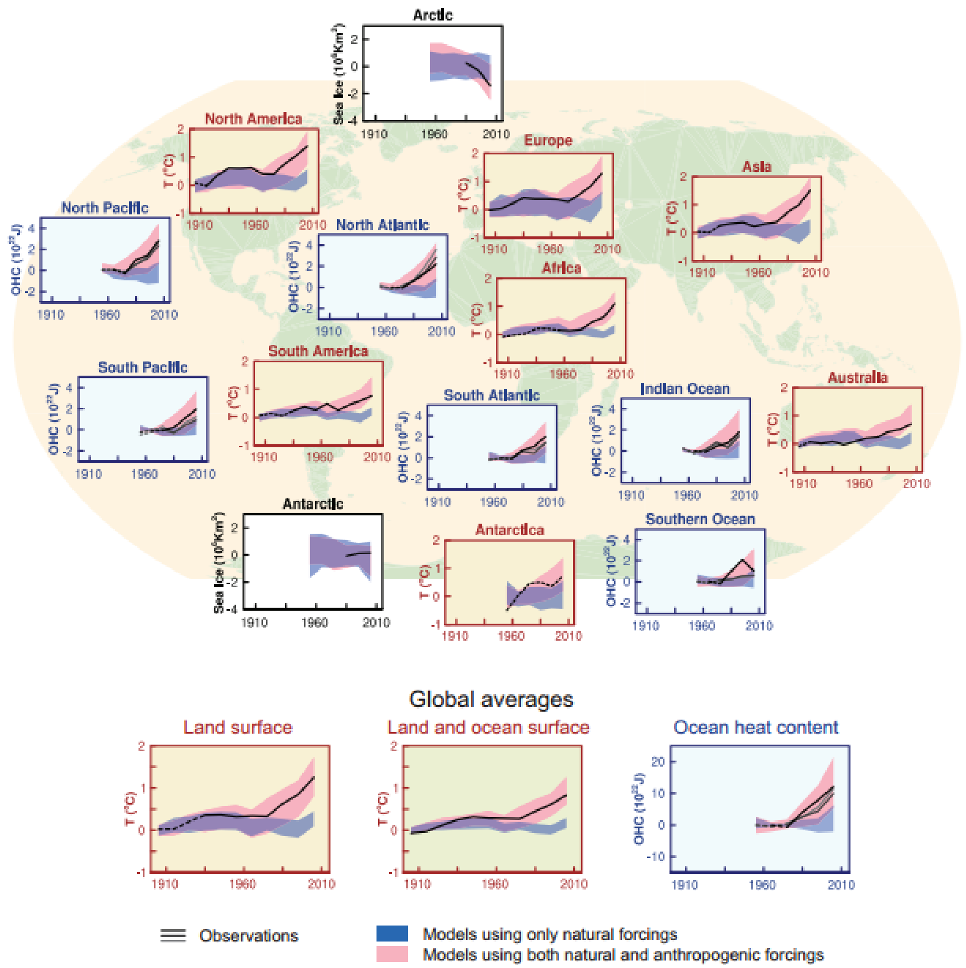


Figure 1.1– Comparison of observed and simulated climate change based on three large-scale indicators: temperature, ocean heat content and sea ice extent (IPCC, 2013b).

In all the regions of the World, with exception of Arctic and Antarctic, the models that use both natural and anthropogenic forcings show results with the same behaviour as the observations. This fact demonstrates how the human activities had impact on climate over the last century (ex. Temperature), and how is important to consider them in the future.

The period 2004-2013 was the warmest decade on record in Europe. Many other changes significant for Europe have been observed across the climate system, including warming oceans, rising sea level and shrinking of the snow cover, ice sheets, sea ice and glaciers (URL1). Over Portugal, since 1972 there is a general trend towards an increase in the mean annual surface air temperature (Santos et al., 2002). From the 30's to the end of the XXth century positive trends on average and maximum temperatures registered in Portugal. However, both parameters show a wave behaviour on the trends. Higher positive trends show up between 1930 and 1945 and in the end of the period under analysis, i.e., 1975-2000; Negative trends were calculated between 1945 and 1975, -0.14 ºC/decade and -0.16 ºC/decade for maximum and mean temperatures, respectively. The increase in the mean temperatures over Portugal between 1975 and 2000 was of +0.48 ºC/decade (Figure 1.2).

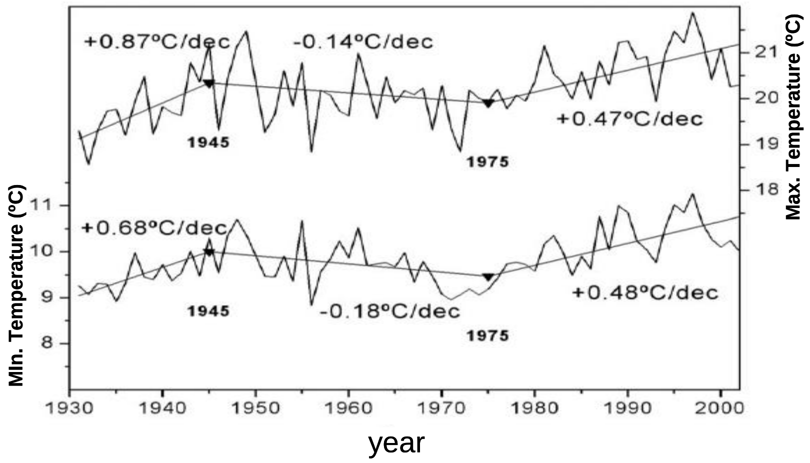


Figure 1.2 – Observed temperature in Portugal between years 1930 and 2000 (Santos et al., 2002).

The mean temperature has risen in all regions of Portugal in the period 1976-2006, at a rate of approximately 0.52 ºC per decade, which more than double the rate of mean annual global temperature increase. Time-series analysis of the mean annual temperature starting at 1941 shows that 1997 was the warmest year and that 8 of the 10 warmest years occurred in the last 20 years of the serie. Also the observation of temperature indices indicates that the increase of the mean temperature was accompanied by a change in the frequency of very hot days and a decrease in the frequency of very cold ones. The heat waves of 1981, 1991, 2003, 2006, 2009, and 2010, becoming more frequent since the beginning of this century and were of particular significance due to their duration and spatial extension (Ferreira et al., 2008; Carvalho et al., 2013).

Climate models are powerful tools to represent possible future weather patterns. A hierarchy of climate models ranging from simple climate models, to models of intermediate complexity, to comprehensive climate models, and Earth System Models were applied by the IPCC to study future changes in the climate system.

In order to determine the impact of climate change in the future, it is necessary to estimate the concentrations of greenhouse gases and other pollutants in the atmosphere to which the climate is sensitive for, in the years to come. These concentrations depend on their emissions from various sources, natural, as well as anthropogenic. The latest are foresee as emissions scenarios describing future releases into the atmosphere of greenhouse gases and other pollutants, which are used as inputs to climate models. Until now, scenarios were developed and applied sequentially in a linear causal chain that extended from the socioeconomic factors that influence GHG emissions through the atmospheric and climate processes and finally to impacts.

In 1992, the IPCC developed long-term emissions scenarios (IS92) to be used for driving global circulation models to develop climate change scenarios. These scenarios were the first global scenarios to provide estimates for the full suite of greenhouse gases and have been widely used in the analysis of possible climate change, its impacts, and options to mitigate climate change. However in 1995 the IS92 scenarios were evaluated and significant changes had been recommended, namely in the understanding of driving forces of emissions and methodologies that should be addressed and consequently, the development of new scenarios. In this sense, IPCC release the Special Report on Emissions Scenarios (SRES), which reproduce a large, global and long-term effort. They cover a wide range of key "future" characteristics such as demographic change, economic development, and technological change. Although these scenarios assume improvements in production technology, they do not include some of the expected changes in the future penetration of abatement measures, like the highly probable impacts resulting from the application of legislation approved at the time (Nakicenovic et al., 2000). The SRES scenarios are constituted by four qualitative storylines yield four sets of scenarios called "families": A1, A2, B1, and B2, and altogether (40 SRES scenarios) have been developed by six modelling teams.

The SRES scenarios specifically do not assume that any action will be taken to limit emissions under international treaties such as Kyoto Protocol. This approach is taken further by another important scenarios developed by International Institute for Applied Systems Analysis (IIASA), which generate the Current Legislation scenario (CLE), and the Maximum Feasible Reduction (MFR) scenario (AQEG, 2007). The CLE scenario assumes the implementation of all presently decided emission related national legislation, such as the large combustion plan directive, directives on quality of petrol and diesel fuels, etc. The MFR scenario assumes full implementation of the presently available most advanced technical emission control measures. It assumes technologies like combustion modification on small biomass boilers, high efficiency in flue-gas desulfurization, etc. In both scenarios the legislation and measures assumed is specific for each pollutant (Amann, 2004).

The creation of this kind of scenarios led to inconsistencies due to the delays between the development of scenarios, their use in climate modelling, and the availability of the resulting climate scenarios for impact assessment. To overcome these delays, a parallel approach was developed to create emission scenarios, but in this case the time between the development and the use of the resulting climate scenarios in impact studies was shorten. Instead of starting with detailed socioeconomic storylines to generate emissions and then climate scenarios, this process begins with the identification of important characteristics for scenarios of radiative forcing for climate modelling, e.g. the level of radiative forcing in the year 2100 (Moss et al., 2010). Figure 1.3 describes the parallel approach.

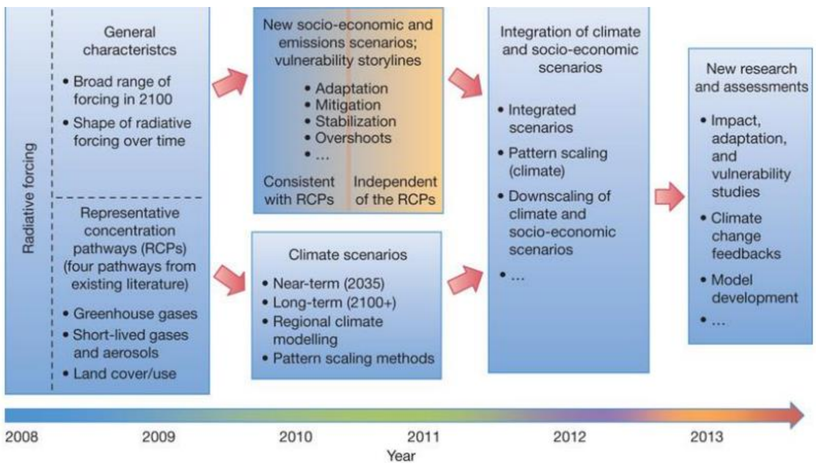


Figure 1.3 - Parallel approach of scenarios development (Moss et al., 2010).

The Representative Concentration Pathways (RCPs) are a set of four new pathways of pollutant emission, which were considered into the global climate models simulations, carried out under the framework of the Coupled Model Intercomparison Project Phase 5 (CMIP5) of the World Climate Research Programme. In all RCPs atmospheric CO_2 concentrations are higher in 2100 relative to present day as a result of a further increase of cumulative emissions of CO_2 to the atmosphere during the 21st century. These pathways are a result of a careful selection process where the needs of both climate scenario developers and users were considered.

The definition of the RCPs follows several design criteria in order to facilitate climate research and assessment (van Vuuren et al. 2011). The final RCP selections were: RCP2.6, RCP4.5, RCP6.0 and RCP8.5 (Moss et al., 2010), where the number in the acronyms refers to the radiative forcing achieved in 2100 the main characteristics of each one are showed in Table 1.1.

Table 1.1– Characteristics of the RCPs scenarios.

Name	Radiative forcing	Concentration (p.p.m.)	Pathway	Model providing RCP
RCP8.5 a)	>8.5 $W.m^{-2}$ in 2100	>1370 $CO_{2equiv.}$ in 2100	Rising	MESSAGE
RCP6.0 b)	≈6$W.m^{-2}$ at stabilization after 2100	≈850 $CO_{2equiv.}$ (at stabilization after 2100)	Stabilization without overshoot	AIM
RCP4.5 c)	≈4.5 $W.m^{-2}$ at stabilization after 2100	≈650 $CO_{2equiv.}$ (at stabilization after 2100)	Stabilization without overshoot	GCAM
RCP2.6 d)	Peak at ≈3 $W.m^{-2}$ before 2100 and then declines	Peak at ≈490 $CO_{2equiv.}$ before 2100 and then declines	Peak and decline	IMAGE

a) Rao and Riahi, 2006 and Riahi et al., 2007; b) Fujino et al., 2006 and Hijioka et al., 2008

c) Clarke et al., 2007 and Smith and Wigley, 2006; d) van Vuuren et al., 2006 and van Vuuren et al., 2007

The results of the application of climate models under the RCP scenarios shown in the AR5 predict that the anthropogenic forcing by the continued emissions of GHG will bring about changes in the magnitude and frequency of all key components and natural cycles of the climate system (IPCC, 2013b), namely:

- Global surface temperature change at the end of the 21st century is likely to exceed 1.5°C relative to 1850 to 1900 for all RCP scenarios except RCP2.6. It is likely to exceed 2°C for RCP6.0 and RCP8.5, and more likely than not to exceed 2°C for RCP4.5;

- The contrast in precipitation between wet and dry regions and between wet and dry seasons will increase, although there may be regional exceptions;

- The global ocean will continue to warm during the 21st century. Heat will penetrate from the surface to the deep ocean and affect ocean circulation;

- It is very likely that the Arctic sea ice cover will continue to shrink and thin and that Northern Hemisphere spring snow cover will decrease during the 21st century as global mean surface temperature rises. Global glacier volume will further decrease;

- Global mean sea level will continue to rise during the 21st century. Under all RCP scenarios, the rate of sea level rise will very likely exceed that observed during 1971 to 2010 due to increased ocean warming and increased loss of mass from glaciers and ice sheets;

- Climate change will affect carbon cycle processes in a way that will exacerbate the increase of CO_2 in the atmosphere (high confidence). Further uptake of carbon by the ocean will increase ocean acidification;

- Cumulative emissions of CO_2 largely determine global mean surface warming by the late 21st century and beyond. Most aspects of climate change will persist for many centuries even if emissions of CO_2 are stopped. This represents a substantial multi-century climate change commitment created by past, present and future emissions of CO_2.

In order to generate regional climate projections for the land-regions worldwide and to harmonise model evaluation activities, the Coordinated Regional Downscaling Experiment (CORDEX, URL2; Giorgi et al. 2006) was created. CORDEX provides an internationally coordinated framework to improve regional climate scenarios.

As part of the global CORDEX framework, the EUROCORDEX initiative (URL3) provides regional climate projections for Europe at 50 km (EUR-44) and 12.5 km (EUR-11) resolution. The regional simulations are conducted through a downscaling of the new CMIP5 global climate projections (Taylor et al. 2012) considering the RCPs scenarios. In Figure 1.4 are some of the results of EUROCORDEX for the RCP4.5 and RCP8.5 scenarios at a horizontal resolution of 12.5 km (Jacob et al., 2013).

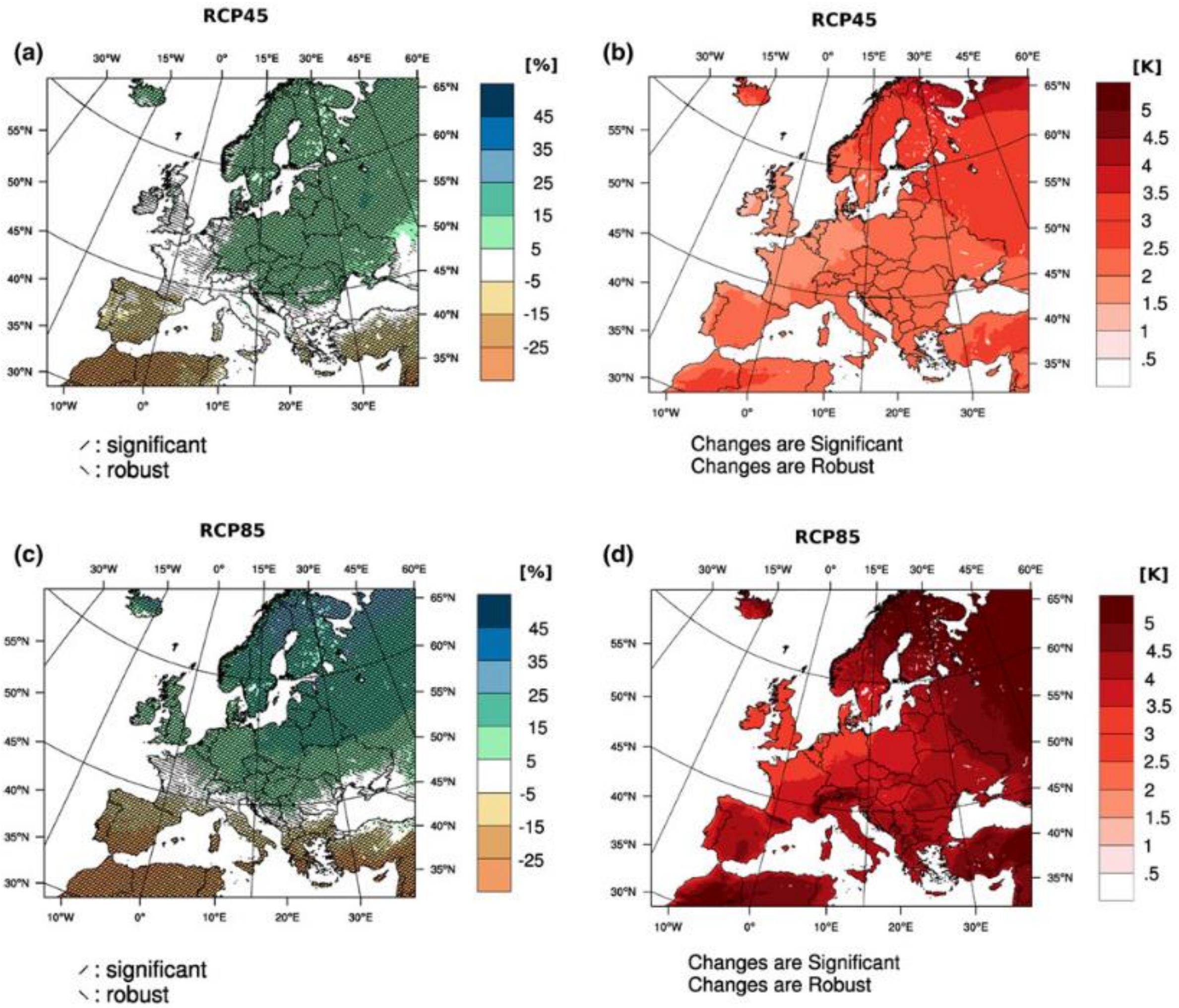


Figure 1.4- Projected changes of total annual precipitation (%) (left) and annual mean temperature [K] (right) for 2071–2100 compared to 1971–2000, RCP8.5 (c, d) and RCP4.5 (a, b) scenarios (Jacob et al., 2013).

In Santos et al. (2001), despite the fact that the emission scenarios driving the results are not the RCP but the SRES scenarios, the regional climate change predictions indicate that the Iberian Peninsula is likely to become hotter and drier by the end of the XXI century, with an increase of temperature and decrease of precipitation (Figure 1.5 and Figure 1.6), which is in accordance to Figure 1.4.

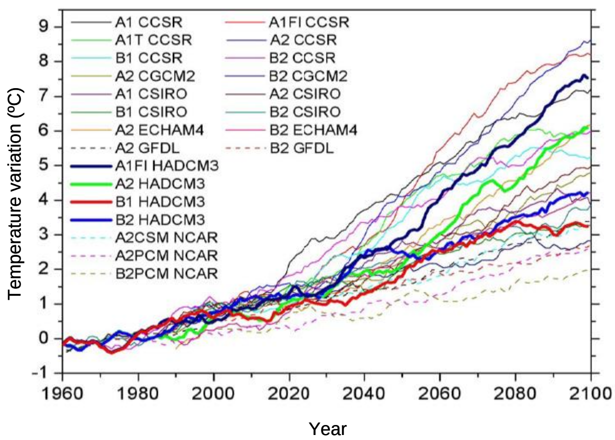


Figure 1.5– Predicted evolution of the mean temperature in the Iberian Peninsula considering different global climate model projections (Santos et al., 2001).

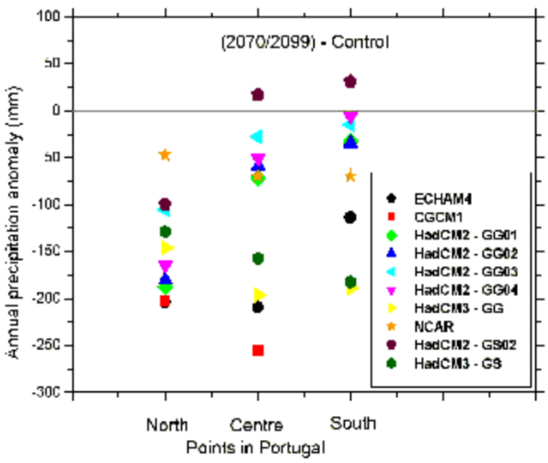


Figure 1.6– Annual precipitation anomalies in 3 points in Portugal considering different global climate model projections (Santos et al., 2001).

By 2100, the projections indicate an increase of temperature in the interval of 4-7 ºC and a decrease of precipitation of the order of 100 mm/year appears to be a likely scenario over Portugal (Santos et al., 2001). In Soares et al. (2014) the mean seasonal precipitation is expected to decrease substantially in all seasons, excluding winter, under the A1B emission scenario. This reduction is statistically significant; it spans from less than 20% in the north to 40% in the south in the intermediate seasons, and is above 50% in the largest portion of mainland in summer.

Climate change will result in an impact on the general weather patterns, in particular, wind, temperature, sunshine hours and rainfall patterns climatologies. This in turn may result in a change on the processes that governs the chemical transformations in the atmosphere, which may lead to air quality changes (AQEG, 2007).

1.1.2 Air quality: origin and evaluation

The problems derived by air pollution have been known for millennia, but the attitude towards them was ambiguous. The world has developed drastically: the global population has highly increase over the last 50 years, the number of people living in cities has increased by more than a factor of four and the global energy consumption by nearly a factor of five. There are now globally about ten times as many cars as 50 years ago, causing problems in the industrialised world, mainly due to the increase of nitrogen oxides, volatile organic compounds and photochemical oxidants. Nevertheless the urban environment in the industrialised world has improved with respect to the classical pollution caused by sulphur dioxide and soot from power and heat production. In addition new hazardous compounds, mainly from industry, have been identified with advanced analytical techniques. Finally agriculture has become an important air pollution source, notably with ammonia from animal production and organic fertilizing. Therefore, the scale of the pollution has increased both in time and magnitude (Fenger, 2009).

Air quality is one of the environmental areas in which the European Union (EU) has been most active, namely in the design and implementation of legislation on air quality and pollutant emissions to the atmosphere. In order to reduce and control the effects of air pollution on human health and in the environment, the air quality Framework Directive (FD) (Directive 2008/50/EC) requires the EU member states to assess air quality through their territory, establishing the obligations of the member states and redefining the guidelines for the assessment and management of air quality. The FD set limits and targets for concentrations of various pollutants for the protection of health and ecosystems, such as annual and daily limit values (LV) and maximum of exceedances for each pollutant. Europe has significantly decrease emissions of several air pollutants in recent decades, greatly reducing emissions and exposure to substances such as sulphur dioxide (SO_2), CO, benzene (C_6H_6) and lead (Pb). Despite improvements over several decades, air pollution continues to damage human health and the environment, namely with particulate matter (PM), ozone (O_3), reactive nitrogen substances and some organic compounds (EEA, 2013). Europe is facing air quality problems in several countries since is surpassing the legislated values of particulate matter with a 50% efficiency cut-off at 10 μm aerodynamic diameter (PM10), nitrogen dioxide (NO_2) and O_3 (Figure 1.7).

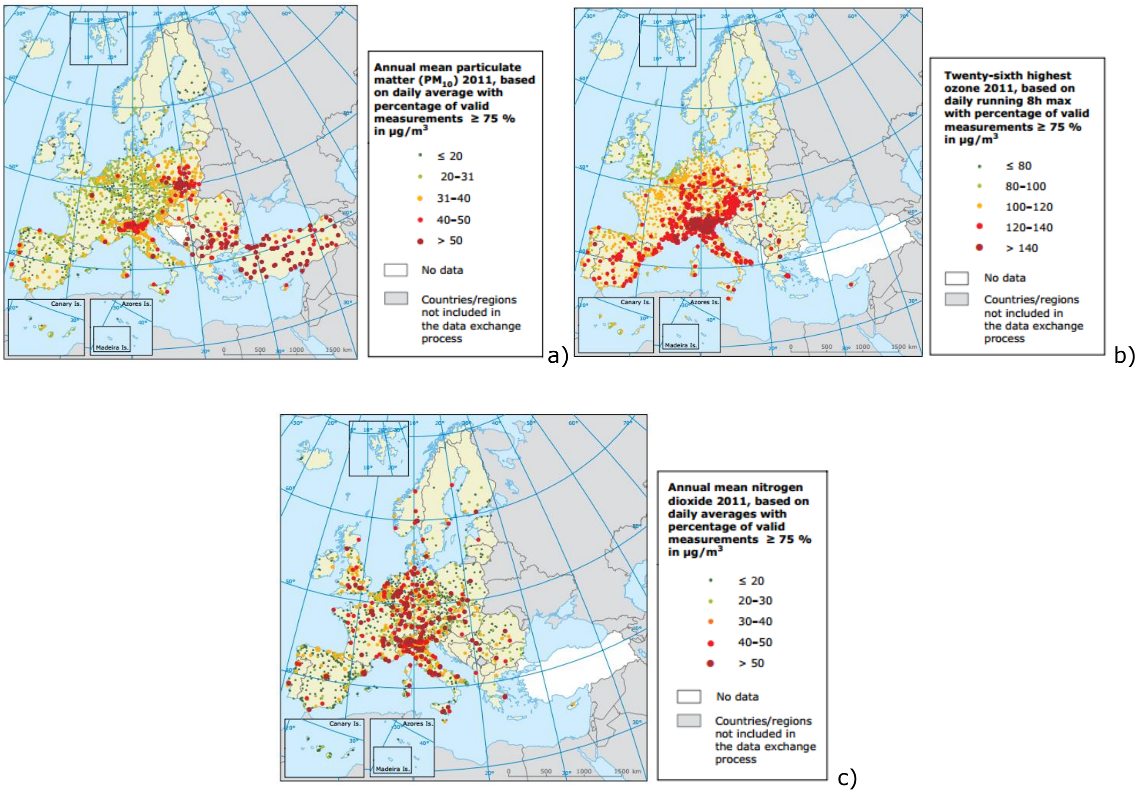


Figure 1.7– Annual mean concentrations of PM10 in 2011 (a); 26th-highest daily maximum 8hrour average O_3 concentration recorded at each monitoring station in 2011 (b); Annual mean concentration of NO_2 in 2011 (c) (EEA, 2013).

Figure 1.7 shows that in Europe the annual LV of PM10 (40 $\mu g.m^{-3}$) and NO_2 (40 $\mu g.m^{-3}$) was surpassed in 2011 in several air quality stations. Also, the maximum number of exceedances allowed (25 times) to the O_3 maximum daily 8-hr mean (120 $\mu g.m^{-3}$) was exceeded in Europe, reaching values higher than 140 $\mu g.m^{-3}$.

Over the last years Portugal is facing air quality problems due to the surpassing of the legislated limit values at several air quality monitoring stations, mainly over two main regions: Lisboa and Vale do Tejo region and the Northern Region, for three air pollutants: PM10, NO_2 and O_3 (Borrego et al., 2012a,b) (Figure 1.8).

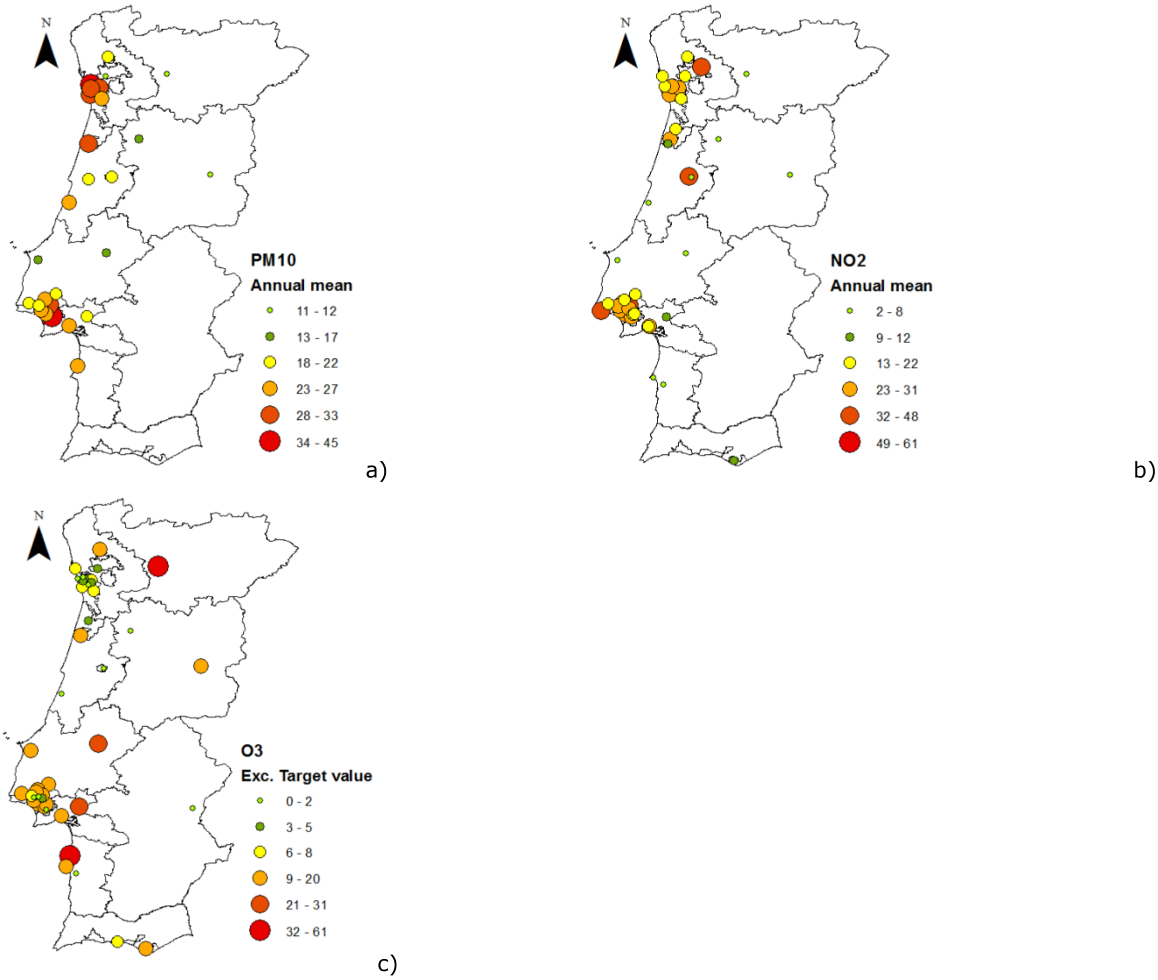


Figure 1.8– Annual mean concentrations of PM10 in 2011 ($\mu g.m^{-3}$) (a); Annual mean concentration of NO_2 in 2011 ($\mu g.m^{-3}$) (b); Number of exceedances to the target value of O_3 in 2011 (c).

The annual mean of PM10 and NO_2 was surpassed in Portugal, mainly in the two biggest urban areas (Porto and Lisboa). Also, there was a high number of exceedances to the O_3 target value (120 $\mu g.m^{-3}$) in several air quality stations in Portugal.

The combination of high emissions and unfavourable weather conditions that promote photochemical formation and reduce atmospheric dispersion, will guide to air pollution episodes. Usually, the main measures for air pollution control are implemented through emission controls (Fenger, 2009). However, to confirm the effectiveness of air quality regulations and improve air quality management efforts, long-term air quality trends must be analysed and explored. Meteorology plays an important role in air pollution formation, dispersion, transport and dilution. Therefore, the variations in local meteorological conditions, e.g., wind speed and direction, temperature and relative humidity, can affect the temporal and spatial variations of air quality pollutants (Duenas et al., 2002; Stasangi et al., 2004; Elminir, 2005). Hence, the strong linkage between weather conditions and pollutant levels can obscure the effects of changing emission levels over time; a better understanding of the effects of meteorology on air quality is important (Flaum et al., 1996).

Climate change is expected to lead to long-term seasonal changes in weather patterns, which are likely to affect the concentrations and dispersion of pollutants in the atmosphere (Thambiran and Diab, 2010). Jacob and Winner (2009) considered that changes in climate affect air quality by perturbing ventilation rates (wind speed, mixing depth, convention and frontal passages), precipitation scavenging, dry deposition chemical production and loss rates, natural emissions and background concentrations.

It is important to foresee the possible impacts of climate change on the concentration and dispersion of air pollutants in order to be ahead and improve the air quality management over a region.

1.1.3 Air quality modelling under climate change

Air quality models are powerful tools to assess the influence of future climate scenarios on the air pollutants concentrations and consequently in air quality management.

Air quality models are able to reproduce and help to understand the physical and chemical transformations and the removal processes of gaseous and particulate pollutants in the atmosphere, and so they have been used worldwide (Bessagnet et al., 2004; Morris et al., 2005; Wyat Appel et al., 2007; Pay et al., 2010).

Air quality models use mathematical and numerical techniques to simulate the physical and chemical processes that affect air pollutants as they disperse and react in the atmosphere. Based on inputs of meteorological data and source information like emission rates and stack height, these models are designed to characterize primary pollutants that are emitted directly into the atmosphere and, in some cases, secondary pollutants that are formed as a result of complex chemical reactions within the atmosphere. The most commonly used air quality models include three big groups of models: dispersion - typically used in the permitting process to estimate the concentration of pollutants at specified ground-level receptors surrounding an emissions source; photochemical - usually used in regulatory or policy assessments to simulate the impacts from all sources by estimating pollutant concentrations and deposition of both inert and chemically reactive pollutants over large spatial scales;

and receptor – consists in observational techniques which use the chemical and physical characteristics of gases and particles measured at source and receptor to both identify the presence of and to quantify source contributions to receptor concentrations.

Photochemical are applied at multiple spatial scales from local, regional, national, and global. There are two types of photochemical air quality models commonly used in air quality assessments: the Lagrangian trajectory model that employs a moving frame of reference, and the Eulerian grid model that uses a fixed coordinate system with respect to the ground. Earlier generation modeling efforts often adopted the Lagrangian approach to simulate the pollutants formation because of its computational simplicity. However, the disadvantage of Lagrangian approach is that the physical processes it can describe are somewhat incomplete. Most of the current operational photochemical air quality models have adopted the three-dimensional Eulerian grid modeling mainly because of its ability to characterize physical processes in the atmosphere in a comprehensive way and predict the species concentrations throughout the entire model domain. Nowadays there are several models that could simulate the chemistry of atmosphere at long-term and at regional scale. As example the American models Comprehensive Air quality Model with extensions (CAMx) and Community Multi-scale Air Quality Model (CMAQ) (Tesche et al., 2006), the Australian model The Air Pollution Model (TAPM) (Hurley et al., 2005) and the European models CHIMERE (Van Loon, 2004; Vautard et al., 2007) and LOTOS-EUROS (Schaap et al., 2008). In these air quality models, the chemical processes are treated independently of the meteorological model, i.e. “offline”. This approach is computationally attractive when weather influences on the air chemistry is not the focus of a study, since it permits to carry sets of chemical transport simulations with a single meteorological dataset.

The analysis of the effect of climate change on air quality will require an increase in model simulations confidence for this purpose. In order to perform this kind of analysis it is important to understand that no single model is capable of reproduce a sufficiently wide range of spatial and temporal scales to address all issues related to air pollution and climate change. Global models have a limit resolution, which currently is 0.5 or 1 degree due to computational requirements; on the other hand, with local and regional models it is difficult to account the processes that occur outside the modelled domain, i.e. at a global scale. Figure 1.9 illustrates the current numerical models classification according to the spatial and time scale of the atmospheric phenomena they are design to perform.

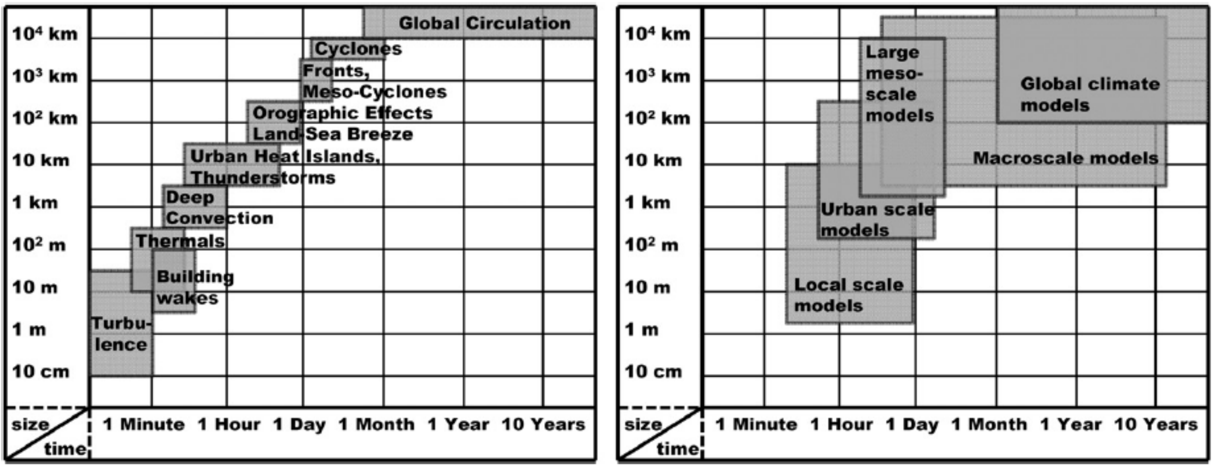


Figure 1.9- Characteristic spatial scales versus characteristic timescales. Left panel: common atmospheric phenomena. Right panel: atmospheric models covering different spatial and temporal scales (Isaksen et al., 2009).

Air quality transcends all scales in the atmosphere from the local to the global with handovers and feedbacks at each scale interaction (Stwart et al., 2004; Monks et al., 2009; Thambiran and Diab, 2010). In the past, scientific analyses of the causes, dynamics and impacts of climate change have predominantly focused on the role of CO_2 and other gases covered by the Kyoto Protocol disregarding the common air pollutants. On the other hand, the research on air pollution only considered the studies at regional and local levels, not taking in account in many cases the global scale. Nowadays there are many studies performed to investigate the effect of changing climate on future traditional air pollutants (Carvalho et al., 2010; Lacressonnière et al., 2014; Penrod et al., 2014; Trail et al., 2014).

There are three main approaches to study air quality under future climate by the application of air quality modelling systems:

- the studies that only consider the effect of climate change, keeping anthropogenic emissions at constant values (Langner et al., 2005; Meleux et al., 2007; Racherla and Adams, 2008; Zhang et al., 2008; Fiore et al., 2011; Tai et al., 2012; Manders et al.; 2012);
- the studies that keep the meteorology conditions invariant (same as the reference year) in future scenarios and change the pollutant emission scenarios (Dentener et al., 2005; Zhang et al., 2010);
- the studies that consider the effect of climate change along with the modification of the anthropogenic emissions in the future scenarios (Unger et al., 2006; Dentener et al., 2006; Stevenson et al., 2006; Tagaris et al., 2007; Nolte et al., 2008; Lei et al., 2012; Coleman et al.,

2013; Doherty et al., 2013; Gao et al., 2013; Jiang et al., 2013; Colette et al., 2013; Penrod et al., 2014; Trail et al., 2014;Lacressonnière et al., 2014).

The vast majority of the studies assess the impacts of climate change and pollutant emissions on O_3. Climate change will affect the factors that contribute toward regulating O_3, such as temperature, water vapour, cloud cover and precipitation, and will contribute to possible variations in O_3 concentration levels (Hogrefe et al., 2004; Lam et al., 2011; Coleman et al., 2013; Gao et al., 2013). As an example of combined impact of climate change and anthropogenic emissions to study O_3, Lacressonnière et al. (2014) simulated changes in future air quality in Europe for the 2030s and 2050s, under the RCP8.5 scenario. Simulations showed an increase in surface ozone in north-western Europe and a decrease in southern areas in the future horizons studied. Over Europe, average O_3 levels steadily increase with at a rate of around 3 $\mu g.m^{-3}$ per decade in summer time. The tropospheric ozone budget is found to be dominated by enhanced stratosphere-troposphere exchanges in future climate while the chemical budget is significantly reduced. Also, the results point out that a NO_x-limited chemical regime will stretch over most of Europe, including especially Western France in the future. Over Portugal the O_3 concentrations may increase by 20 $\mu g.m^{-3}$ in July (Carvalho et al., 2010).

In the last years, the studies besides assessing the impacts on O_3, also evaluate the impacts on PM2.5 (Tagaris et al., 2007; Racherla and Adams, 2008; Zhang et al, 2010; Lam et al., 2011; Tai et al., 2012; Jiang et al., 2013; Colette et al. 2013; Penrod et al., 2014; Trail et al., 2014;) and the impacts on PM10 (Carvalho et al., 2010; Manders et al., 2012). The studies which have examined the impact of climate change on surface PM concentrations showed some differences on the conclusions, including the sign of the effect and in the selection of the most important driving of the sensitivity of PM to climate change. In Carvalho et al. (2010) the results show, throughout Portugal, the maximum increases are foreseen for the northern coastal region in September reaching nearly 30 $\mu g.m^{-3}$.

As mentioned before, there are already previous studies that have included the impacts of both climate and emission changes in several aspects, however most of the studies focused only in O_3 and PM (mainly PM2.5), not considering critical pollutants as PM10 and NO_2. Usually the period of simulation is less than 3 years (or season/month), which should be at least of 5 years to be a period long enough to obtain a climate signal. In terms of spatial resolution, a big part of the air quality studies performed under climate change focus on global or regional level, not reaching a higher detail, as example at the urban level.

1.2 Research objectives and structure of the work

The main goal of this work is to assess the impacts of climate change on air quality in 2050 over Portugal and Porto urban area. To reach this goal, the air quality modelling system, Weather Research & Forecasting (WRF, Skamarock et al., 2008) combined with CAMx, was tested and applied. Several required inputs, such as meteorological fields, air pollutant emissions and chemistry boundary and initial conditions, were meticulous prepared to perform all the simulations.

The structure of this thesis is shown in Figure 1.10.

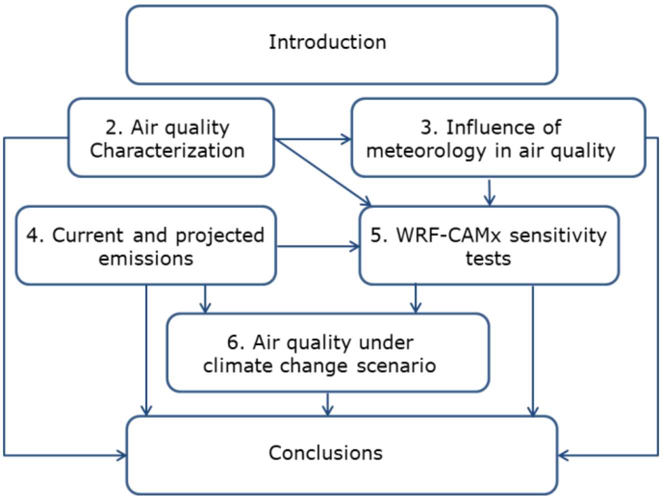


Figure 1.10 – Thesis structure.

The work follows an integrated approach: characterization of air quality in Portugal; evaluation the influence of meteorology in historical air quality data; definition of the national emission scenarios based on the RCP scenarios; selection of the most accurate air quality modelling system to apply over Portugal and Porto urban area; and application the air quality modelling system under climate change to assess air quality in Portugal and Porto urban area in 2050.

The historical data collected through a ten years period, between 2002 and 2012 is analysed in Chapter 2, to verify the national air quality problems. The analysis focused on the most relevant monitored air pollutants: PM10, NO_2, O_3, particulate matter with a 50% efficiency cut-off at 2.5 μm aerodynamic diameter (PM2.5), carbon monoxide (CO), volatile organic compounds (VOCs), SO_2 and C_6H_6. Then, in Chapter 3, the influence of the meteorology on air quality in Portugal is explored and, in this scope, the Kolmogorov-Zurbenko (KZ) filter (Rao and Zurbenko, 1994) is applied to the time-series of the air quality data analysed in Chapter 2 (of PM10, NO_2 and O_3) and to the measured meteorological data. The main outcomes of these two chapters are: the definition of the case study (Portugal and Porto urban area); the identification of the critical air pollutants in Portugal (NO_2, PM10 and O_3); and through historical data, understand that the meteorology can influence the concentration values of some air pollutants.

In Chapter 4 the national current emissions are estimated with a higher detailed disaggregation to improve the performance of the air quality simulations for current conditions. Also, in this chapter, a new emissions model was developed to estimate the national emissions based on the RCP scenarios, being one of the main innovations of this thesis, since allows to keep the consistency of the climate change simulations performed in Chapter 6, in terms of anthropogenic emissions.

In Chapter 5 the air quality modelling system WRF-CAMx is tested and evaluated to adequately assess the best options for its application over Portugal.

The impacts of climate change on air quality over Portugal are evaluated in Chapter 6, through the application of WRF-CAMx modelling system. Two applications are performed, the first only consider the effect of climate change, preserving current anthropogenic emissions, and the other, besides the effect of climate change, includes anthropogenic emissions based on future scenarios. Three 5-year period simulations were performed for historical period and two for future period (2050) over Portugal, Northern Region and Porto metropolitan domains.

Finally, in Chapter 7 the summary of the main results is carried out. Additionally, the general conclusions are explored and possible future developments discussed.

Some of the work presented in this study were performed under some projects of the research group on Atmospheric Emissions Modelling and Climate change (GEMAC) in which the author is integrated, namely the protocol with CCDR-N to develop the Northern Region Air Quality Plans, as well as the SMARTDECISION, MAPLIA and CLICURB projects. In all projects, the author participated dynamically in several tasks: air quality characterization and analysis; preparation of the air quality and meteorological inputs to the air quality modelling simulations; and running all the mesoscale air quality modelling simulations (WRF-CAMx) of these projects.

The vast majority of the contents presented in Chapter 2 to 5 are published or submitted to international peer-reviewed journals and conferences. In most of them, the author was responsible for the study conception and design, as well as, for the manuscript writing. These publications are listed below:

Borrego, C., ***Sá, E.,*** *Carvalho, A., Sousa, J., Miranda, A.I. (2012b) – Plans and Programmes to improve air quality over Portugal: a numerical modelling approach, Int J Environ. Pollut., Vol.48, Nos. 1/2/3/4.*

Borrego C., Monteiro A., ***Sá E.,*** *Carvalho A., Coelho D., Dias D., Miranda A.I. (2012a) - Reducing* NO_2 *Pollution over Urban Areas: Air Quality Modelling as a Fundamental Management Tool, Water Air Soil Pollut. DOI 10.1007/s11270-012-1281-7.*

Sá, E., *Tchepel, O., Carvalho, A., Borrego, C. (submitted). Meteorological driven changes on air quality over Portugal: a KZ filter application. Atmospheric Pollution Research.*

Sá, E., *Ferreira, J., Carvalho, A., Borrego, C. (accepted). Development of current and future pollutant emissions for Portugal. Atmospheric Pollution Research.*

Sá, E., *Martins, H., Carvalho, A., Lopes, M., Miranda, A.I., Borrego, C. (2012) - WRF-CAMx application over an urban area: parameterizations sensitivity study for air quality purposes. AIR QUALITY MANAGEMENT at URBAN, REGIONAL and GLOBAL SCALES, 4th International Symposium and IUAPPA Regional Conference, 10-13 September, Istanbul, Turkey. Eds: S. Incecik, C. Kahya. ISBN: 978-975-561-424-3.*

Coelho, M.C., Fontes, T., Bandeira, J., Pereira, S., Tchepel, O., ***Sá, E.,*** *Amorim, J., Borrego, C. (2014) - Assessment of potential improvements on regional air quality modelling related with implementation of a detailed methodology for traffic emission estimation. Science of the Total Environment, 470–471, 127–137.*

Martins H., ***Sá E.,*** *Freitas S. Amorim J.H., Rafael S., Borrego C. Impacte das alterações climáticas na qualidade do ar do Porto. (2014) - Clima 2014, IV Congresso Nacional sobre Alterações Climáticas. 4-5 Dezembro, Aveiro (Portugal).*

Sá, E.*, Martins, H., Ferreira, J., Almeida, M., Rocha, A., Carvalho, A., Borrego, C. (submitted). Climate change and pollutant emissions impacts on regional and urban air quality in 2050. Atmospheric Environment.*

Chapter 2

2 Mainland Portugal - air quality characterization (2002-2012)

The contents of this chapter were partially published in:

*Borrego, C., **Sá, E.,** Carvalho, A., Sousa, J., Miranda, A.I. (2012b) – Plans and Programmes to improve air quality over Portugal: a numerical modelling approach, Int J Environ. Pollut., Vol.48, Nos. 1/2/3/4.*

*Borrego C., Monteiro A., **Sá E.,** Carvalho A., Coelho D., Dias D., Miranda A.I. (2012a) - Reducing NO_2 Pollution over Urban Areas: Air Quality Modelling as a Fundamental Management Tool, Water Air Soil Pollut. DOI 10.1007/s11270-012-1281-7*

Abstract

The main purpose of the air quality management is protecting human health and the environment as a whole, by controlling the air quality concentration levels, taking in account the existing target values and limit values of ensuring effective protection against harmful effects on human health and vegetation and ecosystems. In this sense, hourly values are measured in each air quality station for several pollutants in order to assess air quality and provide the basis for a correct air quality management. This work intends to evaluate and characterize the air quality over mainland Portugal, for the period between 2002 and 2012, taking into account the analysis of the legislation fulfillment over the monitoring sites. The results show that NO_2, PM10 and O_3 are critical pollutants in Portugal with exceedances every year since 2002. Two Air Quality Plans implemented in the Northern region of Portugal for NO_2 and PM10 are analysed. For NO_2, traffic is the main sector with contribution for the measured concentration values. On the other hand, several sectors, besides traffic, contribute for the PM10 levels, such as residential combustion, industry, among others. With the measures selected for each pollutant, for the sectors with higher contribution, NO_2 and PM10 concentrations decrease, however some air quality stations are still in incompliance.

2.1 Introduction

Air pollution can be defined by being a contamination of the indoor or outdoor environment by any chemical, physical or biological agent that modifies the natural characteristics of the atmosphere. Some of the most common sources of air pollution are household combustion devices, motor vehicles, industrial facilities and forest fires.

Pollutants of major public health concern include PM, carbon monoxide (CO), O_3, NO_2, VOCs, SO_2 and C_6H_6.

PM is the term for solid or liquid particles found in the air. Concentrations of PM comprise primary particles emitted directly into the atmosphere and secondary particles formed by chemical reactions in the air. The origin of PM concentrations is emissions from natural or anthropogenic sources. The most usual natural sources that influence the PM concentrations in Portugal are the Saharan dust, forest fires and sea spray (Borrego et al., 2008). The anthropogenic sources of PM are usually located in the urban and industrial areas. Over the urban areas, the emission of PM10 is related to residential combustion, constructions and traffic (Borrego et al., 2010). The major concerns for human health from exposure to PM include: effects on breathing and respiratory systems, damage to lung tissue, cancer, and premature death. The elderly, children, and people with chronic lung disease, influenza, or asthma, are especially sensitive to the effects of PM. In the Directive, PM is categorized taking in account the size of the particles and is divided into two main groups:PM10 – PM with a 50% efficiency cut-off at 10 μm aerodynamic diameter and PM2.5 – PM with a 50% efficiency cut-off at 2.5 μm aerodynamic diameter.

CO is a primary pollutant, formed from incomplete combustion of carbon containing fuels. The largest source is road transport, with residential and industrial combustion making significant contributions. CO is an odorless, colorless and toxic gas since it is impossible to see, taste or smell. Substantially reduces capacity of the blood to carry oxygen to the body's tissues and blocks important biochemical reactions in cells. People with existing diseases, which affect delivery of oxygen to the heart or brain, such as angina, are at particular risk. At extremely high levels, CO can cause death.

O_3 is a colorless gas found in the air we breathe and, it can be good or bad depending on where it occurs: naturally in the Earth's upper atmosphere (the stratosphere), where it shields the Earth from the sun's ultraviolet rays; or at ground-level, ozone is an air pollutant that can harm human health.

O_3 at ground-level is the one that is important in terms of air quality management because is the one that people contact. It is not emitted directly from any human-made source and because of this is considered a secondary photochemical pollutant. It arises from chemical reactions between various air pollutants, primarily NO_X and VOCs, initiated by strong sunlight. The atmospheric chemistry involved in O_3 formation is complex. It takes time for the ozone to accumulate as the chemical reactions involved are quite slow - O_3 builds up in polluted air masses. This takes several days and is favored by prolonged sunny weather and low wind speeds. The distribution and the formation of O_3 are affected by air movement. Although most of the pollutants that form O_3 (O_3 precursors) are generated in urban areas, concentrations of tropospheric O_3 tend to be higher away from towns. Some people are at especially high risk for health problems associated with O_3. O_3 can affect the health by irritating the respiratory system, causing coughing, throat irritation, and/or an uncomfortable sensation in the chest. Even relatively low levels of ozone can cause health effects and O_3 exposure may also increase the risk of premature death from heart or lung disease.

The two most prevalent NO_x are NO_2 and nitrogen monoxide (NO), which are produced from the reaction of nitrogen and oxygen gases in the air during combustion, especially at high temperatures. All combustion processes in air produce NO_x, being road transport the main source, followed by the electricity supply industry and other industrial and commercial sectors. Taking in consideration the concentrations normally presents in the atmosphere, NO is not considered as a dangerous pollutant. On the other hand, NO_2 is one of the most critical air pollutants because the significant impacts on human health (extremely high-dose exposure or low level exposure), apart from giving rise to acid rain and other air pollutants, such the contribution to the formation of photochemical smog. In terms of human health impacts, NO_2 acts mainly as an irritant affecting the mucosa of the eyes, nose, throat, and respiratory tract.

VOCs are an important class of air pollutants, commonly found in the atmosphere at ground level in all urban and industrial centres. VOCs are emitted as gases from certain solids or liquids and include a variety of chemicals, some could be encounter on a day-to-day basis: paints, paint strippers, and other solvents; wood preservatives; aerosol sprays; cleansers and disinfectants; moth repellents and air fresheners; stored fuels and automotive products; hobby supplies; dry-cleaned clothing. The ability of organic chemicals to cause health effects varies greatly from those that are highly toxic, to those with no known health effect. Many organic compounds are known to cause cancer in animals; some are suspected of causing, or are known to cause, cancer in humans.

SO_2 is a colorless gas with a strong odor at high concentrations. It is an irritant to the mucous membranes of the pollutant eyes and respiratory tract, may cause acute and chronic health effects, especially at respiratory level. In more sensitive groups, such as children, it may be related to the emergence of respiratory problems (as asthma or whooping cough). This pollutant is very water soluble, originating sulfuric acid (H_2SO_4), which contributes to the formation of acid rain, with the consequent acidification of water and soil and plants damage and materials degradation. The industrial sector is the largest contributor to SO_2 emissions, especially through combustion in boilers and refineries, where are burned fuels with high sulfur content.

C_6H_6 is a natural constituent of crude oil, and is one of the most elementary petrochemicals. Benzene is an aromatic hydrocarbon and it is sometimes abbreviated Ph–H. Benzene is a colorless and highly flammable liquid with a sweet smell. It is mainly used as a precursor to heavy chemicals, such as ethylbenzene and isopropylbenzene, which are produced on a billion kilogram scale. Most non-industrial applications have been limited by benzene's carcinogenicity.

Human health and the environment are adversely affected by air pollution, both in the short and long term. It negative effects on health, can range from minor respiratory irritation to cardiovascular diseases and premature death; on ecosystems, leads to corrosion and soiling of materials, including those used in objects of cultural heritage.

The concentrations of the air pollutants are subject to a range of atmospheric processes including atmospheric transport, mixing and chemical transformation, before exposure to humans or ecosystems may occur. Also, air pollutants do not remain in the atmosphere forever, depending on their physical-chemical characteristics and factors such as atmospheric conditions or roughness of receiving surfaces, they may be deposited after either short (local, regional) or long-range (European, inter-Mainland) transport. Pollutants can be washed out of the atmosphere by precipitation (rain, snow, fog, dew, frost and hail) or deposited dry as gases or particulate matter, for example directly on vegetation surfaces such as crop or tree leaves (EEA, 2010a).

The state of air pollution is often expressed as Air Quality, which consists in measure the concentrations of gaseous pollutants and size or number of particulate matter, taking in account, not only the anthropogenic perturbation of the ‘‘natural’’ atmospheric state, but also, considered in the wider context of the interactions with biogenic and other natural emissions that may have feedbacks with atmospheric composition and climate (Monks et al. 2009).

Air pollution can basically be regulated by emission standards, air quality standards, and cost benefit analyses. The most common way is to limit the emission from a source, a sector or an entire country; the pragmatic way is to state how much concentration there may be in a particular ambient air. The first approach has been attempted for centuries although with limited success. The second is fairly new, being dependent upon more or less sophisticated measurement and computational techniques. Further it is important to be noted that the time pattern of air pollution is relevant in relation to impacts. As an example to material damage – and to some extent damage on ecosystems – long term averages determine the impact, whereas for health and wellbeing often short term peak values are decisive. Therefore different air quality standards, and to some extent emission standards, are established (Fenger, 2009).

In order to reduce and control the effects of air pollution on human health and in the environment, the air quality directives require the EU member states to assess air quality through their territory.

In Portugal, the national air quality network had around 80 air quality stations in 2012. These stations monitor for a variety of air pollutants included the pollutants of major public health concern. The distribution by the territory of the air quality stations follows the Directive 2008/50/EC requirements in order to spatial characterize the national air quality.

Portugal, over the last years, is facing air quality problems because has been surpassing the legislated limit values at several air quality monitoring stations (Monteiro et al., 2007, Borrego et al., 2012a, 2012b), mainly over two main regions: Lisboa and Vale do Tejo region and the Northern Region, for three pollutants PM10, NO_2 and O_3.
This chapter intend to analyse the historical data collected through a ten years period, between 2002 and 2012, since was in 2002 that the air quality monitoring network was almost completed and operational. The analysis will focus on the most relevant monitored air pollutants.

2.2 Data and Methods

In order to characterize the air quality in Portugal is necessary to have a thorough knowledge of the legislation applied over the study region and of the monitoring network where the data is collected. Hence, firstly the legislative framework is described and the air quality national network is presented. After, is given an explanation of the applied methodology to characterize the air quality of the study region for the period since the year 2002 until 2012, for seven pollutants: CO, SO_2, C_6H_6, NO_2, PM10, PM2.5 and O_3.

2.2.1 Legislative framework

Air quality is one of the environmental areas in which the European Union (EU) has been most active, namely in the design and implementation of legislation and on the restriction of pollutants emissions to the atmosphere. The air quality Framework Directive (FD) (Directive 96/62/CE of 27 September 1996) established the obligations of the EU member states (MS) and redefined the guidelines for the assessment and management of air quality, namely the creation of an air quality management system comprising measuring networks, pollutant emission inventories and air quality modelling. Also, the FD states the obligation to elaborate and implement Air Quality Plans (AQPs) (previously known as Plans and Programmes) to improve air quality when the legislation standards are not fulfilled. The FD was transposed into the Portuguese legislation by the Decree-Law (DL) nr 276/99 of 23 July 1999. In order to state limits for specific pollutants, four "daughter" directives emerged later, which were also transposed to national legislation by three decree-laws (DL nr 111/2002, DL nr 320/2003, DL nr 351/2007).

In May 2008, a new Directive on Ambient Air Quality and Cleaner Air for Europe (Directive 2008/50/EC) was published, integrating the former FD, three of the four previous “daughter” directives and one Council decision into a single document. The new Directive was transposed to national legislation by the DL nr 102/2010 of 23 September 2010, and nowadays this is the operative document in Portugal. The evolution of the European and national legislation regarding air quality management is illustrated in Figure 2.1.

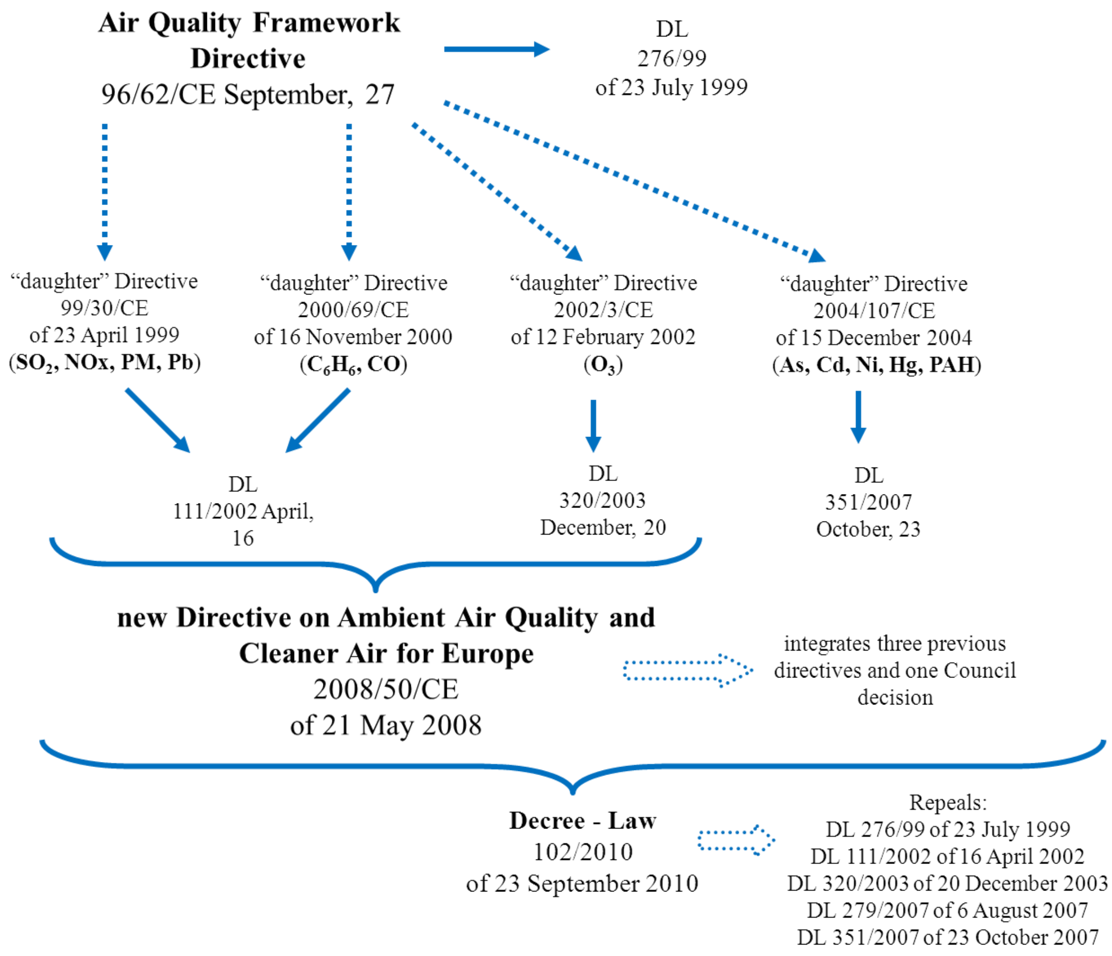


Figure 2.1 - European and Portuguese air quality legislation framework.

The Directive 2008/50/EC introduces new objectives for fine particles, but does not change existing air quality standards for the other pollutants, such as SO_2, oxides of nitrogen (NOx), PM10, PM2.5, O_3, benzene (C_6H_6) and CO. In order to regulate their concentrations in the atmosphere, the following definitions were included on the Directive:

- Limit Value (LV) – shall mean a level fixed on the basis of scientific knowledge, with the aim of avoiding, preventing or reducing harmful effects on human health and/or the environment as a whole, to be attained within a given period and not to be exceeded once attained;

- Margin of Tolerance (MT) – shall mean the percentage of the limit value by which that value may be exceeded subject to the conditions laid down in this Directive;

- Target Value (TV) – shall mean a level fixed with the aim of avoiding, preventing or reducing harmful effects on human health and/or the environment as a whole, to be attained where possible over a given period;

- Long-Term Objective (LTO) – shall mean a level to be attained in the long term, save where not achievable through proportionate measures, with the aim of providing effective protection of human health and the environment;

- Alert Threshold (AT) – shall mean a level beyond which there is a risk to human health from brief exposure for the population as a whole and at which immediate steps are to be taken by the Member States;

- Information Threshold (IT) – shall mean a level beyond which there is a risk to human health from a brief exposure for particularly sensitive sections of the population and for which immediate and appropriate information is necessary.

The air quality requirements defined on the DL nr 102/2010 of 23 September 2010 for these air pollutants are indicated on Table 2.1.

Table 2.1 – Air quality requirements defined on the DL nr 102/2010 of 23 September 2010.

Air pollutants	Criteria	Limit value ($\mu g.m^{-3}$)	Margin of tolerance ($\mu g.m^{-3}$)
CO	Maximum daily 8hr mean	10 000	-
SO_2	One hour	350 [1]	2002 – 90 2003 – 60 2004 – 30 2005 – 0
	One day	125[2]	-
C_6H_6	Calendar year	5	2002 – 5 2003 – 5 2004 – 5 2005 – 5 2006 – 4 2007 – 3 2008 – 2 2009 – 1 2010 – 0
NO_2	One hour	200[3]	2002 – 80 2003 – 71 2004 – 60 2005 – 50 2006 – 40 2007 – 30 2008 – 20 2009 – 10 2010 – 0
	Calendar year	40	2002 – 16 2003 – 14 2004 – 12 2005 – 10 2006 – 8 2007 – 6 2008 – 4 2009 – 2 2010 – 0
PM10	Calendar year	40	2002 – 5 2003 – 3,4 2004 – 1,8 2005 – 0
	One day	50[4]	2002 – 15 2003 – 10 2004 – 5 2005 – 0
		Target value	**Long term objective (LTO)**
O_3	Maximum daily 8hr mean ($\mu g.m^{-3}$)	120[5]	120
	Purpose	**Averaging period**	**Threshold**
	Information ($\mu g.m^{-3}$)	1 hour	180
	Alert ($\mu g.m^{-3}$)		240

- [1] not be exceeded more than 24 times a calendar year
- [2] not be exceeded more than 3 times a calendar year
- [3] not be exceeded more than 18 times in a calendar year
- [4] not be exceeded more than 35 times in a calendar year
- [5] not be exceeded more than 25 times a calendar year averaged over three years

One of the main MS obligations, revealed on the Directive, is the implementation of AQPs when the air pollutant concentrations exceed the air quality standards in zones or agglomerations. The implementation of AQPs should be based on the design of measures to reduce the pollutant atmospheric concentrations and meet the legal requirements. These measures were discussed with the entities involved in their implementation and finally a list of selected measures is designed. The Execution Programme (EP), which is the Portuguese legal document that promotes and enforces the application of the emission reduction measures, describes these selected measures in detail. Figure 2.2 summarizes the entire process from the elaboration of the AQP to the Execution Programme.

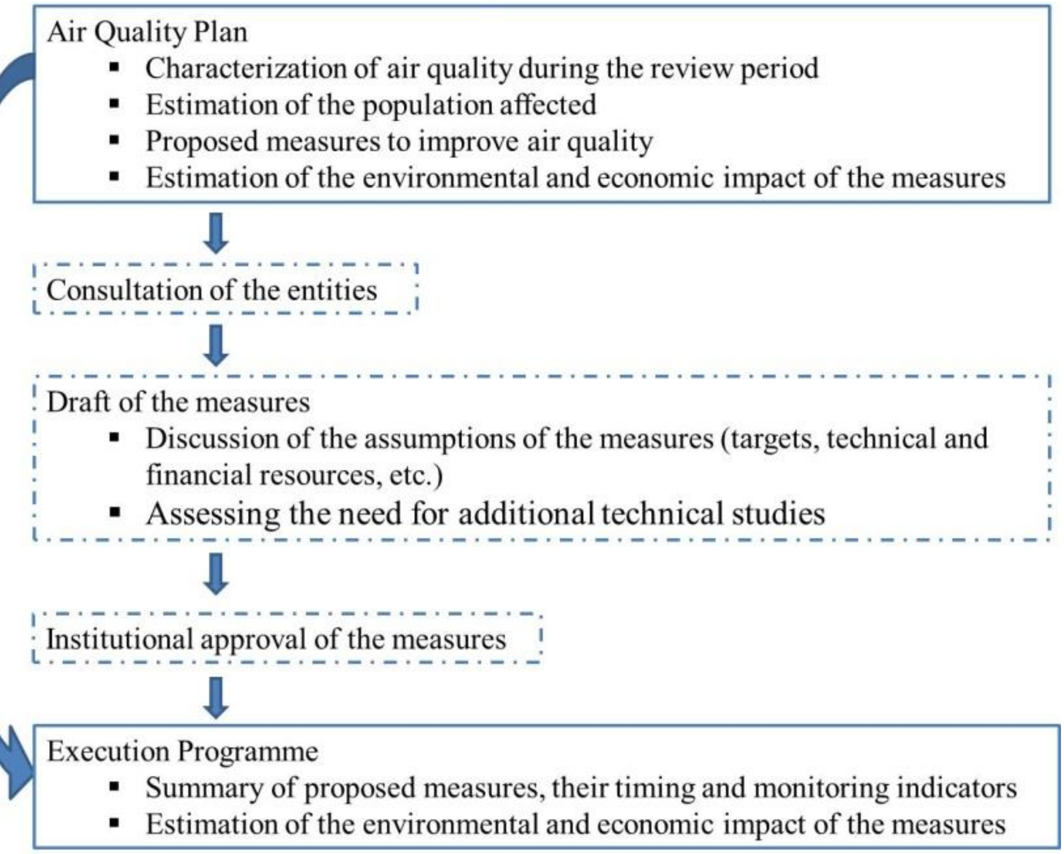


Figure 2.2 - Process related to the design of the Execution Programme of an AQP.

The air quality requirements for each pollutant established on the Directive (as in the DL nr 102/2010 of 23 September 2010) considered standard values for human health protection and protection of ecosystem. In this study only will be focus the human health effects of air pollution, will not be taken in account the protection of ecosystems.

Additionally, this Directive indicates that for most of the pollutants the minimum data capture is of 90% and this value does not include losses of data due to regular

calibration or the normal maintenance of the instrumentation. Only for ozone the criteria for the required proportion of valid data changes (75% or 90%) because depends on the statistical parameter that is to calculate.
On the treatment of the air quality data, there is another important document for the required proportion of valid data, the Decision 2001/752/EC of 17 October 2001, which defined that:

- for one-hour values: minimum data capture 75%;
- for 24-hour values: at least 13 one-hour values available, not more than six successive one-hour values missing;
- for mean and the median: minimum data capture 50%;
- for daily 8 hours mean from hourly running 8 hours: minimum data capture 75%.

This Decision is now being analysed for an update by the European Commission, however at the time of this study there was no new document published.

2.2.2 The air quality monitoring network

In Portugal, the assessment and management of air quality are a legal obligation of the Portuguese Environmental Agency (PEA), at national level, and of the Regionals Coordination and Development Commissions (CCDRs), at regional level. In this sense, in Portugal, there is a national air quality monitoring network, where several air pollutants are continuously measured. The composition of the monitoring network suffers modifications since the beginning of its operation in order to accomplish the criteria of the air quality stations localization mentioned on the legislation.
Taking in account the Directive 2008/50/EC, a zone is a part of the territory of the Member State, delimited by that Member State, for the purposes of air quality assessment and management; an agglomeration is a conurbation with a population in excess of 250 000 inhabitants or, where the population is 250 000 inhabitants or less, with a given population density per km^2 to be established by the Member States. In this sense, the air quality monitoring network in Mainland Portugal is presented in (Figure 2.3):

- 6 Zones in the Northern region, with 3 agglomerations (*Porto Litoral, Vale do Ave e Braga*);
- 5 Zones in the Center region, with 2 agglomerations (*Aveiro/Ílhavo e Coimbra*);

- 4 Zones in the Lisboa and Vale do Tejo region, with 3 agglomerations (*AML Norte, AML Sul and Setúbal*);
- 2 Zones in the Alentejo region;
- 4 Zones in Algarve region, with 3 agglomerations (*Portimão/Lagos, Albufeira/Loulé e Faro/Olhão*).

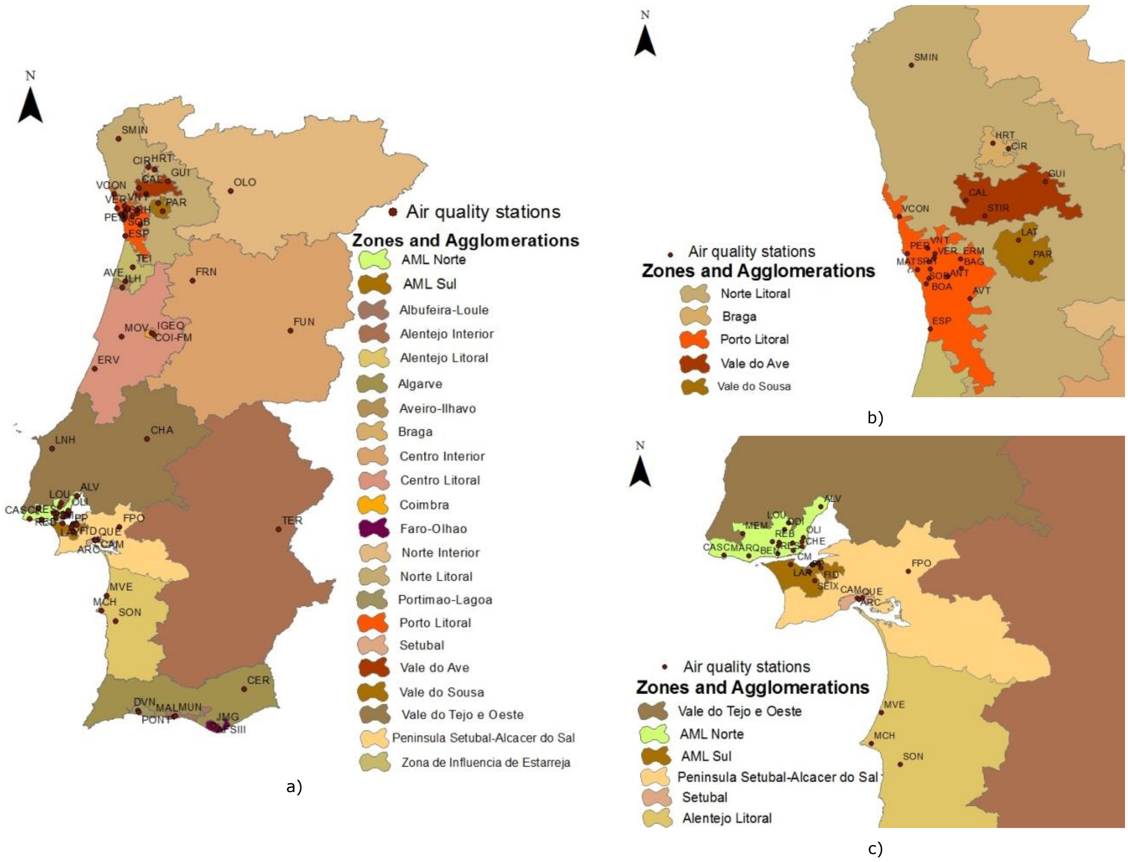
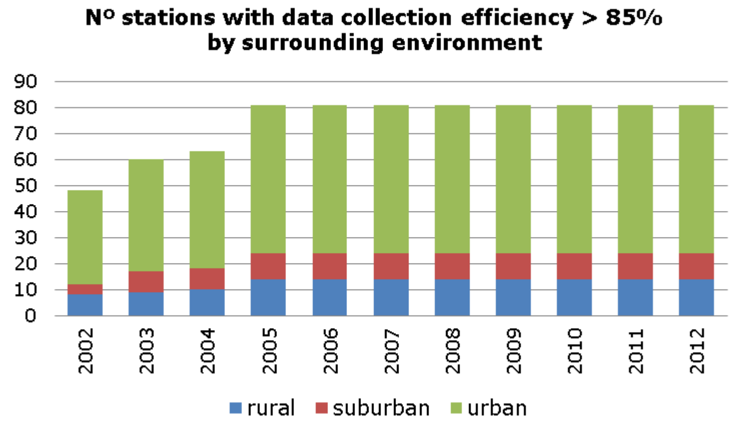
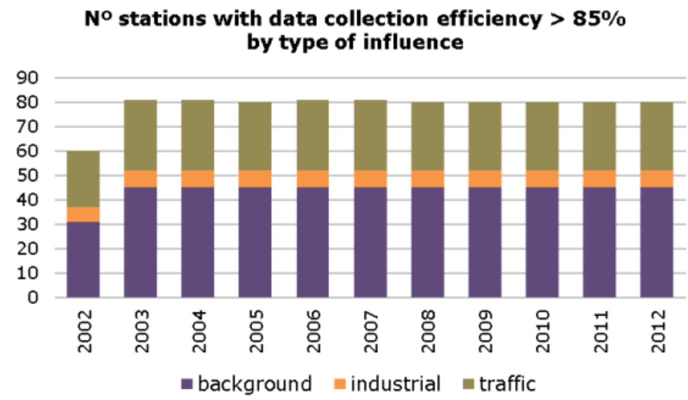


Figure 2.3 - The Mainland Portuguese air quality monitoring network and respective zones and agglomerations (a); part of Northern region(b) and Lisboa and Vale do Tejo region (c).

In Portugal the air quality stations are classified by its type of influence: traffic, industrial and background and the surrounding environment: urban, suburban and rural.Figure 2.4 shows the evolution of the air quality stations that existed in the air quality network from 2002 until 2012 by surrounding environment (a) and type of influence (b). In Appendix A is the classification of all the air quality stations of Mainland Portugal by type of influence and surrounding environment.



a)



b)

Figure 2.4– Number of air quality stations since 2002 until 2012 by surrounding environment (a) and type of influence (b).

The number of urban air quality stations increase between 2002 and 2005, an then it was kept constant. Also, the number of rural air quality stations increase in the same years, having the highest number (12) in the year 2009. The suburban air quality stations were kept constant since 2005. In terms of type of influence the number of background air quality stations increases between 2002 and 2005; the number of industrial air quality stations was constant between 2005 and 2012; and the number of traffic air quality stations do not oscillate significantly between 2003 and 2012. The evolution of the number of air quality stations is due to the fact that the network of national air quality needs to accomplish the requirements established under the Directive. Also, there is a higher number of air quality stations in the litoral region of Portugal in order to fulfil the criteria defined in the Directive, since is where most of the population lives and could be affected by the air quality problems.

2.2.3 Methodology to characterize air quality over Portugal

The air quality characterization is performed through the analysis of the monitoring data from air quality national network, considering the limit and target values defined DL nr 102/2010 of 23 September 2010. The study period starts in 2002 until 2012. The air pollutants considered in this study are: CO, SO_2, VOCs, NO_x, PM (PM10 and PM2.5) and O_3, since these are the pollutants with major public health concern. A long-term evaluation of air pollutants concentrations values registered in Portugal is presented based on hourly values collected.

The analysis of the collected values takes into account the air quality data requirements of the Directive for the minimum data capture and for the proceedings to calculate the exceedances of each pollutant.

Additionally, an analysis of the principal emission sources of the pollutants with more exceedances is presented.

2.3 Results and Discussion

In this section are presented the results from the analysis and comparison of the air quality measurements in the national air quality stations, with the legislated values in order to verify the compliance of the legislation. Also, two AQPs are detailed described for NO_2 and PM10.

2.3.1 Air quality assessment (2002-2012)

All air quality stations over Mainland Portugal were analyzed in terms of data collection efficiency, as is required in the Directive. Figure 2.5 shows the number of air quality stations with annual data collection efficiency above 85%, for CO, SO_2, C_6H_6, NO_2, PM10 and PM2.5 and above 75% for O_3, for the period 2002-2012.

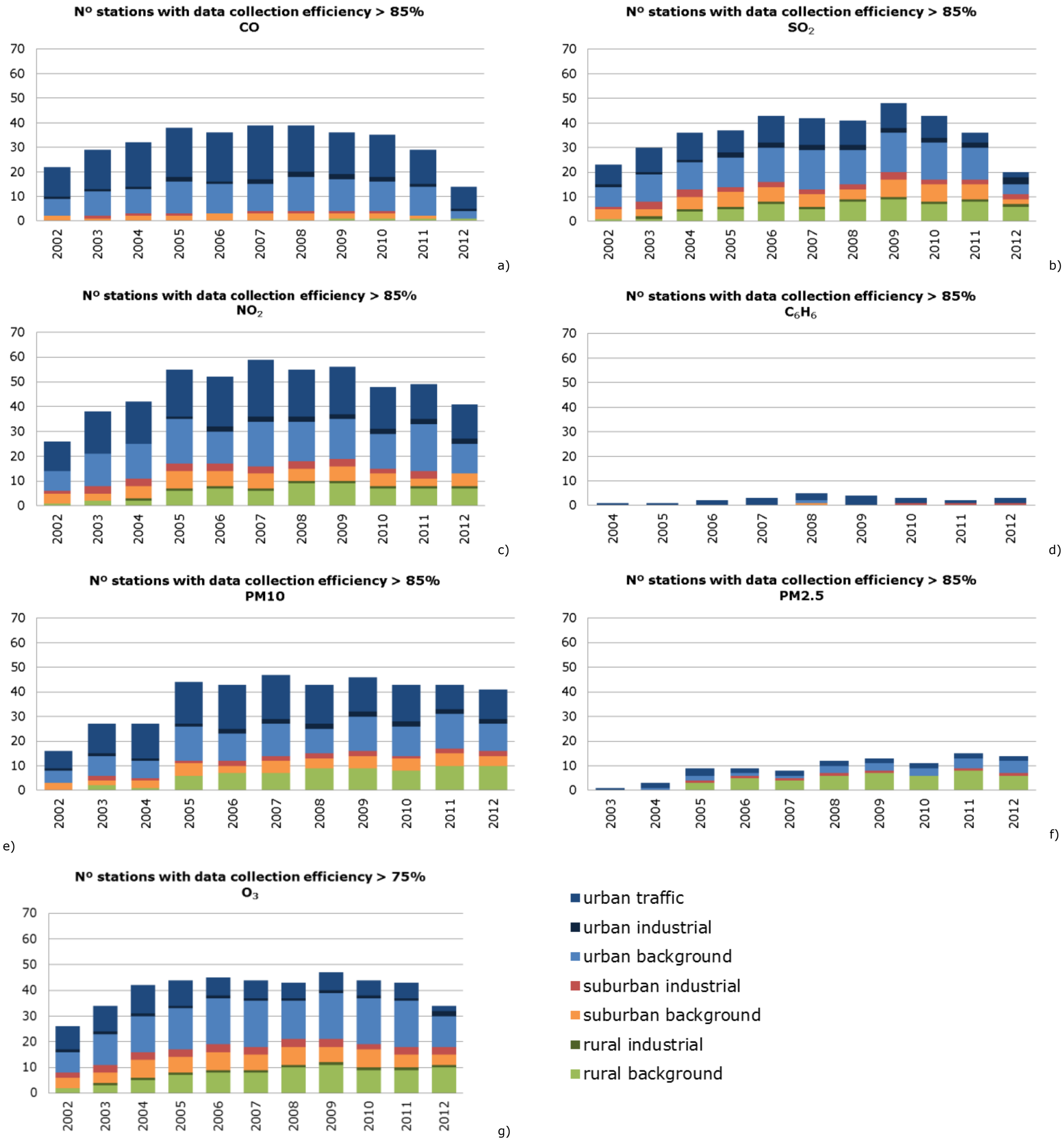


Figure 2.5 - Number of air quality stations in Mainland Portugal for CO(a), SO_2(b), C_6H_6(c), NO_2(d), PM10(e), PM2.5(f) and O_3(g) (2002-2012), with data collection efficiency above 85% and 75% for O_3.

From Figure 2.5 it is possible to verify that over the last years the number of urban background air quality stations increased, namely for SO_2 and O_3 measurements. The number of urban traffic stations decreased for O_3 and were kept constant for the other pollutants. The number of suburban air quality stations remained approximately the same since 2004 for all the pollutants. On the contrary, the number of rural background stations increased for all air pollutants, except for CO. The low number of air quality stations for PM2.5 and C6H6 is justified by the late introduction of air quality requirements for these two pollutants in the legislation. In the last three years, for CO, SO_2, NO_2, PM10 and O_3, there was a decrease of the total number of air quality stations with data collection efficiency higher than 85%, which could be caused by the financial crises that Portugal is facing. It is important to explain that all the air quality stations over Mainland Portugal follow the requirements of the Directive in terms of number and location in order to provide accurate data.

The analysis of the air quality measurements against the legislated air quality requirements will be presented below for each pollutant. In Portugal there are several air quality stations measuring the same pollutant and the purpose of this analysis is to identify the air quality stations in incompliance, so the legends only highlight the air quality stations that are in incompliance.

CO:

Taking in account the air quality requirements for CO, the maximum daily 8hr mean of this pollutant is analyzed for the period 2002-2012 in Figure 2.6.

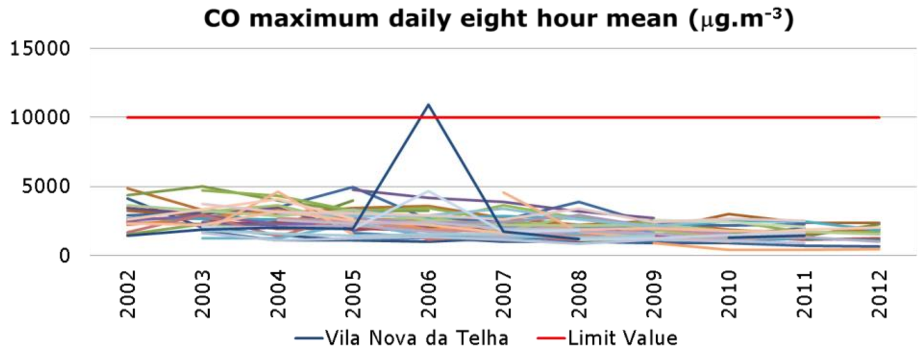


Figure 2.6 – CO maximum 8hr mean concentration values for all air quality stations in Mainland Portugal (2002-2012).

The air quality stations over the study period did not surpassed the limit value of 10 000 $\mu g.m^{-3}$, with the exception of Vila Nova da Telha (VNT) station in 2006 (Figure 2.6). Since the exceedances were just in one year, a detailed analysis of the 8-hr mean CO concentration was performed for that year (Figure 2.7).

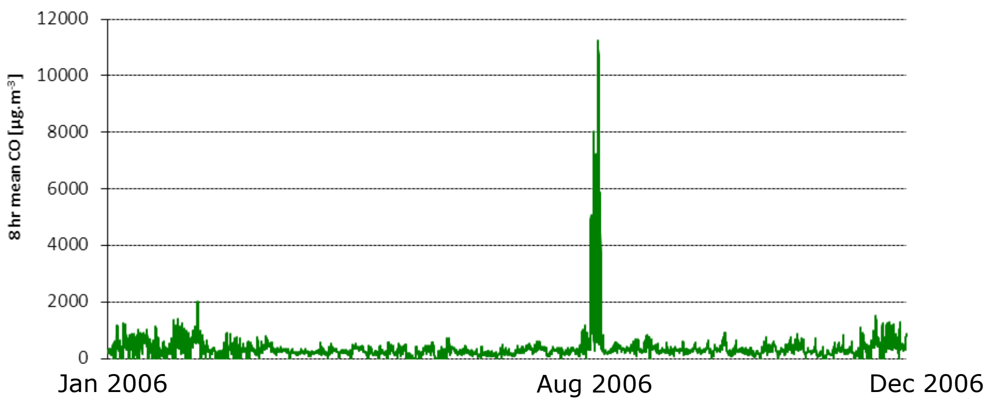
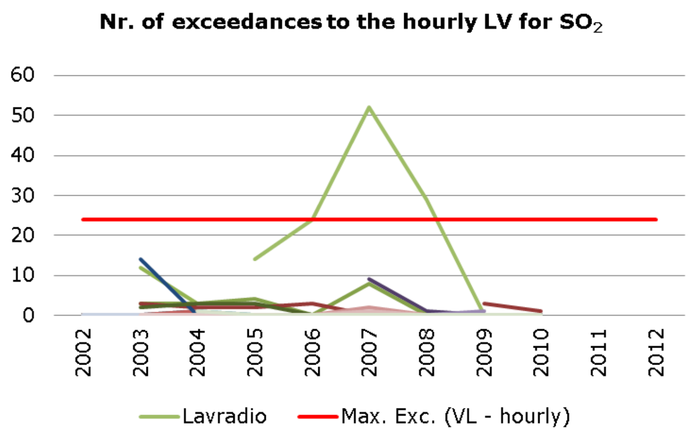


Figure 2.7 - CO 8-hr mean observed in VNT air quality station in 2006.

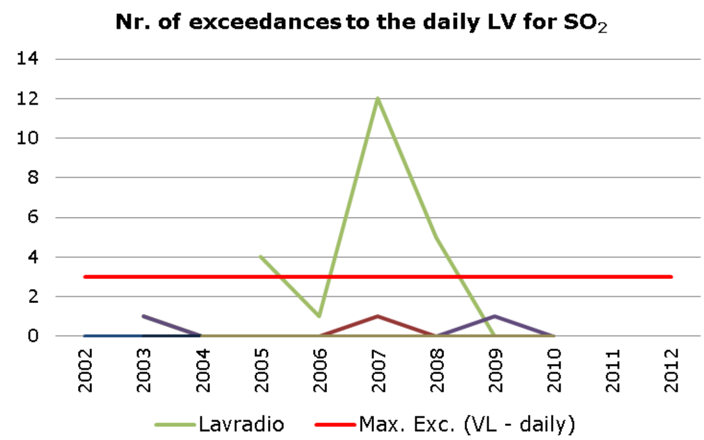
Figure 2.7 allows the identification of an episode occurred in August, 13 of 2006 as the responsible for largest values of CO. Registered data highlight a forest fire occurred in Lidador, a parish of Maia municipality, that started on August, 9. Since that day until August, 14, VNT air quality station registered high concentration values of CO and PM10. Additionally, according to Ferreira et al. (2007), the influence of the forest fires on Porto Litoral agglomeration was detected since August, 5 until August, 13. Even surpassing the LV in 2006, and because it was identified as one time situation, CO is not considered as a critical pollutant.

SO_2:

Figure 2.8 shows the evolution of the number of exceedances to the hourly LV and annual LV for SO_2, from 2002 to 2012.



a)



b)

Figure 2.8 - Number of exceedances to the hourly LV (a) and daily LV for SO_2 (b) (2002-2012).

The hourly and daily LV for SO_2 was surpassed in some air quality stations over Mainland Portugal, however only Lavradio (LAV) air quality station (urban industrial) surpassed the maximum of exceedances allowed by the DL nr 102/2010. These exceedances in LAV station were analysed and it was due to an ammonia industry located in its vicinity. In this sense, the appropriate measures were performed to eliminate the causes of the SO_2 levels measured in that region and it was considered an isolate and controlled episode. For this reason SO_2 was not considered a critical pollutant in Portugal.

C_6H_6:

Figure 2.9 shows the annual mean concentration values of the C_6H_6 measurements for the period with data collection efficiency, 2004-2012.

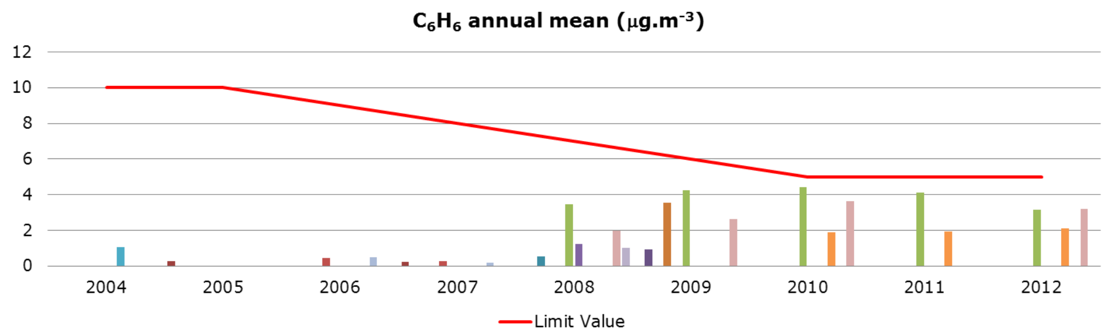


Figure 2.9 – C_6H_6 annual mean concentration values over Mainland Portugal (2004-2012).

Taking in account the benzene LV (5 $\mu g.m^{-3}$) it is possible to observed that this value was not surpassed.

NO_2:

Regarding NO_2, the first part of the analysis was focused on the limit value for one hour, which could not be exceeded more than 18 times in a calendar year. Figure 2.10shows the NO_2 exceedances to the hourly limit value, in each air quality station, since the year 2002.

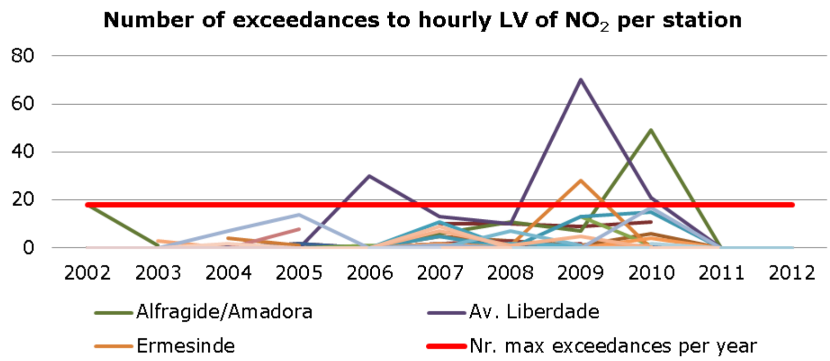


Figure 2.10 – Number of exceedances to NO_2 hourly LV per air quality station in Mainland Portugal (2002-2012).

Three air quality stations surpassed more than 18 times in a calendar year the NO_2 hourly LV, namely Alfragide/Amadora (ALF) (in 2010), Av. Liberdade (AVL) (in 2006 and 2009) and Ermesinde (ERM) (in 2009). The first two air quality stations are located in the Lisboa and Vale do Tejo region and the third in the Northern region. The ALF and ERM are urban background air quality stations and analyzing the exceedances registered in 2010 and 2009, respectively, it seems that were isolated episodes. These exceedances could be justified by the combination of two main factors: specific synoptic conditions and some new and specific emission sources that existed in the period of time when occurred these NO_2 values (Borrego et al, 2012a). In the case of the traffic air quality station (AVL) the exceedances could be justified by the influx of traffic in the surrounding area and for this reason, AQPs should be developed to this air quality station.

The analysis will also be performed considering the legislated values of NO_2 for the calendar year (40 $\mu g.m^{-3}$). Since there are a high number of air quality stations measuring NO_2, the analysis was divided depending on the type of influence of each air quality station. Figure 2.11 shows the comparison between the annual mean, for all the air quality stations, with the LV from 2002 to 2012.

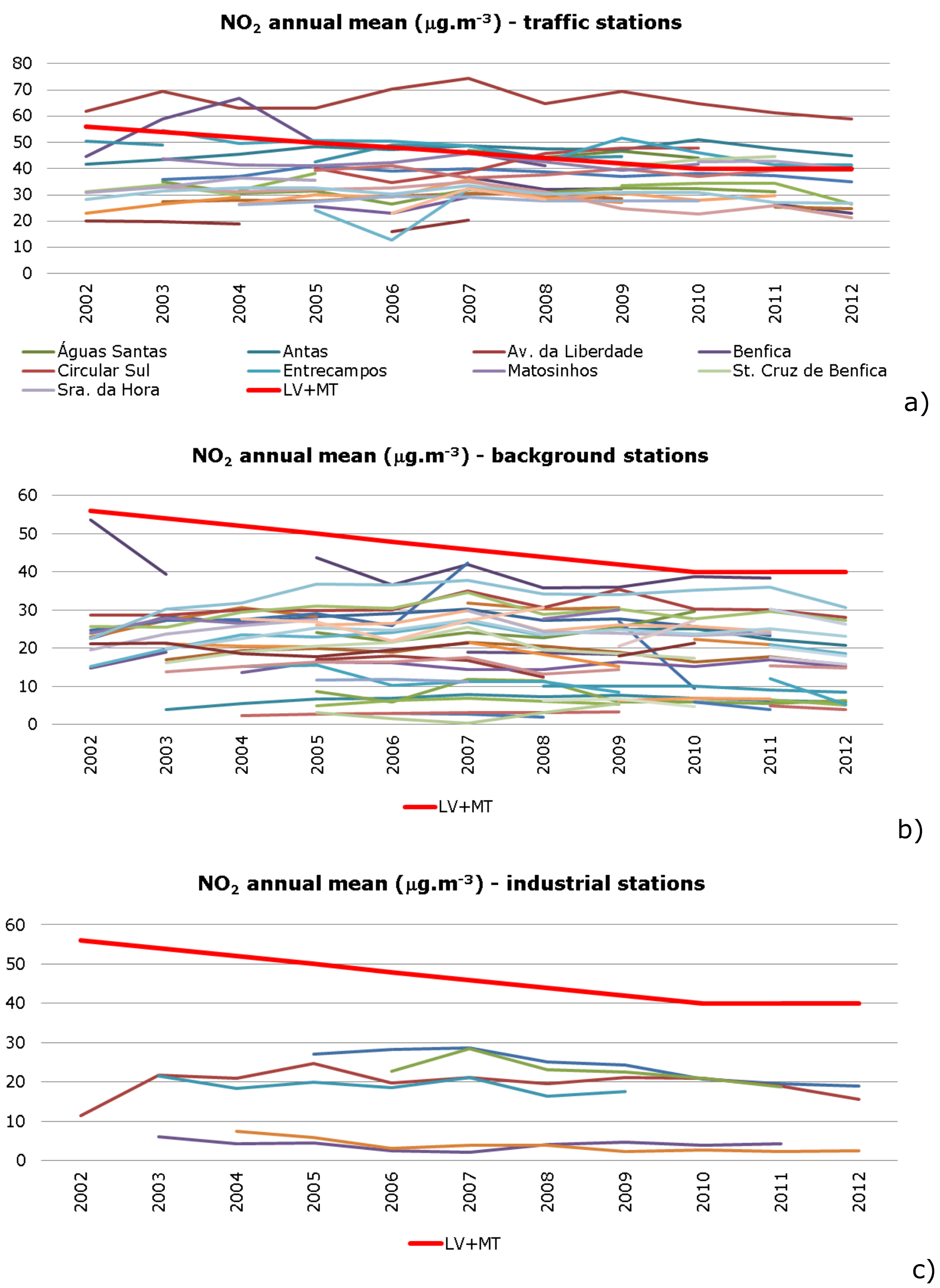


Figure 2.11 – NO$_2$ annual mean concentration values in Mainland Portugal for traffic (a), background (b) and industrial (c) stations, comparing with LV (2002-2012).

According to Figure 2.11, the traffic stations registered exceedances to the NO$_2$ annual LV, namely Águas Santas (ASNT), Antas (ANT), Circular Sul (CSUL), Matosinhos (MAT), Senhora da Hora (SHOR) (all five in the Northern region), Entrecampos (ENT), AVL and Benfica (BEN) (all three in the Lisboa and Vale do Tejo region). For this reason, this pollutant is considered as a critical air pollutant and as required by the legislation AQPs are needed for these two regions (Borrego et al., 2012a).

PM10:

In terms of particulate matter, the analysis was performed for PM10 and PM2.5. Figure 2.12 shows the number of exceedances to the daily LV for PM10 in each air quality station (2002-2012).

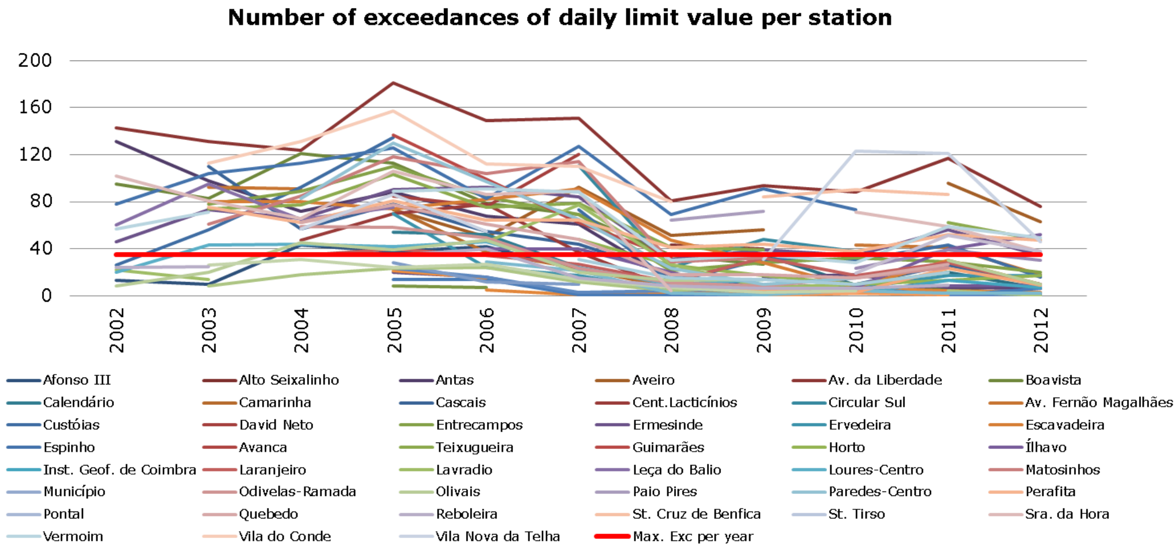
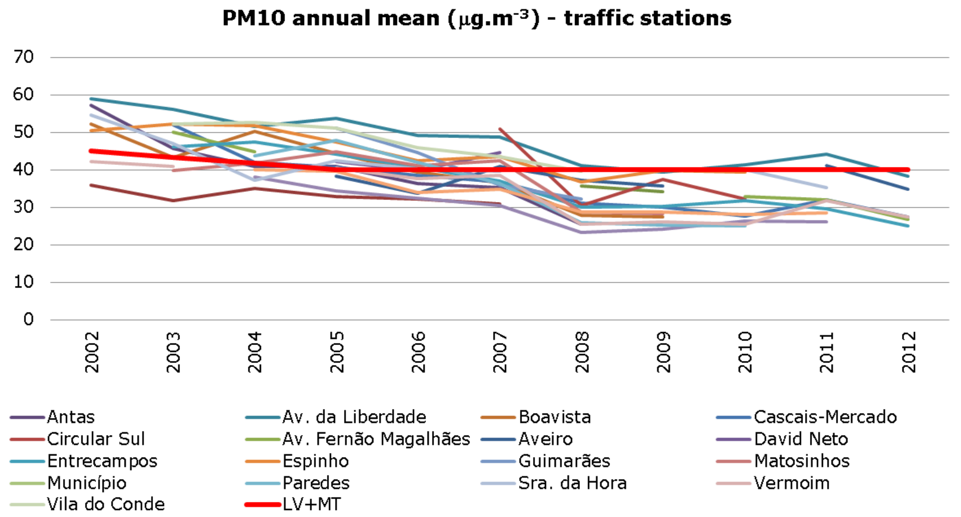


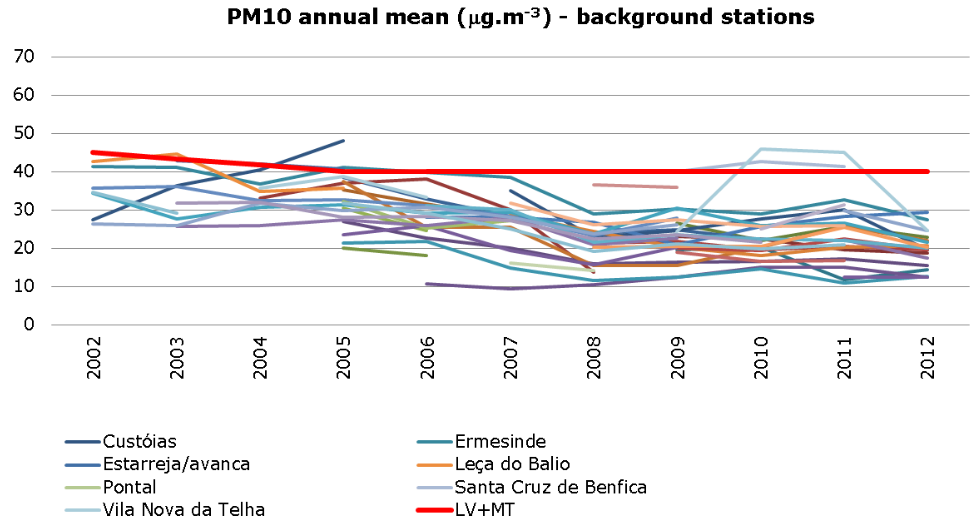
Figure 2.12 - Number of exceedances to the daily LV for PM10 (2002-2012).

Since 2002 there were several air quality stations that surpassed the maximum number of exceedances to the daily LV (35 times, as defined in the legislation). The exceedances were reduced in the last three years, however, AQPs are needed for the affected regions, namely Northern Region, Centre Region and Lisboa and Vale do Tejo region.

A comparison between the annual LV for PM10, defined on the DL 102/2010 of 23 September 2010, and the annual mean calculated from the air quality data measured in the air quality stations was performed for each year of the study period. The analysis was made by type of influence of each air quality station (Figure 2.13).



a)



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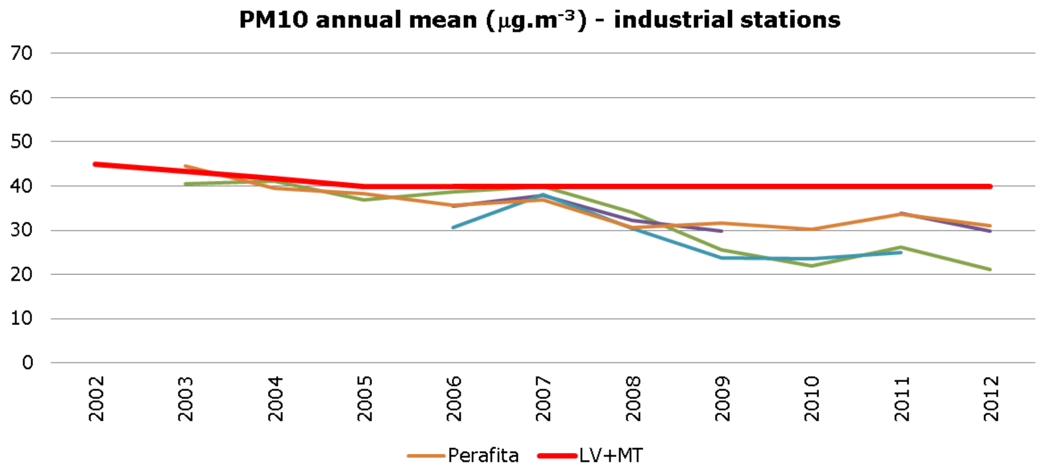


c)

Figure 2.13 – PM10 annual mean concentration values in Mainland Portugal for traffic stations (a), background stations (b) and industrial stations (c), against the LV from 2002 to 2012.

The traffic stations show higher number of air quality stations surpassing the LV+MT, but there are also some background stations surpassing the LV. In terms of industrial stations, just Perafita (PER) surpassed in 2003 the LV+MT, but the observed data was very close to the limit. In general, for all the analyzed period there were air quality stations that surpassed the annual LV even with the MT. From these two analyses it is possible to conclude that PM10 is a critical pollutant in Portugal and AQPs are needed.

PM2.5:

For PM2.5, the annual LV (25 μg.m^{-3}) was only defined on the DL nr 102/2010 of 23 September 2010, and there is a reduce number of air quality stations that have appropriate equipment for the measurements. Figure 2.14 shows PM2.5 annual mean for each air quality station and the LV for each year.

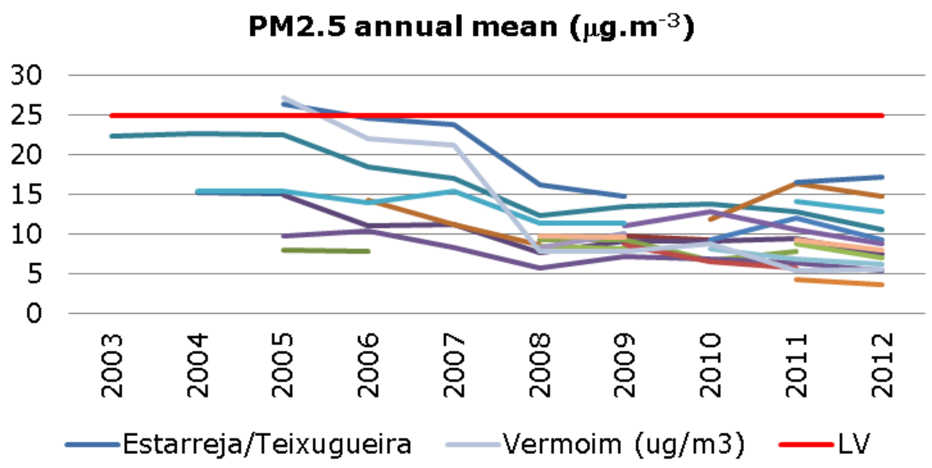


Figure 2.14 – P2.5 annual mean concentration values and LV (2004-2012).

Only in 2005 the annual mean surpassed the annual LV in Estarreja/Teixugueira (TEI) and Vermoim (VER) air quality stations. Since the LV was defined in 2008, PM2.5 is not considered as a critical pollutant.

O_3:

The O_3 air quality data was analysed taking into account the legislated values regarding human health protection. Figure 2.15 shows the Target Value (TV) exceedances registered at the air quality network during the study period.

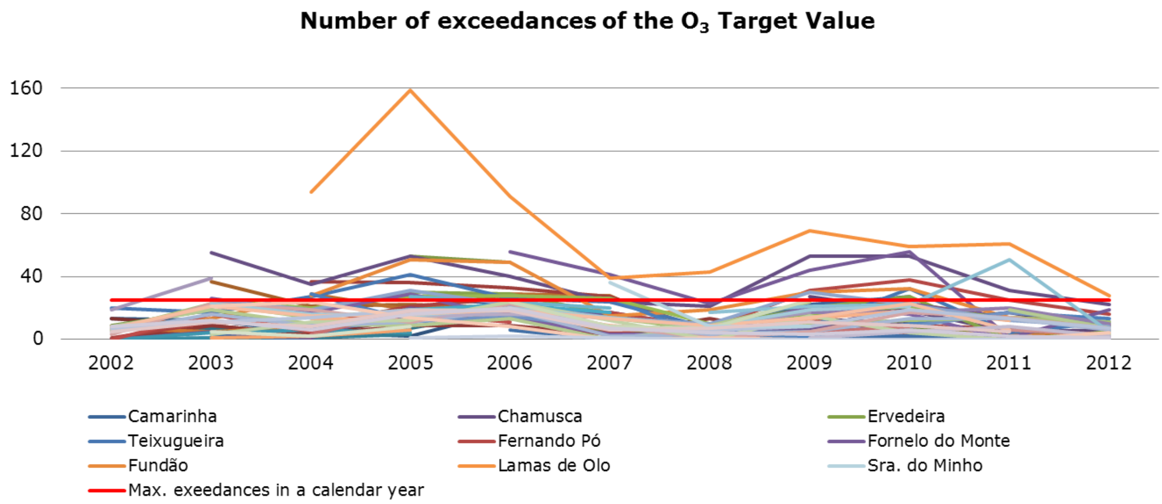
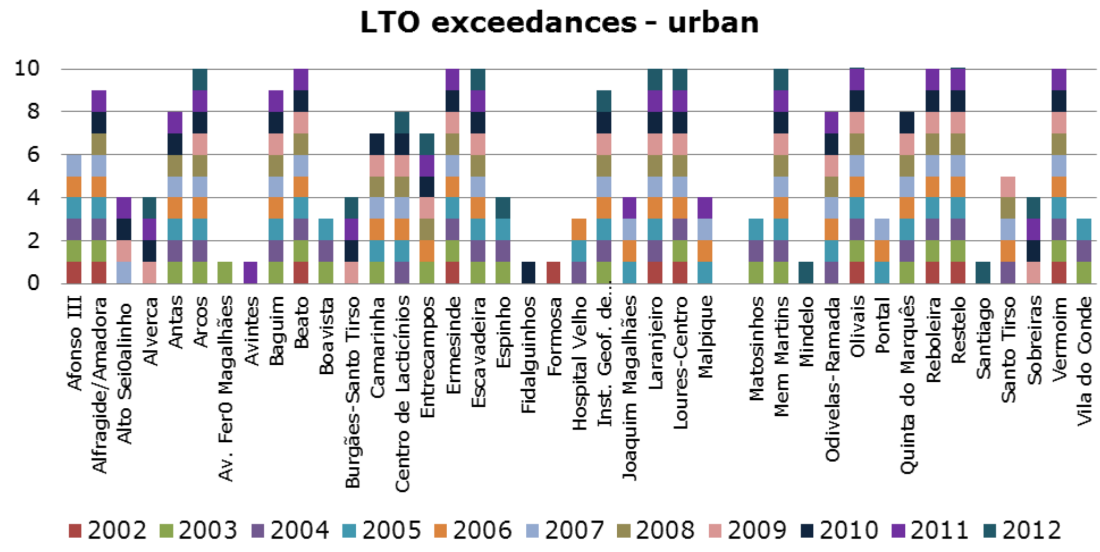


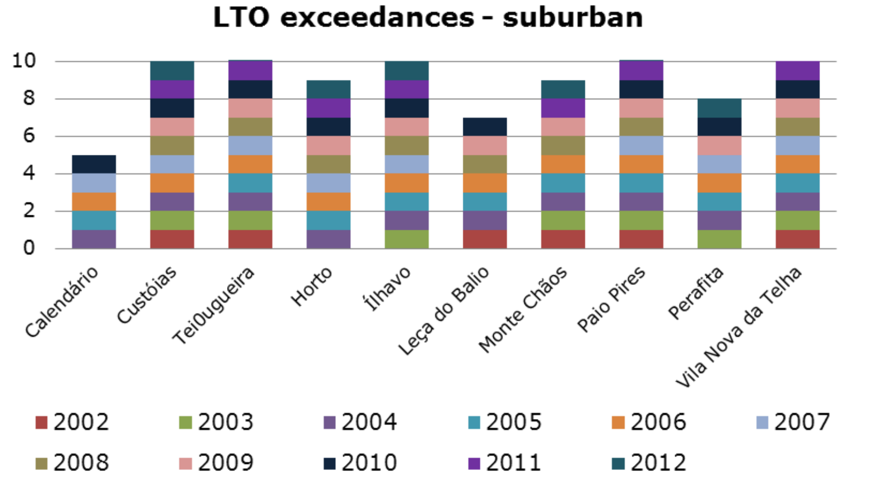
Figure 2.15 - Number of exceedances of the TV for O_3 (2002-2012).

Since 2003, several air quality stations surpassed the TV for O_3 more than 25 times in a calendar year. However the TV is only considered for the air quality assessment in 2010. The number of air quality stations that are surpassing the TV is decreasing from 2010 to 2012. In 2012 only the two rural air quality stations of the Northern region are in incompliance.

Figure 2.16 represents the air quality stations that surpassed the LTO value for the period 2002-2012 by type of environment.



a)



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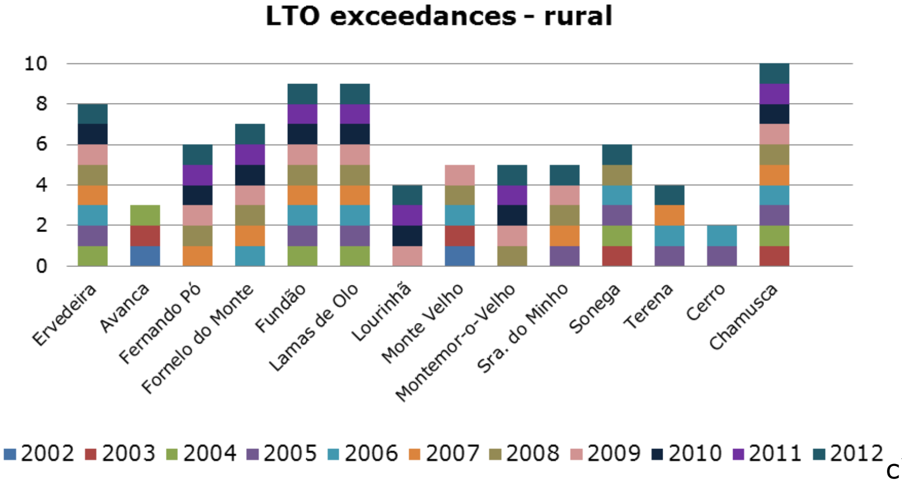


c)

Figure 2.16 – LTO exceedances of O_3 for different air quality stations concerning: urban (a), suburban (b) and rural (c) environments from 2002 to 2012.

Exceedances to LTO were registered in almost all air quality stations between 2002 and 2012. In all types of air quality stations there are several air quality stations that show exceedances for the majority of the analyzed period. For this reason it will be essential the implementation of measures to reduce the emissions of O_3 precursors in Portugal.

Regarding the O_3 thresholds, the number of exceedances regarding the information level is depicted in Figure 2.17 and the alert level in Figure 2.18.

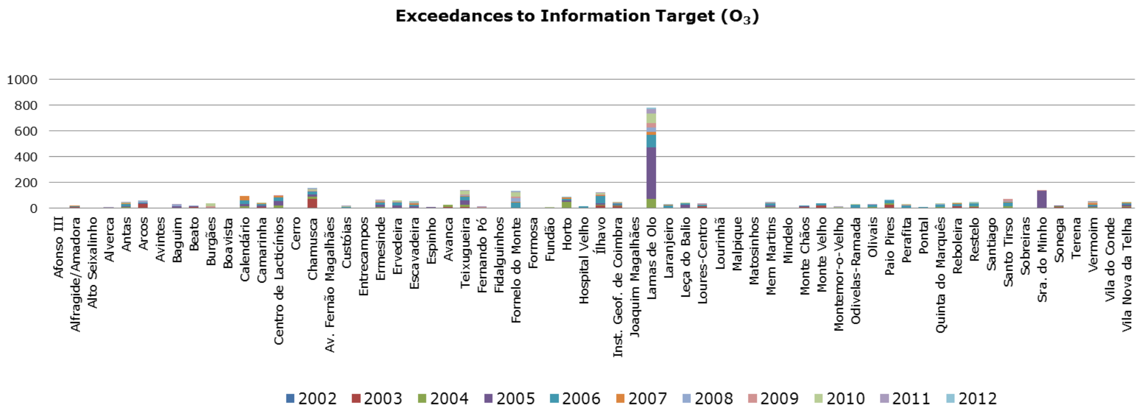


Figure 2.17 - Number of exceedances regarding the O_3 information threshold for the period 2002-2012.

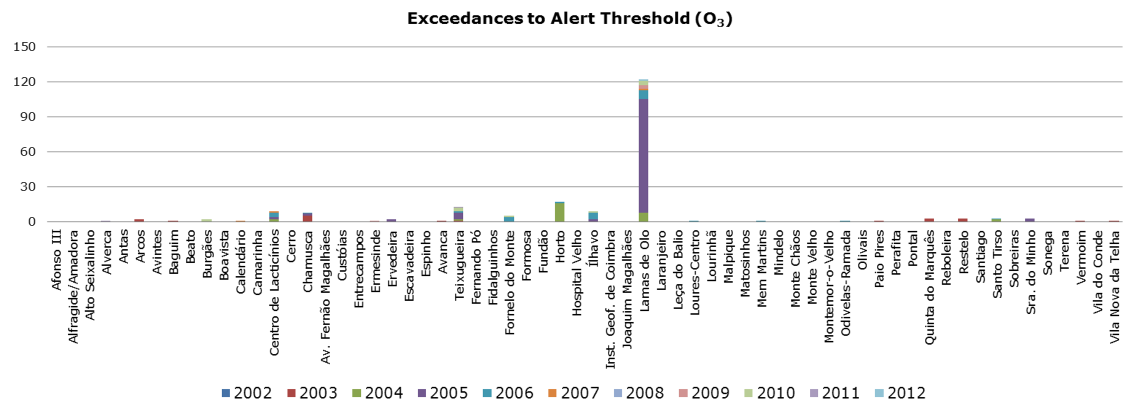


Figure 2.18 - Number of exceedances regarding the O_3 alert threshold for the period 2002-2012.

In the period 2002-2012, the information and alert thresholds are frequently surpassed in Mainland Portugal, namely on the rural air quality stations, but also in some traffic stations. In detail, the year 2005, registered the highest hourly O_3 concentration measured in Europe, on the Lamas de Olo (OLO) air quality station. In this sense, O_3 is considered as a critical pollutant and AQPs should be designed and implemented.

2.3.2 Air quality Plans

As mentioned before, the Directive 2008/50/EC defined as one of the main MS obligations, the implementation of AQPs when the air pollutant concentrations exceed the established air quality standards. From the analysis performed to the air quality data of the national air quality network, NO_2, PM10 and O_3 were identified as the pollutants for each should be AQPs implemented. At this time, there are only

AQPs for NO_2 and PM10 for the Northern and Lisboa and Vale do Tejo regions (Portaria n.º 716/2008, Portaria n.º 715/2008). The Centre region intends to implement an AQP for PM10. Also, for O_3 there is an intention to start the implementation of an AQP briefly.

In order to better know the AQPs already implemented in Portugal, an example of the Northern region AQPs for NO_2 and PM10 is described below.
The air quality plans are divided in four actions:

1. Identification of the air quality stations in incompliance;
2. Estimation of emission sources contribution for the air pollutant registered levels;
3. Definition of the adequate emission reduction measures;
4. Evaluation of the air quality impacts of the selected measures.

On the AQP for NO_2, there were several air quality stations that exceeded the annual threshold value after 2006. Table 2.2 summarises the NO_2 monitoring stations that exceeded LV + MT, the annual measured value, and their data collection efficiency.

Table 2.2 - Air quality stations that exceeded the LV+MT of NO_2 for the reference period of one year, their type and data collection efficiency.

Air quality station	Type of station	Year of exceedance	LV+MT	Annual average	Efficiency (%)
Boavista	Traffic	2006	48	49	99
Boavista	Traffic	2007	46	49	100
Águas Santas	Traffic	2007	46	47	97
Antas	Traffic	2007	46	49	92
Circular Sul	Traffic	2008	44	46	93
Antas	Traffic	2008	44	47	95
Boavista	Traffic	2009	42	45	96
Águas Santas	Traffic	2009	42	47	91
Circular Sul	Traffic	2009	42	48	93
Antas	Traffic	2009	42	47	99
S. Hora	Traffic	2010	40	43	94
Circular Sul	Traffic	2010	40	48	95
Antas	Traffic	2010	40	51	99
Matosinhos	Traffic	2010	40	42	89
Águas Santas	Traffic	2010	40	44	86

As observed in Table 2.2, all the air quality stations that do not meet the NO_2 annual LV + MT are classified as urban traffic stations and are located in the Porto Litoral (Boavista, Antas, Águas Santas, Senhora da Hora, and Matosinhos) and

Braga (Circular Sul) agglomerations. These registered exceedances indicate that the traffic sector could be an important emission source. In order to quantify the traffic contribution, the regional and urban background values were also estimated using a detailed analysis of the NO_2 concentrations measured at different monitoring sites. Air quality data registered between 2006 and 2010 at two different rural stations, 11 suburban/urban sites, and 12 traffic stations were used in this analysis.

To estimate the regional NO_2 background, annual averages were calculated using the rural background stations located in the northern region (Senhora do Minho and Lamas d'Olo stations). Notwithstanding this reduced number of stations, the calculated annual average values can be considered to be representative of the regional background over the northern region of Portugal. Their spatial location and distribution were selected according to the criteria of the EU Directive. The same calculation of annual averages was performed for the urban background stations and the traffic stations. The results are presented in Figure 2.19.

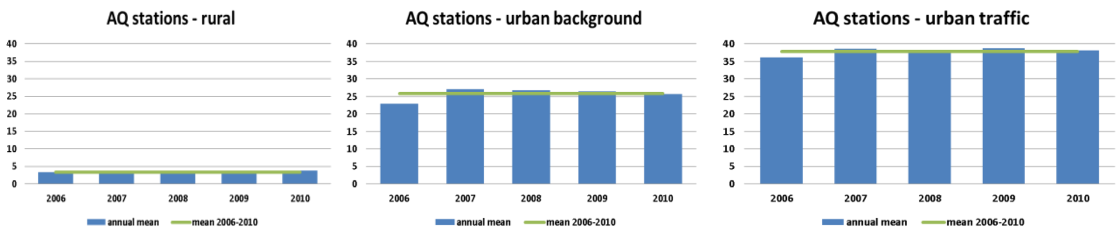


Figure 2.19 - NO_2 annual averages monitored between 2006 and 2010 at rural background, urban background and urban traffic stations placed over the Northern region of Portugal.

The difference between the rural and urban background values provides a quantification of the urban contribution, which is characterised by the influence of anthropogenic activities, such as domestic combustion and traffic.

The difference between the background and urban traffic environments corresponds to the traffic average contribution over the Porto urban area.

Figure 2.20 shows the contribution in percentage of background sources, other sources (such as residential combustion) and traffic, on the concentrations values registered in the air quality stations of the Northern Region network.

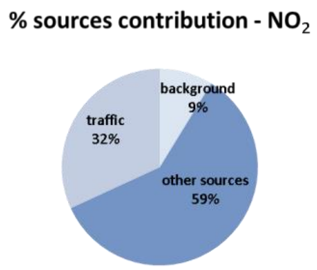


Figure 2.20 - Sources contribution (%) on NO_2 the concentrations values registered in the air quality stations of the Northern Region network.

The traffic contribution has similar weights (percentages of approximately 30%) (WHO, 2005; Lutz, 2011).

These results indicate that the estimated traffic contribution is responsible for the NO_2 annual average exceedances at those specific traffic sites. This kind of information is relevant to the decision makers to select and implement the most appropriate measures to reduce the NO_2 concentration values. Air quality management strategies and measures should thus be focused on this specific sector.

In this sense the selected measures to reduce the emissions of NOx were for the traffic sector. Some of the measures to the traffic sector are described on Table 2.3.

Table 2.3 – Examples of the NO_2 reduction measures selected for the Northern Region (Borrego et al., 2012a).

Sector	**Measure**
Traffic	Introduction of low emission fleet in public transportation and improvement of public transport network
	Renewal of taxis and solid waste collection vehicles
	Decrease of heavy vehicles circulation in the urban areas

In order to estimate the NO_2 air quality impacts of the selected measures, the air quality TAPM was applied. The implementation of all the selected measures in the northern region will lead to a percentage reduction of between 20% and 50 %, which corresponds to a reduction of 4-5 $\mu g.m^{-3}$ in the annual average NO_2 values. Also, will produce a reduction in the NO_2 annual mean concentration values for all the air quality stations that did not meet the legislated limits in 2010, however there are some air quality stations that will not fulfil the air quality requirements.

In terms of PM10, since 2001 the legislation requirements have been surpassed and the first AQP was performed in 2004 (Borrego et al, 2012b). Figure 2.21 shows the number of exceedances in the Northern region from 2001 to 2004.

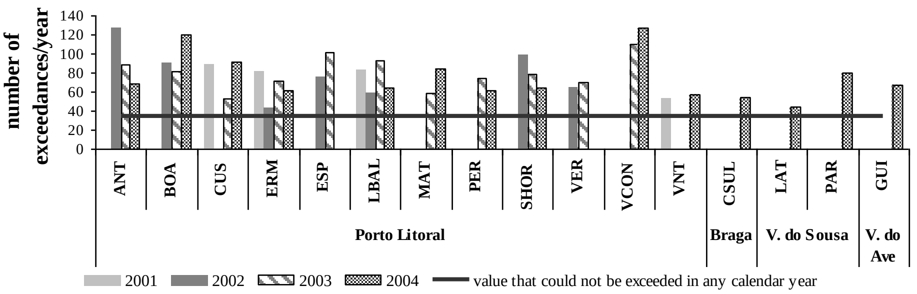


Figure 2.21 - Number of exceedances in Porto Litoral, Vale do Ave, Vale do Sousa and Braga agglomerations to 2001-2004 (Borrego et al., 2012b).

A study was performed to investigate the causes of the high values of PM for the period 2001-2004 (Borrego et al., 2008) showing an increase of the anthropogenic contribution to the exceedances through the years.

The different type of air quality stations with exceedances (Figure 2.21) indicates that is not so evident the sector with higher contribution. Also, in Borrego et al., 2010 it was identified the several sectors that contribute to PM10 emissions (Figure 2.22).

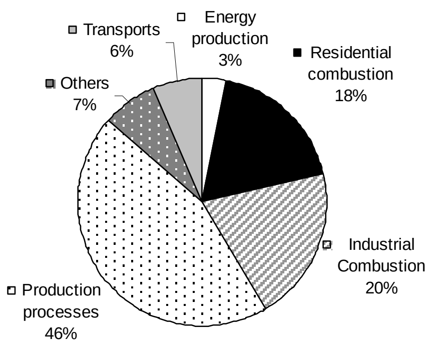


Figure 2.22 – PM10 emissions contribution by sector (Borrego et al., 2010).

In this sense, the measures selected to improve air quality in terms of PM10 had a wider range of sectors: traffic, industry, residential combustion, constructions, agriculture and forests (Borrego et al., 2012b). Some examples of the measures to the first three sectors are described in Table 2.4.

Table 2.4 – Examples of the PM10 reduction measures selected for the Northern Region (Borrego et al., 2012b).

Sector	Measure
Traffic	Introduction of low emission fleet in public transportation and improvement of public transport network
	Renewal of taxis and solid waste collection vehicles
	Decrease of heavy vehicles circulation in the urban areas
	Low Emission Zones
	Banning traffic from selected streets
Industry	Improvement of industrial PM retention systems
	Reinforcement of the inspection of industry sources
	Establishment of emissions standards for industrial clusters and business activities in urban areas
Residential combustion	Use of certified combustion appliances with PM emissions reduction

In order to investigate the impact of the designed PM10 reduction measures on the air quality of the Northern Portugal, TAPM was applied over the northern region. The maximum absolute reduction is achieved over the Porto region and reaches 4.6 $\mu g.m^{-3}$ that corresponds to a 19% reduction of the PM10 average levels in the atmosphere. Although the PM10 annual mean and the number of exceedances decrease in the selected air quality stations, the legislated limit values continue to be surpassed.

2.4 Summary and Conclusions

This chapter analysed all the available monitoring data for CO, SO_2, VOCs, NO_2, PM and O_3 measured in Portuguese air quality network for the period 2002-2012, regarding the legislation fulfilment in terms of protection of human health.

The concentrations of CO, SO_2, VOCs and PM2.5 for the study period were all substantial lower than the LV established and for this reason it was concluded that those pollutants were not considered as critical pollutants in terms of air quality assessment. In the case of CO and SO_2 there were some isolated episodes with exceedances, but they were investigated and causes were identified.

Three of the pollutants analyzed were considered critical in terms of air quality: NO_2, O_3 and PM10. The first one, presented exceedances to the hourly LV, in some air quality stations in 2006, 2009 and 2010; and also, since 2002, there were exceedances to the annual LV in traffic stations, over Northern and Lisboa and Vale do Tejo regions. In terms of O_3, the TV has been surpassed more than 25 times in a calendar year, namely in 2010, also, there were registered exceedances to the LTO in almost all air quality stations since 2002 and information and alert thresholds are surpassed frequently, namely on the rural air quality stations. For PM10, since 2002 there were air quality stations that surpassed the maximum number of exceedances to the daily LV (35 times). The annual LV for PM10 was surpassed in all the years, with a significantly reduction of the number of stations in incompliance since 2008, probably due to the financial crisis that Portugal is facing over the last four years (Borrego et al., 2012c).

Air quality plans already have been developed for NO_2 and PM10, and new AQP for O_3 is being prepared. From the two AQPs presented in this chapter it is possible to verify that in terms of sources contribution, for NO_2, was evident the weight of traffic (around 32%). For PM10, several sectors have contributed to the concentration levels registered in the air quality stations. Based on this information the most appropriate measures to reduce the concentration values of these pollutants were selected. For NO_2 the measures affected mostly the traffic sector, on the other hand for PM10 the measures involved more sectors. Some measures are already implemented for PM10 and are defined for NO_2, but it will be necessary to define and to implement new measures for the O_3 precursors.

The air quality impacts of the implemented measures in NO_2 and PM10 revealed a decrease in the concentration levels of both pollutants, however, even with this reduction, not all the air quality stations fulfill the air quality legislation requirements.

Chapter 3

3 Meteorological driven changes on air quality over Portugal

The contents of this chapter will be published in:

Sá, E., *Tchepel, O., Carvalho, A., Borrego, C. (submitted). Meteorological driven changes on air quality over Portugal: a KZ filter application. Atmospheric Pollution Research.*

Abstract

The Kolmogorov-Zurbenko (KZ) filter method is a relevant tool to improve air quality management, since it determines meteorological effects on air quality concentrations and separate out those effects in order to examine underlying trends. Air quality in Portugal is exceeding the legislated particulate matter (PM10), nitrogen oxides (NO_2) and ozone (O_3) levels; accordingly, measures to reduce pollutant emissions have been designed. The primary objective of this work is to investigate the influence of meteorology on O_3, NO_2 and PM10 levels and the long-term air quality trends. Air quality and meteorological datasets were explored for the period 2002-2012 through the decomposition of time-series using the Kolmogorov-Zurbenko (KZ) filter. To determine the best meteorological predictors for the air quality data, a stepwise regression analysis of the filtered time-series was applied. The KZ filter application revealed that the short-term component has the highest contribution to the total variance of the original air quality data (≈64 % - PM10; ≈52 % - O_3; ≈54 % - NO_2) followed by the seasonal component. The long-term component exhibits the influence of the emission control regulations implemented in each study region. The statistical analysis of the air quality and the meteorological data indicated that O_3 has a statistically significant relationship with temperature in most of the components. The results also indicate that emission control strategies are primary regulators for NO_2 and PM10 levels. Therefore, to establish an accurate strategy to improve air quality further, it will be essential to include meteorological effects. This study highlights that the KZ filter is a useful tool to support the design and implementation of adequate air quality strategies.

3.1 Introduction

The importance of air pollution has increased over the last decade as an important social, economic and environmental issue because of the phenomenon of diminishing air quality and the associated serious human health effects (Fenger, 2009). The European Union (EU) strategy reflects this concern by designing and implementing the guidelines for successful air quality management. To reduce air pollution and control the effects on human health and the environment, air quality directives require EU member states to assess and manage air quality over their territories.

Generally, air pollution results from the combination of high emissions and unfavourable weather conditions (Jacob and Winner, 2009). Meteorology plays an important role in air pollution formation, dispersion, transport and dilution. Therefore, the variations in local meteorological conditions, e.g., wind speed and direction, temperature and relative humidity, can affect the temporal and spatial variations of air quality pollutants (Duenas et al., 2002; Satsangi et al., 2004; Elminir, 2005). Usually, the main measures for air pollution control are implemented through emission controls (Fenger, 2009). However, to confirm the effectiveness of air quality regulations and improve air quality management efforts, long-term air quality trends must be analysed and explored. Moreover, the strong linkage between weather conditions and pollutant levels can obscure the effects of changing emission levels over time; a better understanding of the effects of meteorology on air quality is important (Flaum et al., 1996).

The statistical approaches to air quality adjustment for meteorology may be aggregated into three groups, with considerable variety within each: regression-based modelling, extreme value approaches and space-time models (Thompson et al., 1999). Rao et al., 1997 argued that process changes due to policy or climate changes may be very small and difficult to detect in time series unless they are separated from weather and seasonality.

To understand the contribution of underlying physical processes and the influence of emission sources on the variability of air pollutant concentrations, the different process scales represented in the air quality and meteorological time series should be separated. One of the methodologies that could be applied for this purpose is based on the Kolmogorov-Zurbenko (KZ) filter proposed by Rao and Zurbenko (Rao and Zurbenko, 1994). This method implements low-pass filtering of time-series through repeated iterations of a moving average. In Eskridge et al., 1997, the KZ filter was compared with anomalies and PEST methods; the results demonstrated that the KZ filter had the same level of accuracy as the wavelet transform method and, unlike other methods, could be directly applied to datasets containing missing observations and is very easy to implement.

The KZ filter method is usually applied for situations in which changes in air pollutant concentrations related to emission reductions were much smaller than changes related to meteorological conditions (Wise and Comrie, 2005). Several studies have applied the KZ filter to assess O_3 trends in the United States (Rao and Zurbenko, 1994; Rao et al., 1995; Milanchus et al., 1998; Yang and Miller, 2002; Wise and Comrie, 2005), Australia (Anh et al., 1997) and Spain (Ibarra-Berastegi et al., 2001). All of these studies assessed only O_3 except for Anh et al. 1997 and Wise and Comrie, 2005, which also assessed NO_2 and PM10, respectively.

Ozone concentrations are strongly linked to meteorological conditions, being positively correlated with high temperatures, solar radiation and light winds. In Europe, the highest O_3 concentrations occur in summer under stable high-pressure systems with clear skies (Duenas et al., 2002). Most previous studies have concluded that PM10 is not as weather-dependent as O_3. However, wind speed, mixing height, and relative humidity are the meteorological variables believed to mostly influence PM10 concentrations. Stagnant conditions are thought to correlate with high PM10 concentrations because they allow particulates to accumulate near the earth’s surface. Although high wind speeds can increase ventilation, they are normally correlated with high PM10 concentrations because they allow the resuspension of particles from the ground and long-range transport of particulates between regions. High PM10 concentrations are normally associated with dry conditions due to the increased potential for the resuspension of dust, soil, and other particles. In the Southwestern United States, the moisture level, namely the relative humidity, is the strongest predictor of PM10 concentrations (Wise and Comrie, 2005).

Tchepel and Borrego, 2010 explored the underlying physical processes and the effects of emission sources on the variability of the air pollutant concentrations for traffic related pollutants (CO and PM10) in Portugal. They applied Fourier series analysis to explore the time series of meteorological and air quality data (Ghil et al., 2002). Moreover, to better understand the model prediction uncertainty related to short-term variations in pollutant concentrations, a methodology using spectral analysis and the KZ filter was used to estimate uncertainties of the results obtained with the CHIMERE model for Portugal (Tchepel et al., 2008). However, no studies have been performed with the KZ filter to assess the long-term air quality trends and the influence of meteorological variables on the measured concentrations over Portugal.

In Portugal, the pollutant concentrations registered on the national air quality network indicate that the critical pollutants are particulate matter with a 50 % efficiency cut-off at an aerodynamic diameter of 10 µm (PM10), nitrogen dioxide (NO_2) and ozone (O_3) in some agglomerations. Over the last few years, NO_2 and PM10 levels surpassed the air pollution control legislated values and air quality plans have been designed to guarantee air quality improvements (Borrego et al., 2012a, 2012b). The air quality requirements for O_3 started to being considered in air quality management in 2010, following the recommendations from the decree-law nr 102/2010 of 23 September 2010. Since 2010, that O_3 is surpassing the legislation requirements, therefore, a new air quality plan for O_3 is being prepared in Portugal. However, considering the higher registered levels of O_3 that surpassed the legislated values, air quality plans need to be implemented in the near future. In particular, this situation is verified in the agglomerations that included the two main urban areas of Portugal: Lisbon and Porto. In terms of air quality management and health effects, these areas have great relevance due to the high number of inhabitants exposed to air pollution.

This study implements a methodology to quantify the contribution of different processes to air pollution fluctuations in Portugal in two cities dealing with air quality problems over the last decade. In this sense, a KZ filter was applied to separate time-series into its temporal components and to allow the investigation of air quality trends. Then, in order to understand the relation between meteorology variables and air quality a stepwise regression was applied to the KZ filter outputs. Moreover, this study provides essential information relevant for air quality management in these two cities. It is important to assess the effects of weather conditions on air quality. Accurate planning of measures to improve air quality is strongly dependent on this information.

3.2 Data and Methods

3.2.1 Methodology

In this study, to analyse the air quality trends and to explore the influence of meteorological data on the air quality data, the KZ filter method was applied to separate the original time series into different scales. Thereafter, a statistical analysis was performed to the filtered data.

The KZ filter method was used to separate each variable into its temporal components. The air quality and meteorological datasets were clearly separated by appropriate filtering techniques into short-term, seasonal and long-term variations. According to Rao and Zurbenko (Rao and Zurbenko, 1994), time series data can be represented by

$$A(t) = e(t) + S(t) + W(t), \qquad \text{(Eq. 3.1)}$$

where*A(t)* is the original time series and *e(t)*, *S(t)* and *W(t)* are the long-term, seasonal and short-term components, respectively. In terms of the air quality components, the short-term component is attributable to weather and short-term fluctuations in precursor emissions, the seasonal component is a result of changes in the solar angle, and the long-term trend results from changes in overall emissions, pollutant transport, climate, policy, and/or economics.

The KZ filter is a low-pass filter produced through repeated interactions of a moving average, i.e., KZ(*m, p*), a filter with a window length *m* and *p* iterations. Adjusting the window length and the number of iterations makes it possible to control the filtering of different scales of motion. To filter periods of less than *N* days, the following criterion is applied to determine the filter's effective width (Wise and Comrie, 2005):

$$m \times p^{1/2} \leq N. \qquad \text{(Eq. 3.2)}$$

The applied filter lengths are explained bellowconsidering both the effective widths and cut-off periods that already have been used in previous KZ filter studies (Eskridge et al., 1997; Ibarra-Berastegi et al., 2001; Wise and Comrie, 2005).

A $KZ_{(15,5)}$ filter (15-day length with 5 interactions) was applied to the meteorological and air quality data to extract the sum of the long-term and seasonal components:

$$KZ_{(15,5)} = e(t) + S(t). \qquad \text{(Eq. 3.3)}$$

The residuals of the filter $(A\,(t) - KZ_{(15,5)})$ contain high frequency processes that are referred to as short-term variations, i.e., the short-term component.

The long-term trend from the data can be obtained by the application of a KZ filter choosing a larger window size, e.g., $KZ_{(365,3)}$. The result is the long-term component of the original air quality time series:

$$e(t) = KZ_{(365,3)}. \qquad \text{(Eq.3.4)}$$

According to the combination of equation 3 and equation 4, the seasonal component of the data is obtained by subtracting the long-term component from the baseline components:

$$S(t) = KZ_{(15,5)} - KZ_{(365,3)}. \qquad \text{(Eq. 3.5)}$$

After generating the time-series of each temporal component through the application of the KZ filter to the air quality and meteorological datasets, a statistical analysis was applied to explore the relationships between air quality and meteorological variables. Because we are interested in considering several meteorological variables at the same time in this study, stepwise regression was implemented using *SPSS predictive analytics software* version 19, producing a set of statistical models for O_3, PM10 and NO_2. The stepwise regression essentially does multiple regressions a number of times, each time removing the weakest correlated variable, obtaining at the end the variables that explain the distribution best. The statistical models are a combination of the meteorological variables for each temporal component, regressed with the respective temporal component of each air pollutant. The meteorological variables used in this study were the maximum daily values of temperature (T), dew point temperature (Td) and wind components: east-west component (U) and north-south component (V). These variables were then introduced in the stepwise regression and the terms were accepted only if they met the 0.05 significance level.

3.2.2 Air quality and meteorological database

In Portugal, there are two critical urban agglomerations with no compliance with the air quality requirements because they each have several air quality monitoring stations registering PM10 and NO_2 levels that surpass the legislated limits: Area Metropolitana de Lisboa Norte (AMLN) and the Porto Litoral (PL) (Figure 3.1). Because these regions contain the most relevant national urban areas, i.e., Lisboa and Porto, they have the highest number of inhabitants, which enlarges the importance of air quality management.

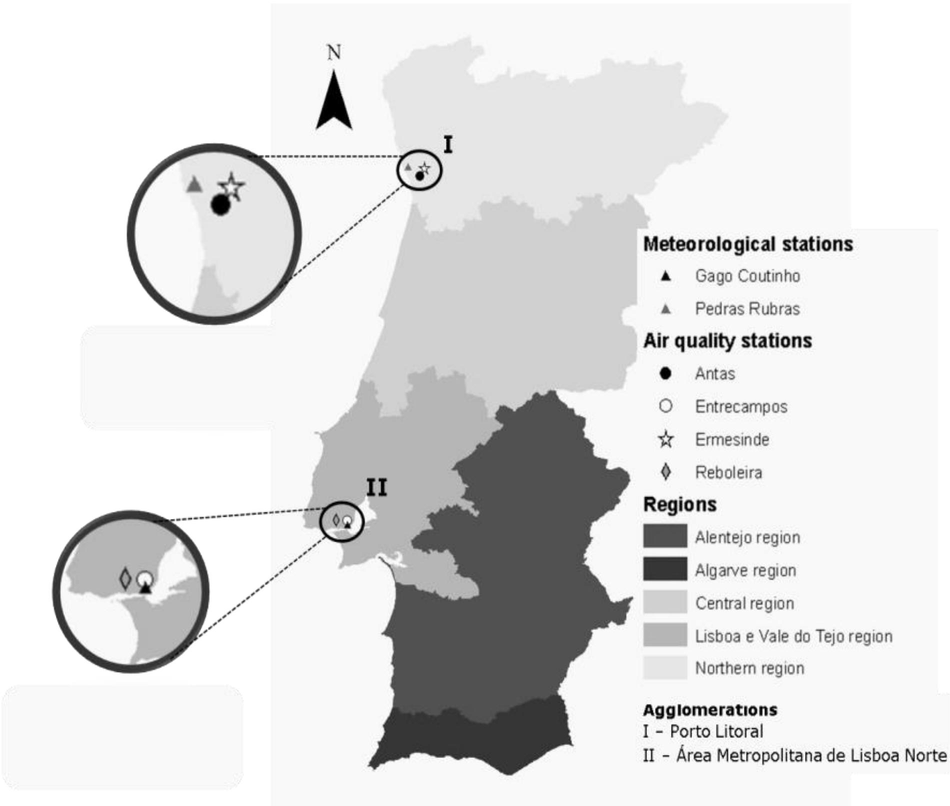


Figure 3.1 - Identification of the study regions and location of the meteorological, air quality stations.

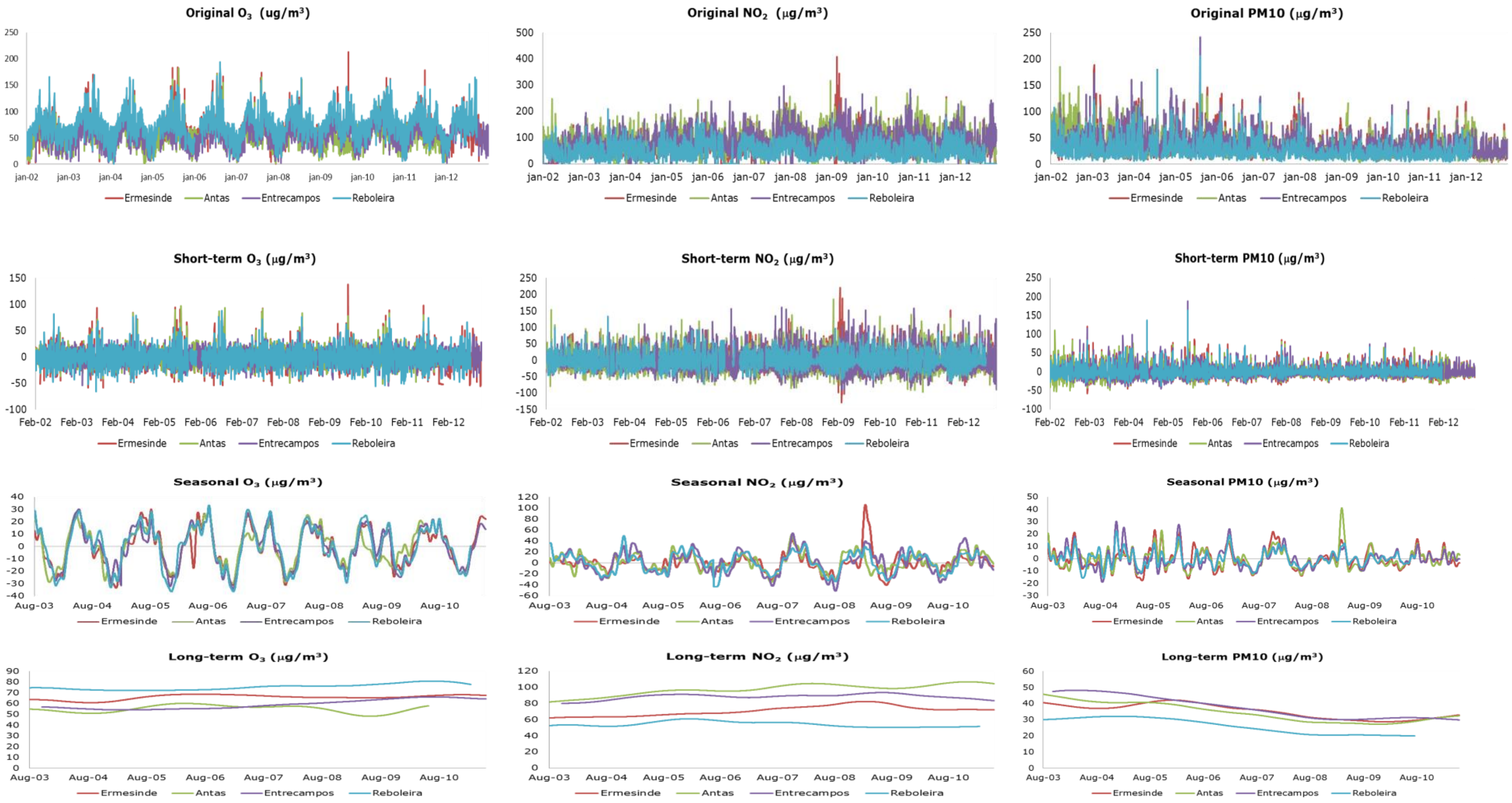
Two air quality monitoring stations were selected in each agglomeration based on the type of influence and the availability of data. The percent of minimum data captured was 85%, considering that the Directive 2008/50/EC defined the minimum to be 90% without regular calibration or normal maintenance of the instrumentation (corresponding to a 5% loss of data) for all the pollutants except for ozone, which has a minimum of 75% as established in the legislation. In the PL agglomeration, the Ermesinde (urban background) and Antas (urban traffic) air quality stations were selected. For the AMLN agglomeration, the Reboleira (urban background) and Entrecampos (urban traffic) air quality stations were selected (Figure 3.1). Hourly data of O_3, PM10 and NO_2 concentrations for the period 2002-2012 were collected from these four air quality stations. Moreover, the daily averaged PM10 concentrations, daily maximum NO_2 concentrations and the daily maximum of 8hr moving average O_3 concentrations for each day were derived from the original datasets.

The meteorological data were obtained from the National Oceanic and Atmospheric Administration (NOAA) for two meteorological stations: Pedras Rubras, located in PL and, Gago Coutinho, located in AMLN (Figure 3.1). These two meteorological stations are the closest to the air quality stations used in this study. The analysed meteorological variables were temperature, dew point temperature and U and V wind components. Hourly data were collected for the period 2002-2012; a subset of meteorological data containing the daily maximum of each meteorological variable was estimated.

3.3 Results and Discussion

3.3.1 KZ filter analysis

The application of the $KZ_{(15,5)}$ filter allows the removal of the short-term signal, resulting from day-to-day weather variations. The long-term trend component was isolated using the $KZ_{(365,3)}$ filter. Subsequently, the seasonal component was estimated. The separation of the original air quality time-series into the three components is depicted in Figure 3.2 for the daily maximum of the 8-hr moving average O_3 concentration (a), daily maximum NO_2 (b) and 24-hr average PM10 (c).



a) b) c)

Figure 3.2 - Decomposition of a daily maximum of 8-hr moving average O_3 (a), daily maximum NO_2 (b) and 24-hr average PM10 (c) time-series by the KZ filter into short-term, seasonal and long-term components.

For all air quality stations selected in this study, the legislated target value for O_3 (120 $\mu g.m^{-3}$) is surpassed several times in the period 2002-2012 (Figure 3.2a – Original O_3). For NO_2, the original data represents the daily maximum for the period 2002-2012. Even though the legislation does not provide a limit for the daily maximum, it has limits for the hourly mean (200 $\mu g.m^{-3}$) and annual mean (40 $\mu g.m^{-3}$). Considering that the daily maximum oscillates between 0 and 400 $\mu g.m^{-3}$ (Figure 3.2b– original NO_2), it is possible to conclude that the legislated values are exceeded at all air quality stations, demonstrating that NO_2 is a critical pollutant in Portugal. The legislated values for PM10 suggest that the daily mean should not surpass 50 $\mu g.m^{-3}$ more than 35 times in a calendar year. By analysing the original data, it is possible to verify that throughout the period 2002-2012, there are several times when this limit is exceeded (Figure 3.2c– Original PM10).

For O_3, there is no significant variation in the long-term component throughout the study period. However, the changes are evident for the other two pollutants. The long-term trend results are related to changes in emissions, pollutant transport, climate, policy, and/or economics. In the case of PM10, the decrease of the long-term component concentration values is evident at all air quality stations since the end of 2005. This can be explained by the implementation of several measures regarding PM10 levels that were considered within the air quality plans for both urban areas. At the same time, measures for NO_2 were also implemented in Lisboa, which is also evident in the long-term component for this pollutant from the decrease in the concentrations at the Entrecampos and Reboleira air quality stations. In Porto, the air quality plans for NO_2 were only developed at the end of 2010. Therefore, the effects of the measures on the long-term components could not be ascertained at the Ermesinde and Antas air quality stations.

Only through the long-term component one is able to verify differences between the urban traffic air quality stations and the urban background air quality stations concentration values. In terms of O_3, higher concentrations are registered in the urban background air quality stations; lower NO_2 concentrations are also registered at these stations. This is expected because one of the major NO_2 emission sources is the traffic sector (Borrego et al., 2012a). For PM10, the urban background air quality station at AMLN (Reboleira) has lower concentration values than the other three air quality stations. In the PL agglomeration, there are no relevant differences between the two types of air quality stations, consistent with the fact that concentration values at all types of air quality stations in PL surpass the legislated limits (Borrego et al., 2012b).

To understand the contribution of each temporal component to the original air quality data, the variance of each generated time-series was calculated and the contribution to the total variance of the original data was determined (Table 3.1).

Table 3.1 - Contribution to the total variance of each component created by the KZ filter to the original air quality data.

		Contribution to the total variance (%)			
		Ermesinde	Antas	Entrecampos	Reboleira
O_3	Short-term	59	52	48	48
	Seasonal	41	42	49	46
	Long-term	1	2	3	1
NO_2	Short-term	70	69	64	59
	Seasonal	24	17	26	32
	Long-term	3	3	1	1
PM10	Short-term	68	56	65	66
	Seasonal	15	12	14	15
	Long-term	4	7	11	8

The results from Table 3.1 reveal that the contribution of the short-term component to the total variance is quite large, followed by the seasonal component, which demonstrates the importance of decomposing the datasets into temporal components to better extract information from the original data. The strong relationship between weather conditions and pollutant levels can obscure the effects of changing emission levels over time. Therefore, the meteorological signal must be analysed.

The contribution of the long-term component to the total variance in the original O_3 time-series is very small when compared with short-term and seasonal variations. In the case of the Porto urban area, the contribution of the short-term component to the total variance is 10 % higher than the seasonal component. However, in the Lisboa urban area, the contribution of the seasonal component to the total variance is similar to the short-term component.

The NO_2 short-term component is the largest contributor to the total variance of the original data, followed by the seasonal component. In this case, the seasonal component has a lower contribution to the total variance when compared to O_3. The long-term component continues to have lowest contribution to the total variance of the original data.

The PM10 seasonal component has the lowest contribution to the variance observed in the original data (approximately 14 %) compared with the other two pollutants. However, the long-term component has the highest contribution (approximately 7.5 %). For the other two pollutants, the seasonal contribution is approximately 2 %. The PM10 short-term component remains the one with the highest contribution to the total variance of the original data.

3.3.2 Statistical modelling

A stepwise regression procedure was applied to evaluate the influence of the meteorological variables on air quality, producing a set of statistical models for O_3, NO_2 and PM10 determined at a 95 % confidence level. This method was applied to each temporal component provided by the KZ filter method and for each air quality station. All the meteorological variables were used (temperature (ºC) – T; dew point temperature (ºC) – Td; U and V wind components ($m.s^{-1}$)). Table 3.2 presents the statistical model suggested by the stepwise regression and the corresponding explained variance (%).

Table 3.2 - Regression model selected by the stepwise regression for the air quality data concerning the short-term, seasonal and long-term components.

		Regression Model	Variance explained (%)	N	P value
		Short-term			
O_3	ANT	0.05+1.38T-0.70Td-0.39U	18%	3493	<0.01
	ERM	-0.92+1.80T-1.08Td-0.33U+0.12V	25%	3853	<0.01
	ENT	0.05+0.84T-0.69Td-0.48U+0.30V	10%	3636	<0.01
	REB	-0.006+1.33T-0.89Td-0.68U+0.36V	18%	3730	<0.01
NO_2	ANT	0.06+2.53T-1.22Td+0.30V	20%	3828	<0.01
	ERM	-0.06+2.29T-0.81Td+0.29V-0.23U	17%	3810	<0.01
	ENT	-0.03+1.87T-0.51V-1.02Td-0.51U	9%	3770	<0.01
	REB	0.02+1.40T-0.7V-0.78Td	11%	3490	<0.01
PM10	ANT	-0.06+1.18T+0.14U	14%	3760	<0.01
	ERM	-0.055+0.94T+0.11U	8%	3767	<0.01
	ENT	-0.002+0.88T-0.22V-0.12U	9%	3827	<0.01
	REB	0.035+0.88T-0.24V-0.32Td	12%	3515	<0.01
		Seasonal			
O_3	ANT	-0.036+2.30V-3.26U+1.12T-1.13Td	60%	2490	<0.01
	ERM	0.212+2.05V-3.35U+2.1T-2.06Td	72%	2849	<0.01
	ENT	0.246+2.19T-2.94Td-3.69U+1.32V	70%	2772	<0.01
	REB	0.088+2.67T-3.71Td-4.11U+1.11V	73%	2755	<0.01
NO_2	ANT	0.636-3.26Td+1.98T-1.21V+0.70U	44%	2856	<0.01
	ERM	0.309-4.50Td+2.89T-0.42V-0.70U	36%	2849	<0.01
	ENT	0.142+0.99T-3.51V-3.22Td+0.29U	54%	2771	<0.01
	REB	0.344-0.27T-2.55V+1.55U-0.96Td	61%	2752	<0.01
PM10	ANT	0.154+0.42U+1.76T-1.99Td-0.39V	38%	2856	<0.01
	ERM	-0.204+0.89U-1.88Td+1.64T-0.54V	40%	2849	<0.01
	ENT	0.72-0.99Td+1.07U-1.32V+0.48T	34%	2785	<0.01
	REB	0.123+1.48U-1.30V+0.52T-0.78Td	37%	2542	<0.01
		Long-term			
O_3	ANT	-411.19+7.28T-0.96Td+2.59V-1.88U	73%	2490	<0.01
	ERM	-290.64+5.29T+2.99V-1.96U-0.59Td	90%	2849	<0.01
	ENT	-1464.47+17.02T+13.88V+3.60Td-3.65U	72%	2770	<0.01
	REB	-775.20+8.94T+7.70V+2.74Td-1.98U	59%	2755	<0.01
NO_2	ANT	-532.97+8.74V+12.15T-4.69Td-0.57U	85%	2849	<0.01
	ERM	-221.06+8.81V-2.47U+3.11T	66%	2849	<0.01
	ENT	-521.08-1.05Td+8.45T-4.54U+6.99V	80%	2771	<0.01
	REB	748.82+6.06U-9.26V-7.24T-1.59Td	35%	2752	<0.01
PM10	ANT	371.82-6.40V-5.63T+1.69Td	49%	2849	<0.01
	ERM	62.36-3.72U+1.61Td-1.43V-1.79T	51%	2849	<0.01
	ENT	2765.16-29.03T-27.96V-7.95Td+3.78U	80%	2783	<0.01
	REB	1824.85-17.97T-19.78V-6.47Td+3.63U	72%	2542	<0.01

ANT – Antas; ERM – Ermesinde; ENT – Entrecampos; REB – Reboleira

The results shown in Table 3.2 depend on the analysed component, the pollutant considered and the air quality station selected. For all air quality stations, the relationship between the air quality and the meteorological variables for the short-term component is very low. For this component, temperature is the meteorological variable that presents the highest explained variance for all analysed pollutants.

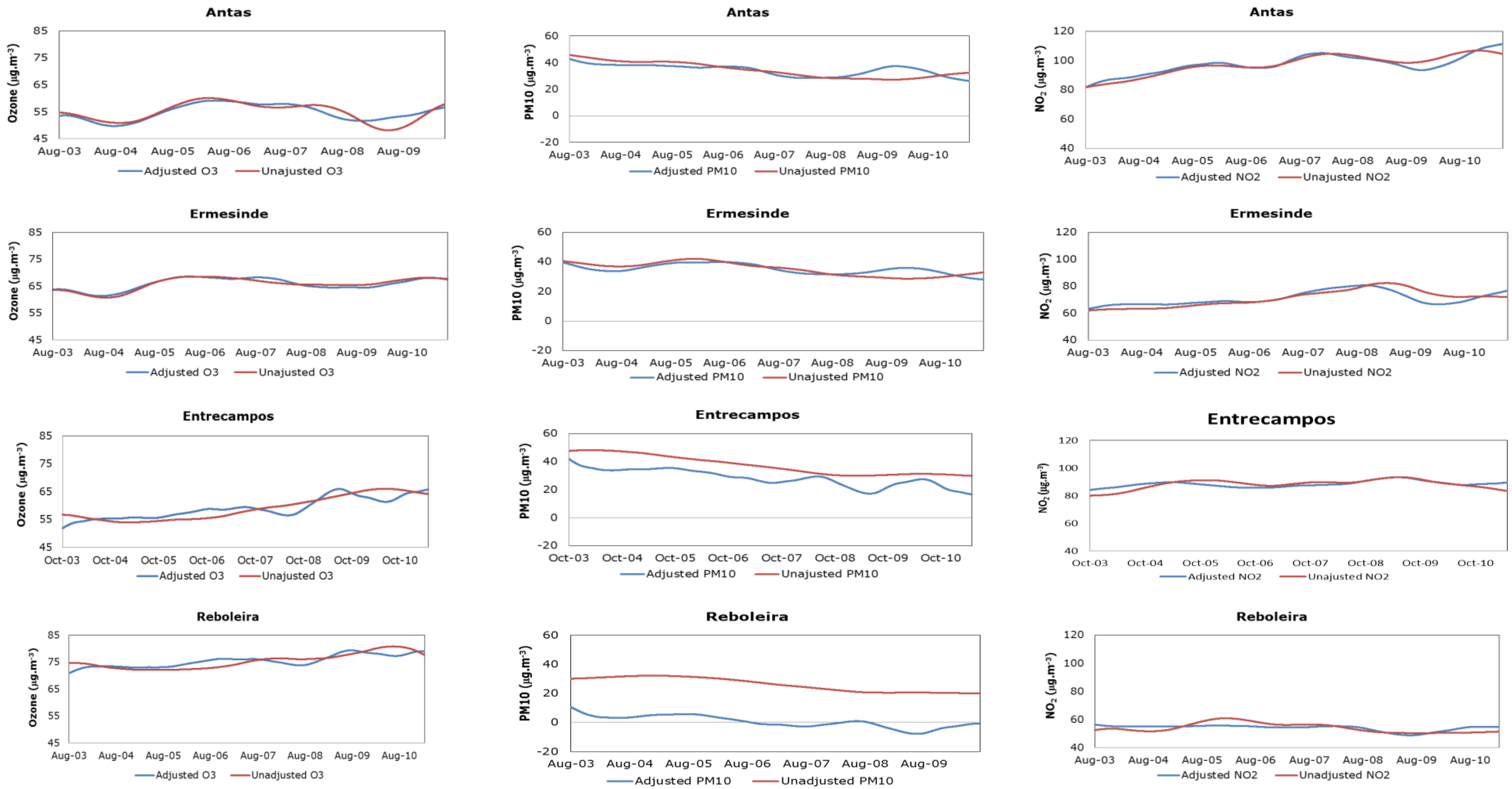
In general, the long-term component has the highest explained variance, followed by the seasonal component, achieving values higher than 58 %. For O_3, in terms of the seasonal and long-term components, the variability of the data can be explained by the four meteorological variables, ranging from 59 % to 90 %. For the other two pollutants, the percentage of the data variability is lower, i.e., from 35 % to 85 % for NO_2 and between 34 % and 80 % for PM10. As expected, the range of values indicates, as expected the largest effect of meteorology is on O_3 when compared to the other two pollutants.

The meteorological variable that has the highest contribution to the variability of the O_3 seasonal component depends on the analysed urban area, i.e., the V component of the wind in the Porto urban area and the temperature in the Lisboa urban area. This is in agreement with the fact that in northern Portugal, the northerly winds (meridional wind or V-wind component) are dominant. For the O_3 long-term component, temperature mostly contributes to the variability in both urban areas, which is in agreement with other studies (Flaum et al., 1996; Yang and Miller, 2002; Wise and Comrie, 2005). The correlations with all meteorological variables are higher for the long-term component with the exception of the Reboleira air quality station where the seasonal component has a higher correlation value. For the seasonal and long-term components, the temperature and V-wind component are directly related to O_3 concentrations. However, the U-wind component and Td are not directly related with the exception of the air quality stations located in the Lisboa urban area.

In the Porto urban area, the meteorological variable that has the highest contribution to the variability of the NO_2 data depends on the analysed component. For the short-term, seasonal and long-term components, the temperature, dew point temperature and V-wind component are dominant, respectively. On the contrary, for all the three components in the Lisboa urban area, temperature was the meteorological variable that most contributed to the variability of the NO_2 data with the exception of the long-term component at the Reboleira air quality station in which the U-wind component was dominant. Moreover, the long-term component at this air quality station as the lowest variance explained between the air quality data and the meteorological variables (35 %). At all other air quality stations, the variance explained for the long-term component is greater than 65 %. The temperature and dew point temperature are directly related to NO_2 concentrations for all temporal components and air quality stations. However, the U- and V-wind components depend on the analysed air quality station. The Reboleira air quality station differs from the other stations in terms of the long-term component because the U-wind is directly related to NO_2 and the V-wind is not directly related to NO_2. The Ermesinde air quality station is the only air quality station in which the U-wind meteorological variable is not directly related to NO_2 concentrations in terms of the seasonal component.

For the variance explained between meteorological variables and PM10 air quality data, the long-term component has the highest values, followed by the seasonal component. In the Lisboa urban area, the variance explained for the long-term component is significantly higher than in the Porto urban area. In Lisboa, temperature contributes more to the variability of the PM10 long-term component; in Porto, the U- and V-wind components are dominant depending on the air quality station. Unlike the other two pollutants, PM10 concentrations are not always directly related to temperature. This finding is verified for the long-term component at all the air quality stations where the coefficients for temperature are negative. For the same component, Td is directly related to PM10 in the Porto urban area and indirectly related in the Lisboa urban area. Moreover, the correlation between the U-wind component and PM10 concentrations differs for each analysed urban area, e.g., directly related for Lisboa. The V-wind component is indirectly related to PM10 concentrations in both urban areas.

Figure 3.3 provides a comparison between the O_3, NO_2 and PM10 long-term data and calculated O_3, NO_2 and PM10 based on the best model of the meteorological variables from the stepwise regression. From Figure 3.3, it is possible to conclude that the variability of the O_3 and NO_2 long-term data are better justified by the meteorological variables analysed in this study than the PM10 long-term data because there are larger differences between the original long-term data and the calculated long-term data for PM10. These results are in agreement with the results from the stepwise regression for the explained variances, which are higher for O_3 and NO_2 than for PM10.



a) b) c)

Figure 3.3 - Representation of the original long-term O_3 (a), NO_2 (b) and PM10 (c) (blue) and the respectively calculated O_3, NO_2 and PM10 based on the best model of meteorological variables from the stepwise regression (red line).

The air quality stations with urban traffic influences have a different behaviour between the original and calculated O_3 long-term data since 2007 based on the best model of the meteorological variables from the stepwise regression. The Ermesinde air quality station has the lowest differences between original and calculated O_3 long-term data, with a higher absolute difference of 2 $\mu g.m^{-3}$ throughout the entire study period. This result is expected because the four meteorological variables justify 90 % of the variability of the O_3 long-term data (Table 3.2).

The urban traffic air quality stations have higher correlations for NO_2 for the long-term component than the urban background air quality stations. However, there are no significant differences between the original and calculated NO_2 long-term data for all air quality stations.

There are evident differences in PM10 between the representations of the original and calculated long-term data between the air quality stations in the Porto and Lisboa urban area. In the Porto urban area, the difference between the original and calculated long-term data is lower than in the Lisboa urban area.

3.4 Summary and Conclusions

The objective of this study was to analyse air quality trends and to estimate the influence of the meteorological conditions on O_3, NO_2 and PM10 concentrations measured in two of the most critical agglomerations of Portugal: Lisboa and Porto. The application of the KZ filter allowed the separation of different components present in the meteorological and air quality datasets: short-term, seasonal and long-term. The obtained air quality components allowed the estimation of the contribution of each component to the total variance of the original air quality time-series. Moreover, with an application of stepwise regression modelling to the filtered time-series, the best meteorological predictors for the air quality data were estimated, and the differences between the original and calculated long-term air quality data were verified.

For the three analysed pollutants, the short-term component has the highest contribution to the total variance of the original time-series, followed by the seasonal component. These results suggest that the short-term component must be removed to not obscure the trends in the original data; the effects of meteorology should also be removed to better analyse the effects of long-term changes in emission levels over time, which is present in the air quality data. Air quality long-term trend analysis identified that the changing emission levels of implemented policies through air quality plans affected PM10 and NO_2 over the last several years.

It is clear from the obtained results that NO_2 and PM10 were not as weather-dependent as O_3 for the different analysed components. However, the method is adequate for determining the meteorological variables that mostly influence NO_2 and PM10 and for examining long-term trends.

The analysis showed that O_3 concentration has a statistically significant relationship to temperature for most of the components. Therefore, to establish an accurate air quality strategy to further improve air quality in terms of O_3, it will be essential to include the effects of meteorology on the analysis. For NO_2 and PM10, a meteorological variable with higher influence was not identified. However, this finding depends on the analysed temporal component and the selected air quality station. In the case of NO_2, the long-term component for the two air quality stations that are influenced by urban traffic explains the most variance. The highest explained variance for PM10 is found at both Lisboa air quality stations. The emission mitigation strategies play an important role in the PM10 and NO_2 concentrations; this should be further explored and considered in the long-term air quality strategies and policies.

This study demonstrated that the KZ filter analysis of air quality time series is a powerful technique that can provide important information about the cause-effect relationship in pollutant concentration variations. The findings from the current work contributes to better understand the nature of the processes behind the measurements in the Lisboa and Porto urban areas, which is required for accurate air quality management. Moreover, air pollutant measures to reduce pollutant emissions can only be effective to a certain extent because additional variables, e.g., meteorology, may play a crucial role in the air quality conditions of a certain region.

Chapter 4

4 Development of current and future pollutant emissions for Portugal

The contents of this chapter will be published in:

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Abstract

Air pollutant emissions are a fundamental input for accurate air quality simulations. Therefore, a detailed estimation of current emissions should be performed, mainly for the activity sectors that have higher contributions to emission totals. In order to estimate air quality under climate change at regional scale, it is extremely important to provide the most accurate emission inventories based on the emission scenarios used as input for the global climate models. The Representative Concentration Pathways (RCPs) are the most recent developed emission scenarios. Emission inventories used in air quality simulations at regional scale for future periods should be based on these recent developments. In this sense, an Emission Projections under RCP scenarios (EmiPro-RCP) model was developed to assist the estimation of future emission inventories for GHG and common air pollutants. This paper describes the methodology developed under EmiPro-RCP model and presents the estimation of current and projected emissions for Portugal for CO, PM2.5, PM10, SOx, NOx, NMVOC and NH_3, which will be used as input in air quality modelling systems. A comparison between the inventories was performed and the results indicated that all the RCPs scenarios predict a decrease in most of the air pollutant emissions until 2100, with the exception of NH_3 that increases. The main decreases are found in the coastal zone of Portugal, mainly in Porto and Lisboa urban areas, while the NH3 increases are located not only in the coastal zone but also in the southern inland of Portugal.

4.1 Introduction

Atmospheric dispersion is a complex process which depends on topography, land use, meteorology and emissions (Seinfeld and Pandis, 2006). In the last decades, these data have been systematized allowing a general use of air quality models and an increasingly complex estimation of air concentrations (Zhang et al., 2012a; Zhang et al., 2012b). However, despite the complexity behind the development of these models, the accuracy and precision of their results are often high and usually associated with the emission inventories used as input in those models (Taghavi et al., 2005).

Emission inventories quantify the mass of a primary pollutant emitted from different sources over a period of time and they are a fundamental part of most air quality and climate modelling, forecasting and policy assessments. Inventories are the means by which the relative importance of different sources to emissions of a given pollutant can be expressed and therefore allow an appreciation of how measures brought in to control one pollutant might affect another (AQEG, 2007).

The European Commission requires that all 28 MS of the EU report annual information concerning emissions and projections for four main air pollutants: SO_2, NOx, non-methane volatile organic compounds (NMVOC), and ammonia (NH_3) in order to provide better assessment and evaluation of acidifying gases emissions trends. In addition the European Commission also required that the MS prepare the National Inventory of Greenhouse Gas (GHGs) Emissions and sinks to comply with the international commitments under the UNFCCC and the Kyoto Protocol (EEA, 2010b). The UNFCCC Guidelines require that Parties set up a National Inventory Report (NIR) as one part of their annual submissions. The NIR should contain detailed and complete information related to methodologies, emission factors, activity data, and should give explanations concerning any recalculations of historical inventories, in order to ensure transparency and enable the inventory review (UNEP, 2009). The application of a common methodology is fundamental for the Intercomparison of the GHG inventories.

Atmospheric emissions inventories are usually quantified using one of two approaches: (i) top-down, based on the disaggregation process of total emissions from a certain area to smaller administrative units or a regular grid with higher resolution (Ossés de Eicker et al., 2008); and (ii) bottom-up, based on emissions estimation using detailed data of each emission source (Zhao et al., 2011).

The Portuguese national emission inventory (INERPA) (PEA, 2011), which is a combination of top-down and bottom-up approaches, provides quantitative information for the main atmospheric pollutants: CO, CH_4, NOx, nitrous oxide (N_2O), SOx, NMVOC, NH_3, PM10 and PM2.5.

In the early 90s, there was an increase of the Portuguese emissions. Thereafter, the growth of emissions has been more moderate and even a sort of stagnation tended to happen in the most recent years. This situation is, in part, a result of the implementation of some measures, such as the introduction of natural gas (1997), the installation of combined cycle thermoelectric plants using natural gas (1999), the progressive installation of co-generation units, the amelioration of energetic and technologic efficiency of industrial processes, the improvement in car efficiency and the improvement of fuels' quality. However, the positive effect of the implementation of these measures has been outweighed by the overall increase of energy consumption which mainly relies on fossil fuel sources. Furthermore, in most recent years there has been an impressive development and installation of equipment for the use of renewable energy sources with a particular expansion of windmills (PEA, 2010a). Despite the fast growing trends of the transport sector (mainly roads) since the 90s, the introduction of new petrol-engine passenger cars, with catalyst converters and stricter regulations on diesel vehicles emissions, limited the increase of the emissions or even resulted in their decrease. In fact, the situation started to change in the last years, as transport emissions growth has first stabilised and even started to decline in the most recent years (PEA, 2010b). Additionally to these facts, the financial crisis that Portugal has been facing over the last four years has several implications in the behaviour of the national emissions in these years (Borrego et al., 2012c).

Understanding the past developments in emissions of pollutants and greenhouse gases is a crucial step for understanding and modelling climate change, as well as in the development of future air pollution control policies (Isaksen et al., 2009).

The most recent emission scenarios are the Representative Concentrations Pathways (RCPs), which are a result of a careful selection process, where the needs of both climate scenario developers and users were considered. As pointed out by van Vuuren et al. (2011) the definition of the RCPs follow several design criteria in order to facilitate climate research and assessment:

1) The RCPs are based on scenarios published in the existing literature, developed independently by different modeling groups and, as a set, be 'representative' of the total literature, in terms of emissions and concentrations; At the same time, each of the RCPs should provide a plausible and internally consistent description of the future;

2) The RCPs provide information on all components of radiative forcing that are needed as input for climate modeling and atmospheric chemistry modeling (emissions of greenhouse gases, air pollutants and land use). Moreover, they should make such information available in a geographically explicit way;
3) The RCPs have harmonized base year assumptions for emissions and land use and allow for a smooth transition between analyses of historical and future periods;
4) The RCPs cover the time period up to 2100, but information also needs to be made available for the centuries thereafter.

The final RCP selections were four RCPs: RCP2.6, RCP4.5, RCP6.0 and RCP8.5 (Moss et al., 2010) (Table 4.1).

Table 4.1 – Characteristics of the RCPs scenarios.

Name	Radiative forcing	Concentration (p.p.m.)	Pathway	Model providing RCP
RCP8.5 **a)**	>8.5 $W.m^{-2}$ in 2100	>1,370 $CO_{2equiv.}$ in 2100	Rising	MESSAGE
RCP6.0 **b)**	≈6$W.m^{-2}$ at stabilization after 2100	≈850 $CO_{2equiv.}$ (at stabilization after 2100)	Stabilization without overshoot	AIM
RCP4.5 **c)**	≈4.5 $W.m^{-2}$ at stabilization after 2100	≈650 $CO_{2equiv.}$ (at stabilization after 2100)	Stabilization without overshoot	GCAM
RCP2.6 **d)**	Peak at ≈3 $W.m^{-2}$ before 2100 and then declines	Peak at ≈490 $CO_{2equiv.}$ before 2100 and then declines	Peak and decline	IMAGE

a) Rao and Riahi, 2006 and Riahi et al., 2007; b) Fujino et al., 2006 and Hijioka et al., 2008

c) Clarke et al., 2007 and Smith and Wigley, 2006; d) van Vuuren et al., 2006 and van Vuuren et al., 2007a

In Moss et al. 2008, it was indicated that the RCPs should be used to initiate climate model simulations for developing climate scenarios to support a broad range of climate-change related research and assessment and were requested to be "compatible with the full range of stabilization, mitigation and baseline emissions scenarios available in the current scientific literature". Figure 4.1 shows how the air pollutant emission scenarios are not only relevant to the climate model simulations, but also for the regional air quality simulation under climate change.

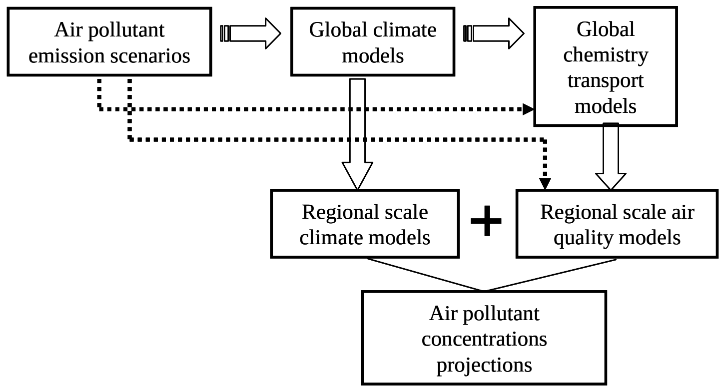


Figure 4.1 – Flowchart of global and regional air quality simulation under climate change.

In this sense, it is extremely important that the estimation of future emissions for regional air quality modelling application considers the emission scenarios used as input for the global climate models.

There are two main goals in this chapter: the first is to estimate current emissions for Portugal with a higher detailed disaggregation to improve the performance of the air quality simulations; the other consists in estimating projected emissions for Portugal based on the RCPs emissions scenarios in order to improve the results and keep the consistency of the climate change simulations.

4.2 Data and Methods

The 2012 INERPA will be analyzed and a new model was created to estimate national emission projections. A new methodology was developed for the estimation of current emissions and a new model was created to estimate national emission projections.

4.2.1 National emissions inventory

Emissions inventory are developed to provide a picture of the important sources of emissions and their trends (e.g. emissions increasing or decreasing). They also provide the information needed to track progress of emission reduction targets agreed, between many countries, for greenhouse gas emissions and emissions of harmful air pollutants.

The European Environment Agency (EEA) proposes as guidance on estimating emissions from both anthropogenic and natural emission sources the EMEP

CORINAIR methodology. The EMEP/EEA air pollutant emission inventory guidebook (formerly referred to as the EMEP CORINAIR emission inventory guidebook) is designed to facilitate reporting of emission inventories by countries to the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) and the EU National Emission Ceilings Directive (EEA, 2009). These guidelines define the format for reporting emission data and offer guidance on how to provide supporting documentation, through an Informative Inventory Report, which describes the activity data, emission factors, methodologies applied in the calculation, and explanation of the whole process of inventory preparation.

In Portugal, as a Party in the CLRTAP, the emission data is obtained from the methodology EMEP/EEA air pollutant emission. The procedures were defined according to Good Practice and Uncertainty Management Guide (IPCC, 2000) and adapted to the specific INERPA characteristics. The national emissions are reported for 11 main source categories "Selected Nomenclature of Air Pollution" (SNAPs) (Table 4.2). In this work only anthropogenic emissions will be analysed and in this sense SNAP11 will not be included.

Table 4.2– SNAP (Selected Nomenclature of Air Pollution) sector IDs, full name and short name.

SNAP	Full name	Short name
1	Combustion in energy and transformation industries	Energy industries
2	Non-industrial combustion plants	Residential/commercial combustion
3	Combustion in manufacturing industry	Industrial combustion
4	Production processes	Industrial processes
5	Extraction and distribution of fossil fuels and geothermal energy	Extraction/distribution of fossil fuels
6	Solvent and other product use	Solvent use
7	Road transport	Road transport
8	Other mobile sources and machinery	Other mobile sources
9	Waste treatment and disposal	Waste
10	Agriculture	Agriculture
11	Other sources and sinks	Other

The inventory provides quantitative information for the main atmospheric pollutants: CO, CH4, NOx, N_2O, SOx, NMVOC, NH_3, PM10 and PM2.5. Until 2009, the yearly emission data was available at both national level and spatial disaggregation to the municipality level (NUT IV), by activity and by pollutant. Since 2010, only total emission data in s Portugal, per activity and pollutant, have been made available for each year.

4.2.2 Representative Concentrations Pathways database

A RCP database is available in URL4, providing documentation of the emissions, concentrations, and land-cover change projections of the four RCPs. The emissions estimation for the four RCP scenarios is clustered in the webpage for the World and for the regions (OECD90, REF, ASIA, MAF and LAM), for all the scenarios (RCP2.6, RCP4.5, RCP6.0 and RCP8.5) and for several GHG and other common air pollutants.

The database is divided in regional aggregations of 5 regions (URL4):

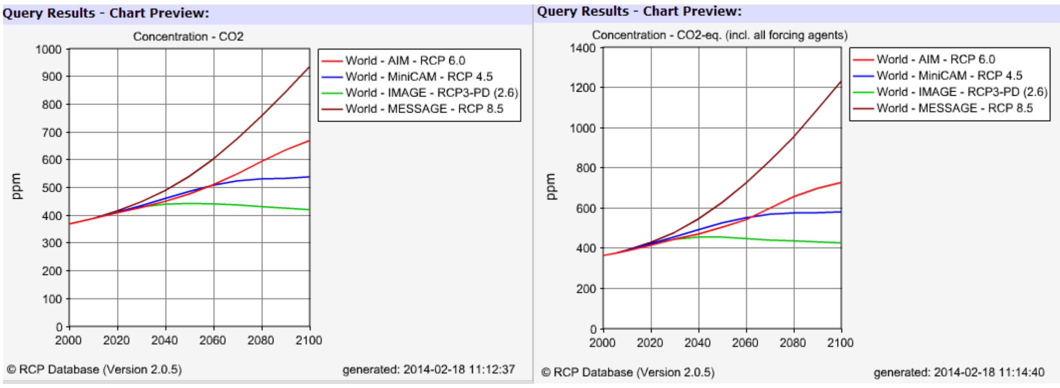
- OECD90 - Includes the OECD 90 countries, therefore encompassing the countries included in the regions Western Europe (Austria, Belgium, Denmark, Finland, France, Germany, Greece, Iceland, Ireland, Italy, Luxembourg, Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, United Kingdom), Northern America (Canada, United States of America) and Pacific OECD (Australia, Fiji, French Polynesia, Guam, Japan, New Caledonia, New Zealand, Samoa, Solomon Islands, Vanuatu) .
- REF - Countries from the Reforming Economies region (Albania, Armenia, Azerbaijan, Belarus, Bosnia and Herzegovina, Bulgaria, Croatia, Cyprus, Czech Republic, Estonia, Georgia, Hungary, Kazakhstan, Kyrgyzstan, Latvia, Lithuania, Malta, Poland, Republic of Moldova, Romania, Russian Federation, Slovakia, Slovenia, Tajikistan, TFYR Macedonia, Turkmenistan, Ukraine, Uzbekistan, Yugoslavia).
- ASIA - The countries included in the regions China + (China, China Hong Kong SAR, China Macao SAR, Mongolia, Taiwan) , India + (Afghanistan, Bangladesh, Bhutan, India, Maldives, Nepal, Pakistan, Sri Lanka) and Rest of Asia (Brunei Darussalam, Cambodia, Democratic People's Republic of Korea, East Timor, Indonesia, Lao People's Democratic Republic, Malaysia, Myanmar, Papua New Guinea, Philippines, Republic of Korea, Singapore, Thailand, Viet Nam) are aggregated into this region.
- MAF - This region includes the Middle East (Bahrain, Iran (Islamic Republic of), Iraq, Israel, Jordan, Kuwait, Lebanon, Oman, Qatar, Saudi Arabia, Syrian Arab Republic, United Arab Emirates, Yemen) and African (Algeria, Angola, Benin, Botswana, Burkina Faso, Burundi, Cote d'Ivoire, Cameroon, Cape Verde, Central African Republic, Chad, Comoros, Congo, Democratic Republic of the Congo, Djibouti, Egypt, Equatorial Guinea, Eritrea, Ethiopia, Gabon, Gambia, Ghana, Guinea, Guinea-Bissau, Kenya, Lesotho, Liberia, Libyan Arab Jamahiriya, Madagascar, Malawi, Mali, Mauritania, Mauritius, Morocco, Mozambique, Namibia, Niger, Nigeria, Reunion, Rwanda, Senegal,

Sierra Leone, Somalia, South Africa, Sudan, Swaziland, Togo, Tunisia, Uganda, United Republic of Tanzania, Western Sahara, Zambia, Zimbabwe) countries.

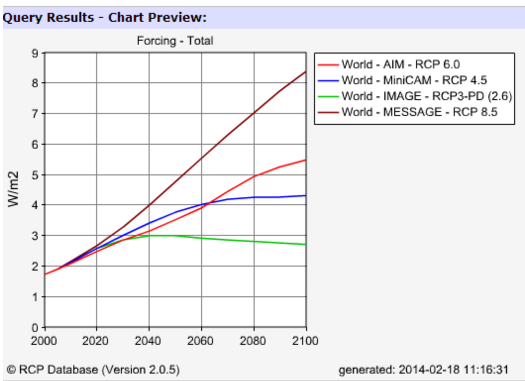
- LAM - This region includes the Latin American countries (Argentina, Bahamas, Barbados, Belize, Bolivia, Brazil, Chile, Colombia, Costa Rica, Cuba, Dominican Republic, Ecuador, El Salvador, Guadeloupe, Guatemala, Guyana, Haiti, Honduras, Jamaica, Martinique, Mexico, Netherlands Antilles, Nicaragua, Panama, Paraguay, Peru, Puerto Rico, Suriname, Trinidad and Tobago, Uruguay, Venezuela).

The projections from the RCP database are divided in twelve years (2000, 2005, 2010, 2020, 2030, 2040, 2050, 2060, 2070, 2080, 2090 and 2100) and it is also possible to select an extension to the year 2300.

Figure 4.2 shows some examples of the climate indicators of the RCPs: concentration (a) and radiative forcing (b).



a)



b)

Figure 4.2 – Climate indicators of RCPs (URL4).

From the climate indicators it is possible to understand the differences between the four RCP scenarios.

4.2.3 Estimation of current emissions

Most of the air quality studies require emissions with high spatial disaggregation. In this sense, a top-down methodology is applied to spatially disaggregate the total anthropogenic emissions, taking in consideration several assumptions.

The annual national emissions from anthropogenic area sources for each pollutant and activity sector were spatially disaggregated to the sub-municipality level for SNAP2 (S2), SNAP3 (S3), SNAP4 (S4), SNAP6 (S6), SNAP9 (S9), SNAP10 (S10) and to the municipality level for SNAP5 (S5), SNAP7 (S7) and SNAP8 (S8). The data are then converted to a regular grid, using GIS, for the spatial resolution required for the selected simulation domains. The disaggregation into the higher resolution grid was conducted by taking in consideration the area (km^2) of the municipality or sub-municipality, depending on the sector.

Since the traffic sector (SNAP7) is a major contributor to the concentration levels that are registered in Portugal it is relevant to better estimate the current emissions of the traffic sector. Also, the traffic sector is a line source and by the top-down methodology is considered as an area source. For these reasons, a bottom-up methodology combined with the top-down methodology was applied to this sector.

The main objective of the application of this methodology is to transform an area source into a line source. Figure 4.3 illustrates the sequence of this methodology.

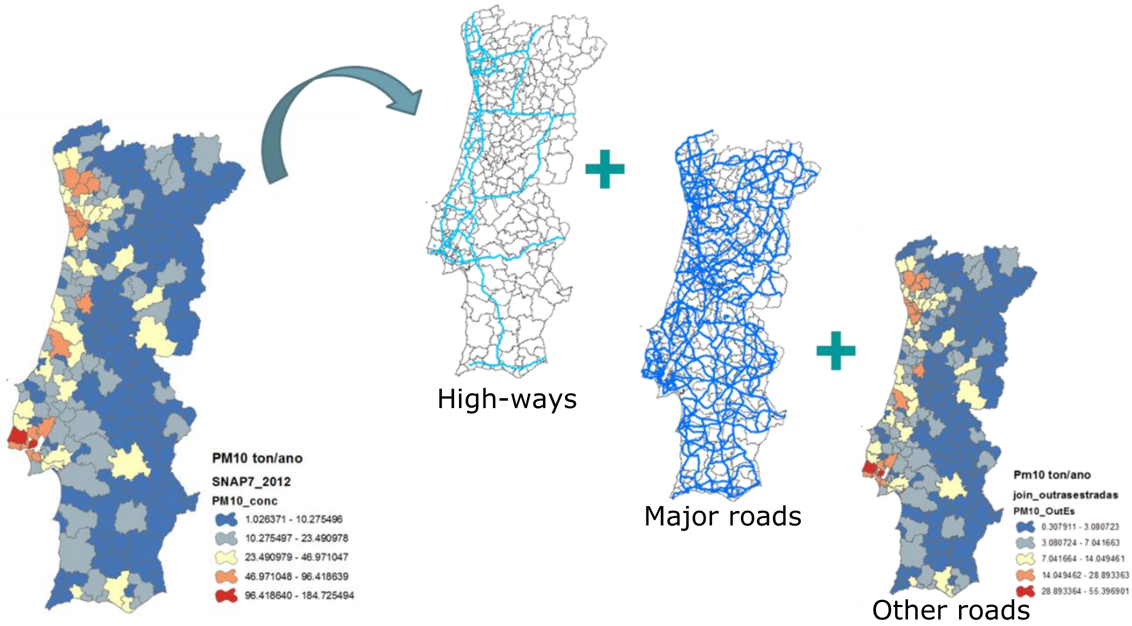


Figure 4.3 – Representation of SNAP7 methodology to transform area source into line source.

The emissions from SNAP7 will be divided in high-ways, major roads and other roads, the first two being considered as line sources and the last one as an area source because of the high number of this kind of roads.

Since the SNAP7 was already spatially distributed by municipality, the first part of this methodology is to calculate the traffic emissions for all the national high-ways. The traffic emissions from the high-ways are estimated by the TREM model. The Transport Emission Model for Line Sources (TREM) was developed at the University of Aveiro, to support the quantification of emissions induced by road traffic with high temporal and spatial resolution to be used in air quality modeling. The model is based on the emission functions derived from the MEET/COST methodology (Borrego et al., 2003). The emission rates for several atmospheric pollutants and fuel consumption are estimated as a function of average speed. Different technologies (engine type, model year) and engine capacities are distinguished. The model is particularly designed for line sources. Therefore, roads are considered as line sources and emissions induced by vehicles are estimated individually for each road segment considering detailed information on traffic flux. Total emission of the pollutant *p* (*Ep*) for each road segment is estimated by the model as following:

$$E_p = \sum_i (e_{ip}(v) \times N_i) \times L \quad \text{(Eq. 4.1)}$$

where$e_{ip}(v)$ is the emission factor for pollutant *p* and vehicle class *i* as a function of average speed *v*, N_i is the number of vehicles of class *i* and *L* is the road segment length. In the current model version the calculation algorithm is implemented for the following pollutants: CO; NOx; VOC; CO_2; SOx and PM. Furthermore, fuel consumption is estimated to provide additional data to be compared with statistical information as a measure of model evaluation. A schematic of TREM model is shown in Figure 4.4.

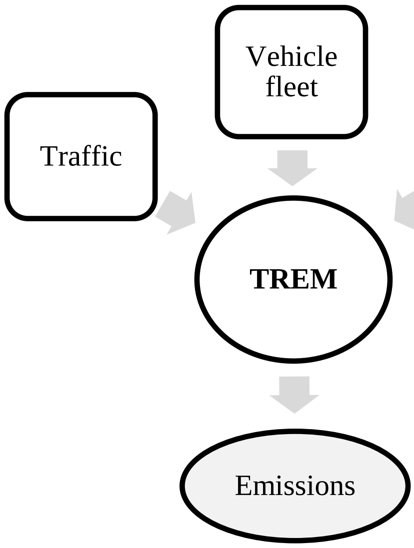


Figure 4.4– Inputs and outputs of the TREM model.

By the application of the TREM model, the emissions of VOC, CO, PM10, SOx were calculated for each national high-way. The first output from this methodology is the emission of high-ways as a line source. The second step consists in using the GIS tool to calculate the emissions of the high-ways associated to each municipality. Then, for the output from the top-down methodology for SNAP7, it is necessary to subtract, in each municipality, the emissions from the high-ways.

Knowing the Portuguese behavior in terms of traffic flow (Bandeira et al., 2011), it was assumed that the circulation of vehicles on the major roads corresponds to 70% and the other roads to 30%. Therefore, to the result of the subtraction performed before 70% of the emission value by municipality was calculated, obtaining the emission of the national major roads by municipality. Using the GIS tool, the area emission of the major roads was transformed in line sources, and the emission from other roads (30%) was converted to a regular grid for the spatial resolution required for the selected simulation domains.

4.2.4 Estimation of emissions projections

An Emission Projections under RCP scenarios (EmiPro-RCP) model was developed to estimate the future emissions at regional level used as input in the air quality models, based on the emission scenarios used as input for the global climate models.

The climate change simulations discussed in the next chapter will consider the latest available scenarios; therefore the model was developed based on the RCPs scenarios.

The model consists of a software package, based on the Python language, to estimate the emissions for several study regions. EmiPro-RCP is a user-friendly, PC-based model that can be used in Windows or Linux server.

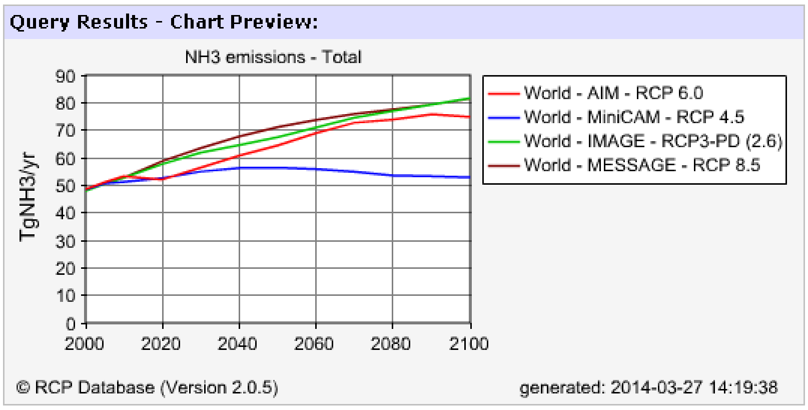
The emission projections and historical data in the RCP database are organized by sectors, which differ from the SNAP classification. A correspondence between the RCP division and the SNAPs classification was performed to allow use the national emissions in the EmiPro-RCP model (Table 4.3).

Table 4.3– Correspondence of the RCP database division and the SNAPs classification.

RCP database division	SNAPs
Surface Transportation	7 and 8
International Shipping	8
Aviation	8
Power Plants, Energy Conversion, Extraction, and Distribution	1 and 5
Waste (landfills, wastewater, non-energy incineration)	9
Industry (combustion & processing)	3 and 4
Residential and Commercial	2
Agriculture (Animals, Rice, & Soil)	10
Agriculture (waste burning on fields)	10
Grassland burning	11
Forest burning	11
Solvents	6

Twelve years were selected for the EmiPro-RCP model, being the first three (2000, 2005 and 2010) considered as the base years and the remaining nine (2020 until 2100) as the projection years.

As an example, Figure 4.5 illustrates the worldwide projections of RCPs for NH_3 and NOx.



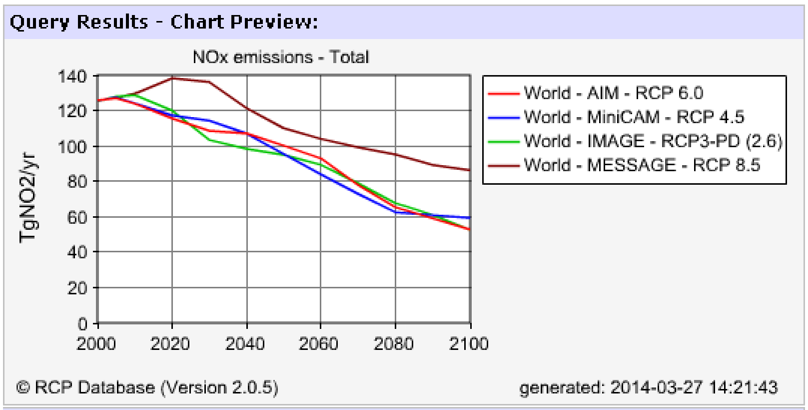


Figure 4.5 – Emissions projections of NH_3 and NOx based on the RCPs scenarios from the website.

The EmiPro-RCP model is based on a questionnaire that should be answered to select the base year, the future year, the RCP scenario, the study region and if the emissions provided as input are total or by SNAPs. With the information provided from the RCP database the EmiPro model calculates the emission factor for each pollutant, depending on the base year, the SNAP (or total emission), the study region, the scenario and the selected year of projection. Then, using as input the current emissions for the study region, the model computes the future emissions taking in consideration the variable chosen. Figure 4.6 shows the schematic representation of the EmiPro-RCP model.

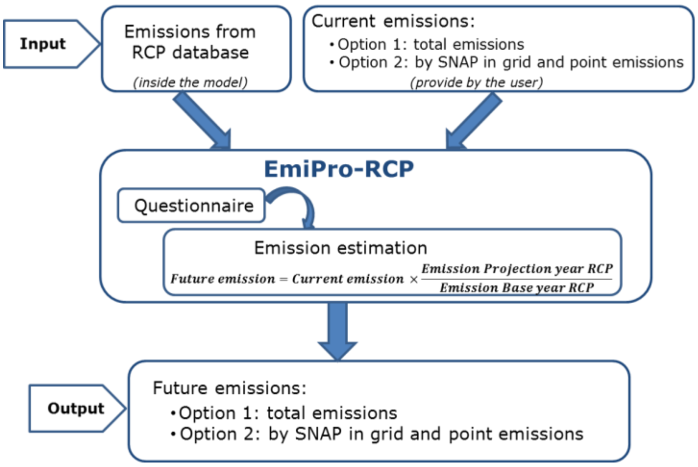
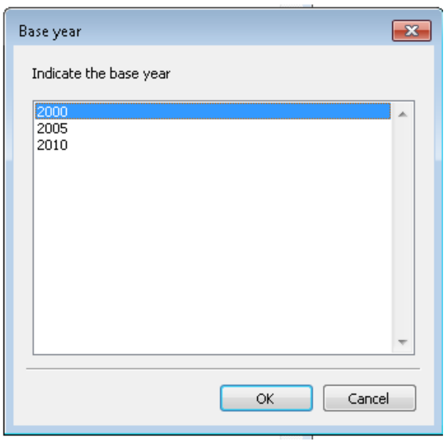
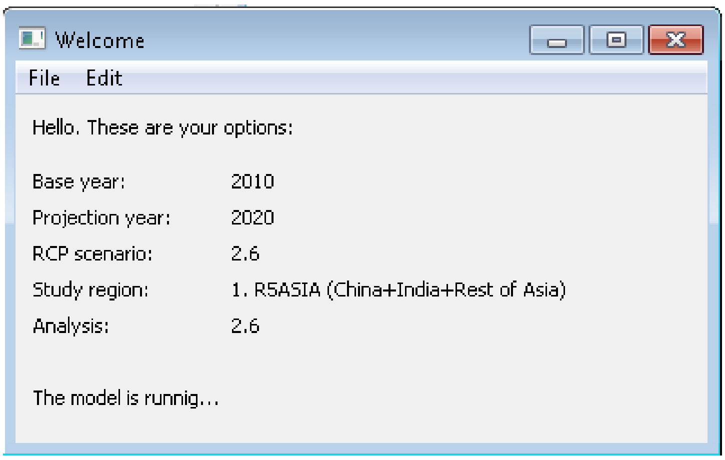


Figure 4.6 – Flowchart of EmiPro-RCP model.

An example of one of the questions of the questionnaire is in Figure 4.7a) and in Figure 4.7b) is the latest information of the model, where all the answers of the questionnaire are presented.



a)



b)

Figure 4.7– Questionnaire of the EmiPro model.

This methodology allows that the grid emissions required by the air quality models can be use directly in the EmiPro-RCP model, however this methodology do not consider the socio-economic or land-use changes that could happen in the study domain. One of the reasons is to keep the trends defined in the RCP scenarios for OECD90. Moreover, there is no available data to characterize consistently the land-use changes in Portugal, based on the RCPs scenarios.

4.3 Results and discussion

4.3.1 Current emissions

The national 2012 total annual emissions by SNAP and for each considered pollutant are presented in Table 4.4. Also, with the information of Figure 4.8 it is possible to identify, for each pollutant, the SNAP with higher emission value.

Table 4.4 – Total annual emissions by SNAP for Portugal in 2012 (ton).

SNAP	**CO**	**SOx**	**NOx**	**NH_3**	**NMVOC**	**PM2.5**	**PM10**
1	5109	8765	24219	4	1427	487	714
2	119182	2243	6600	0	17995	24265	24853
3	19381	20051	29570	462	6721	6940	7615
4	28426	9929	5927	1894	49078	13861	26662
5	0	0	0	0	3822	0	0
6	0	0	0	0	59094	1606	1606
7	86263	97	67500	1019	15302	3684	4412
8	8703	1980	24312	2	2771	2832	2832
9	13639	207	1019	2079	9804	0	2530
10	24808	98	2075	42020	2484	2307	2307

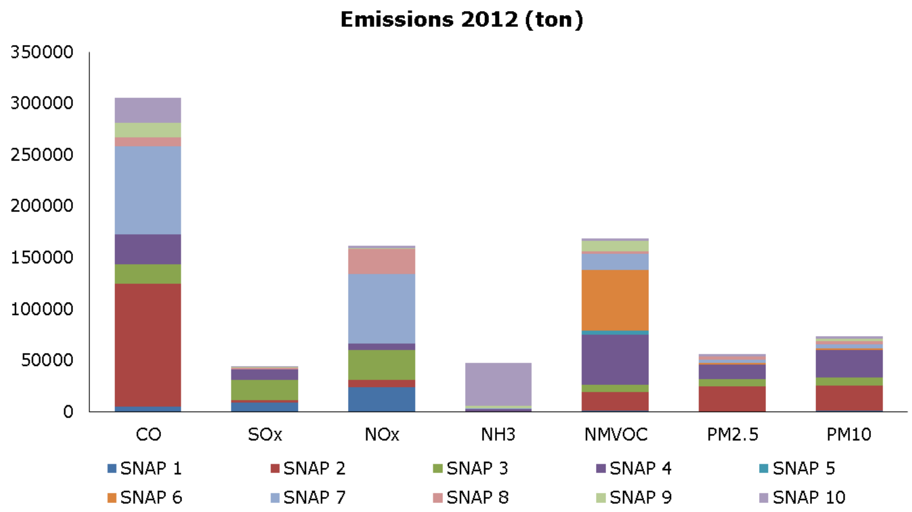


Figure 4.8– Distribution of the SNAPs emission value for each pollutant (ton).

The higher emission values of CO and PM2.5 could be found in the SNAP2, which results from the residential/commercial combustion processes. CO also has higher values in SNAP7 (traffic sector). The other pollutant with higher contribution to the SNAP7 emissions is NOx which is, accordingly with the analysis from Chapter 2, where traffic is the most responsible sector of NOx concentration values.

As expected, the SNAP with higher SOx emission values is SNAP3 (industrial combustion), for NH_3 emissions is SNAP10 (agriculture) and NMVOC emissions is SNAP6 (solvent use).

For PM10, the higher emission values are found in SNAP2 and SNAP4 (industrial processes).

The spatial distribution of the national emissions by municipality and sub-municipality (depending on the sector) for each pollutant are represented in Figure 4.9. In order to focus the analysis, there are only represented, for each pollutant, the sector with higher emission value: CO (S2), PM2.5 (S2), SOx (S3), PM10 (S4), NMVOC (S6), NOx (S7) and NH_3 (S10). The maps with the spatial distribution of the emission values of all the pollutants for all the SNAPs are collected in Appendix B.

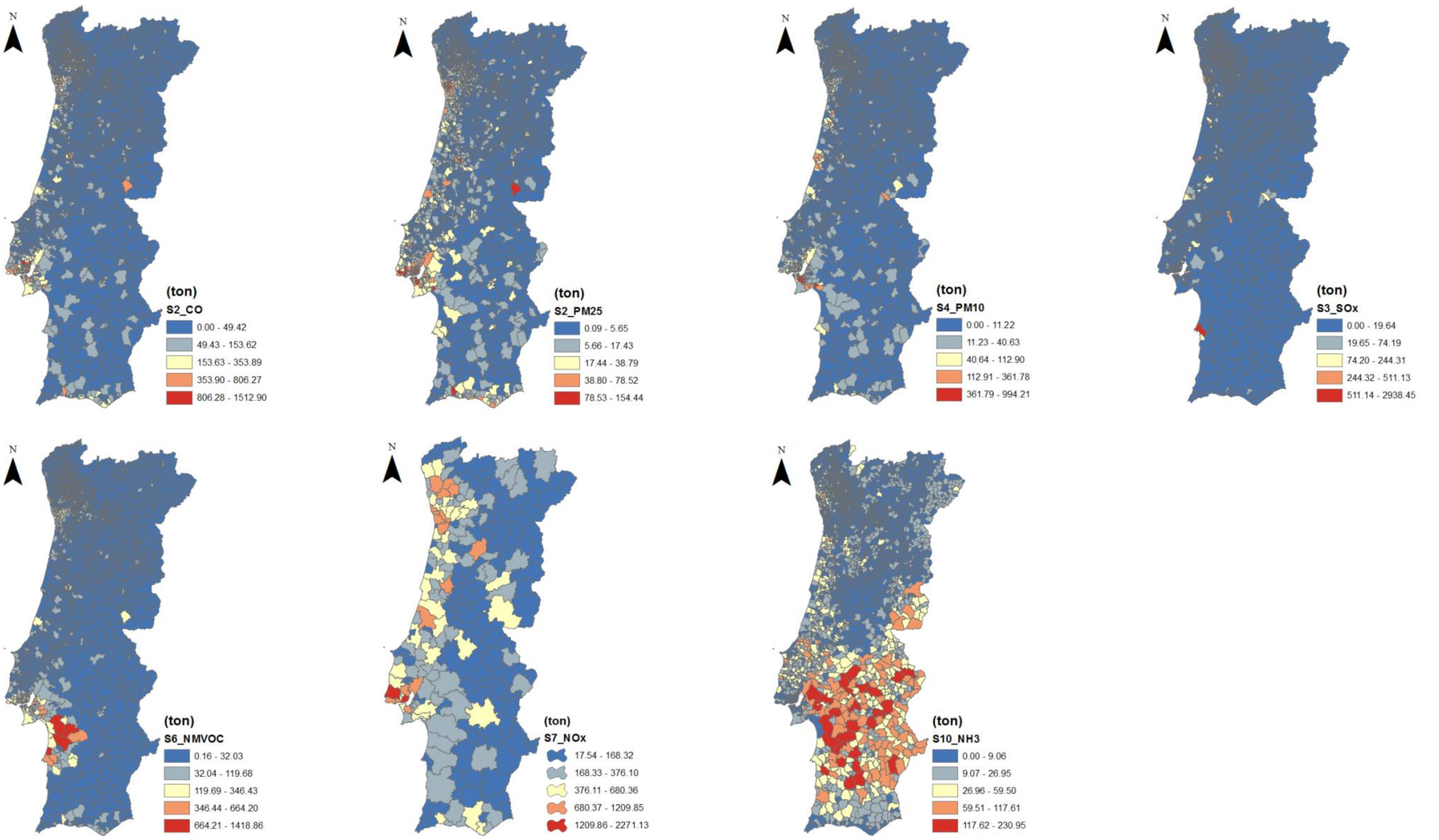
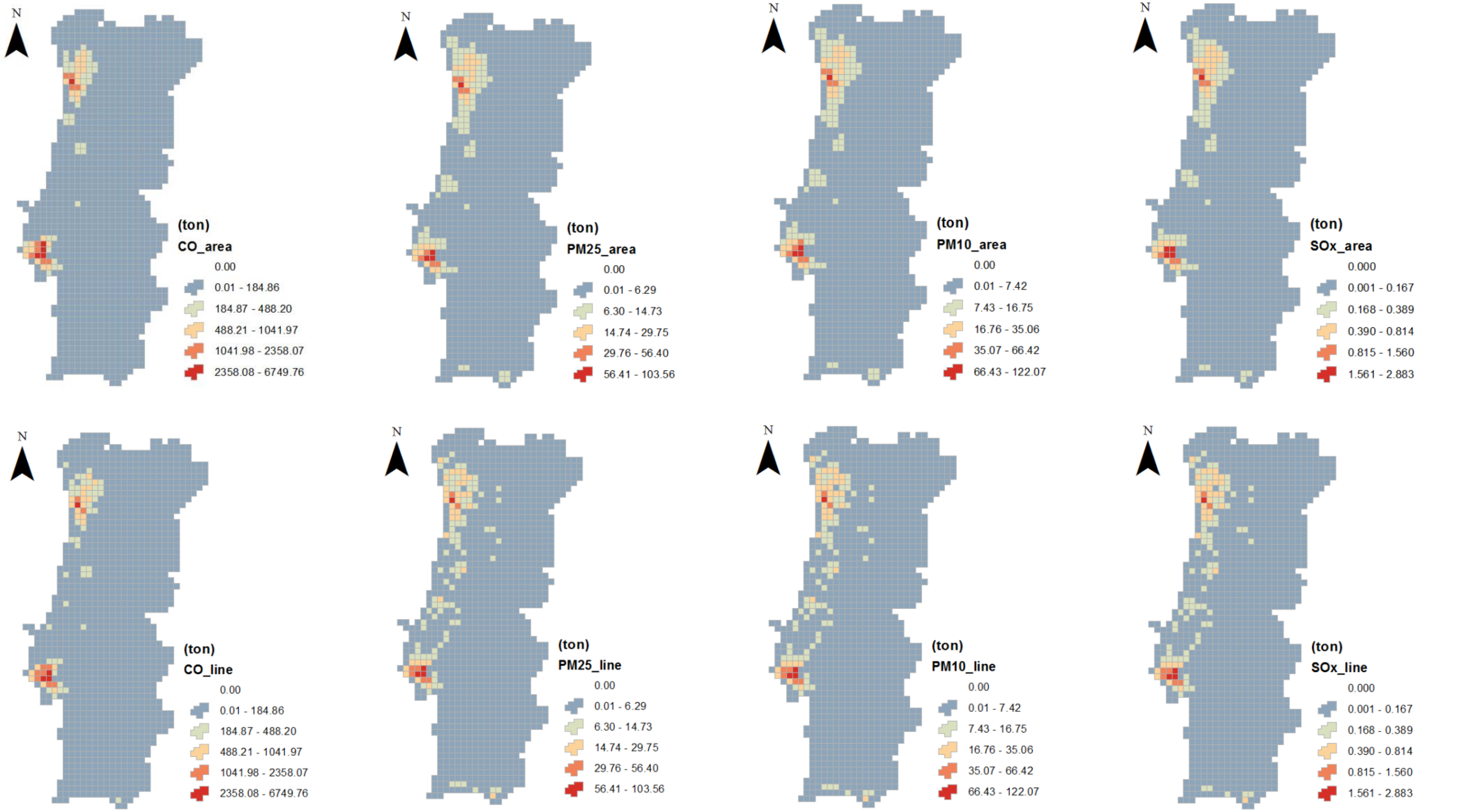


Figure 4.9– Spatial distribution of the emission value by municipalty and sub-municipality (depending on the SNAP analysed): CO (S2), PM2.5 (S2), SOx (S3), PM10 (S4), NMVOC (S6), NOx (S7) and NH_3 (S10).

The spatial distribution of the emissions depends on the pollutant and the SNAP analysed. However, in general, from Figure 4.9, it could be concluded that the highest emission values related with traffic (NOx), residential/commercial combustion (CO and PM2.5) and the industrial areas (SOx, PM10 and NMVOC) are in the coastal region (mainly in the urban areas of Porto and Lisbon). The only exception is for NH_3 since it is most related with rural areas.

For SNAP7 a detailed work was performed to improve how the traffic emissions were spatial distributed in Portugal, Figure 4.10 shows the comparison between the results from the two methodologies to spatial distribute traffic emissions. The maps above are the result of the spatial distribution by municipality and then converted to a regular grid (top-down approach). The other maps are the result of a combination of top-down and bottom-up approaches.


N
(ton)
CO_area
0.00
0.01 - 184.86
184.87 - 488.20
488.21 - 1041.97
1041.98 - 2358.07
2358.08 - 6749.76
(ton)
PM25_area
0.00
0.01 - 6.29
6.30 - 14.73
14.74 - 29.75
29.76 - 56.40
56.41 - 103.56
(ton)
PM10_area
0.00
0.01 - 7.42
7.43 - 16.75
16.76 - 35.06
35.07 - 66.42
66.43 - 122.07
(ton)
SOx_area
0.000
0.001 - 0.167
0.168 - 0.389
0.390 - 0.814
0.815 - 1.560
1.561 - 2.883
(ton)
CO_line
0.00
0.01 - 184.86
184.87 - 488.20
488.21 - 1041.97
1041.98 - 2358.07
2358.08 - 6749.76
(ton)
PM25_line
0.00
0.01 - 6.29
6.30 - 14.73
14.74 - 29.75
29.76 - 56.40
56.41 - 103.56
(ton)
PM10_line
0.00
0.01 - 7.42
7.43 - 16.75
16.76 - 35.06
35.07 - 66.42
66.43 - 122.07
(ton)
SOx_line
0.000
0.001 - 0.167
0.168 - 0.389
0.390 - 0.814
0.815 - 1.560
1.561 - 2.883

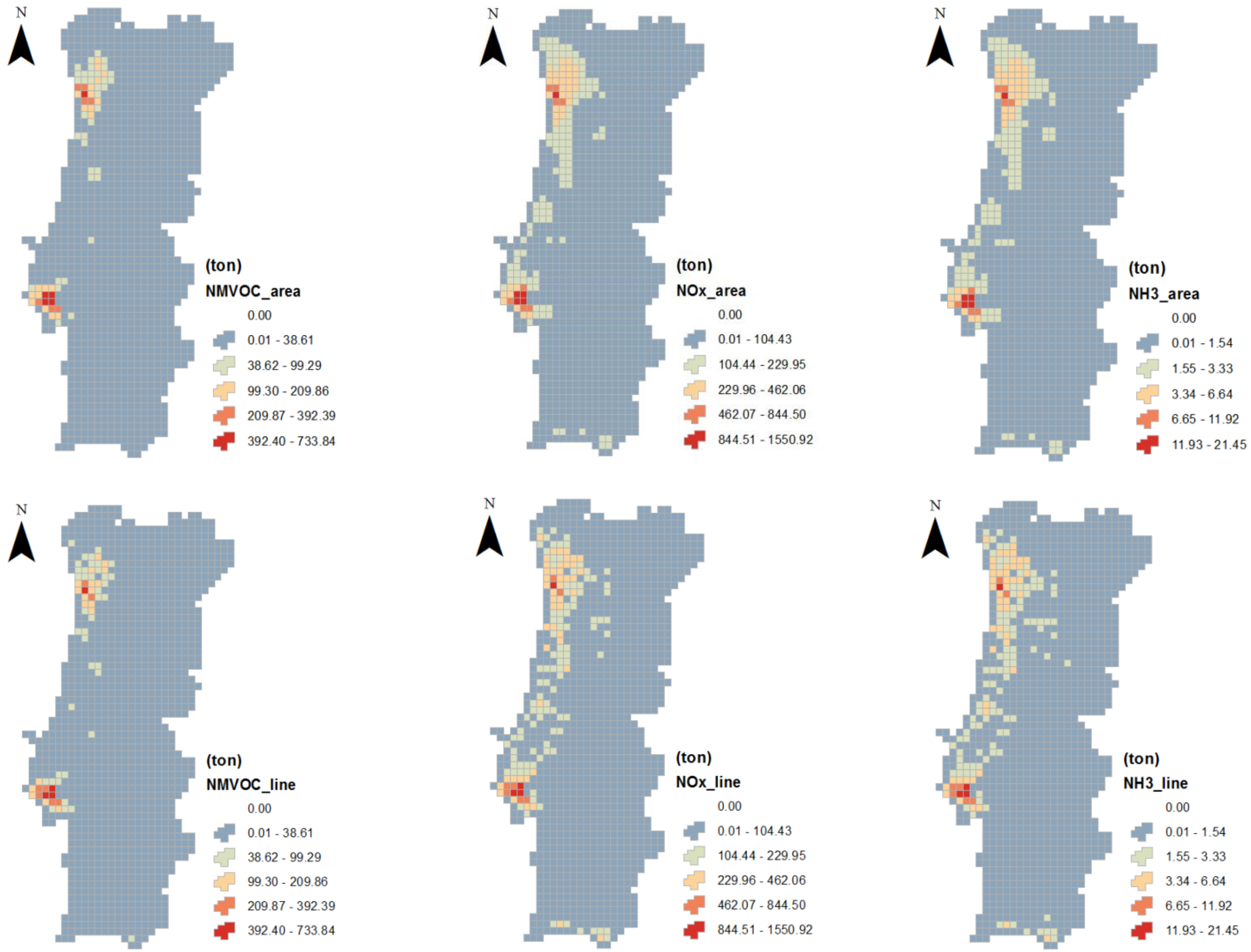


Figure 4.10– Comparison of SNAP7 emissions using top-down approach (left maps) and combination of top-down and bottom-up approaches (right maps) for CO, PM2.5, SOx, PM10, NMVOC, NOx and NH_3.

The improvement of the SNAP7 emissions spatial distribution is higher in NH_3, NOx, PM10, PM2.5 and SOx. In the case of CO and NMVOC, the difference is not so evident since the emissions of these pollutants are concentrated in the urban areas: Porto and Lisbon (Appendix A – S7 – CO and NMVOC). The other pollutants have their emissions more dispersed through Portugal (Appendix A – S7) and the added value of using line sources is more evident. With this methodology, the cells that usually had the same value of the surrounding cells, now have a more accurate value.

4.3.2 Comparison between current and future emissions

The application of the EmiPro-RCP model will deliver emission scenarios over Portugal focused in 2020, 2050 and 2100, for all RCP scenarios. In this section a comparison between the current emissions for 2012 and the emissions results from the EmiPro-RCP model, for the selected years will be performed.

First a comparison for each pollutant (CO, PM2.5, PM10, SOx, NMVOC, NOx and NH_3) of annual total emissions (Gg) for the year 2012, with the emissions of 2020, 2050 and 2100 of each RCP scenarios (2.6, 4.5, 6 and 8.5) for the OECD90 region, was accomplished (Figure 4.11).

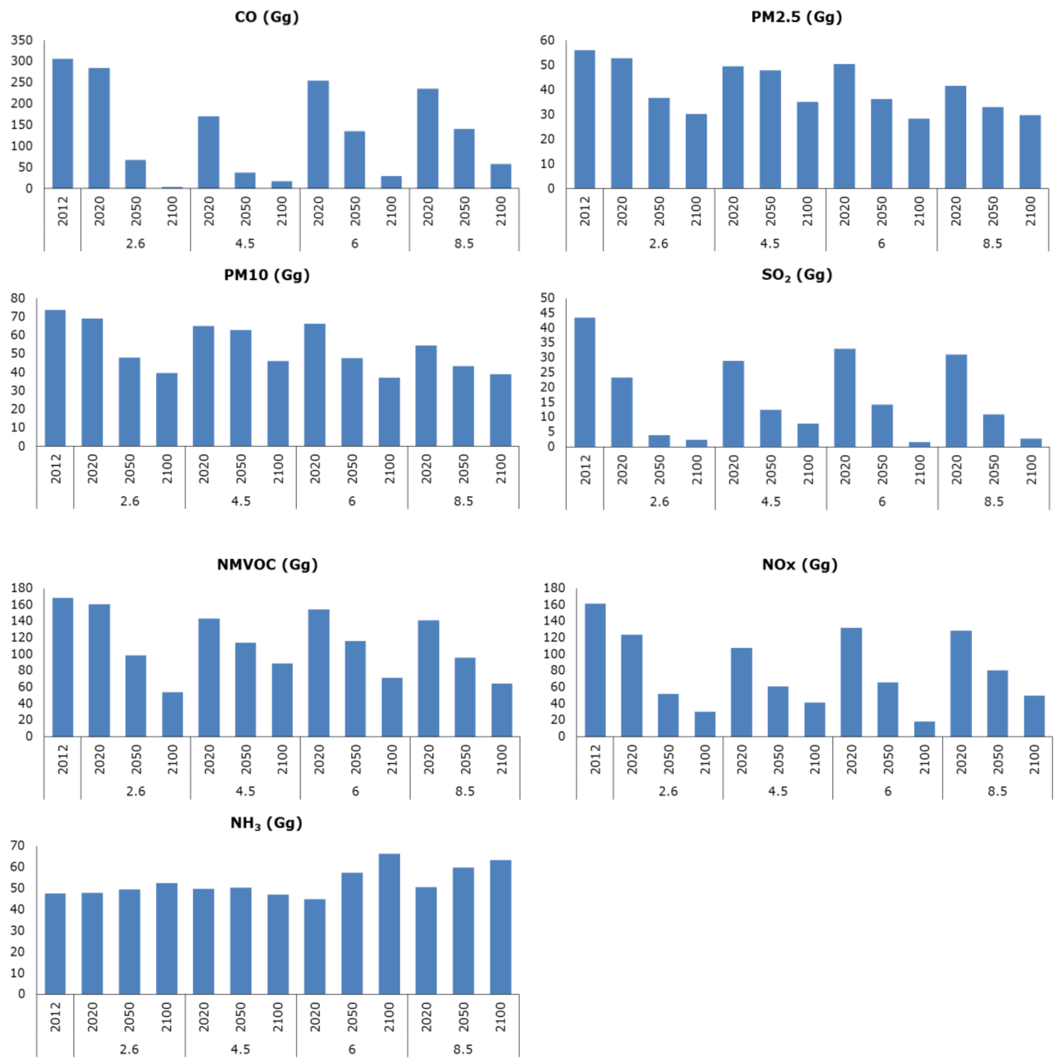


Figure 4.11– Comparison of 2012 total emissions (Gg) for CO, PM2.5, PM10, SOx, NMVOC, NOx and NH_3 with 2020, 2050 and 2100 for each RCP scenario.

For all the RCP scenarios all the pollutants emission values progressively decrease until 2100, with the exception of NH3 that will gradually increase for RCP 2.6, RCP6 and RCP 8.5. The change on the emission values between the year 2012 and the year 2020 for all the scenarios is higher in SOx when compared with the other pollutants. Also, for SOx, NOx and CO the decrease between the year 2020 and 2050 is higher than the difference between the year 2050 and the year 2100. On the other hand, for NMVOC, PM10 and PM2.5 the reduction is similar for all periods.

The EmiPro-RCP model also predicted the point emissions for each RCP scenario, an analysis of all the most important Portuguese point was performed for the same years and the same region analysed before, but just for the two most extremes scenarios (RCP2.6 and RCP8.5) (Figure 4.12).

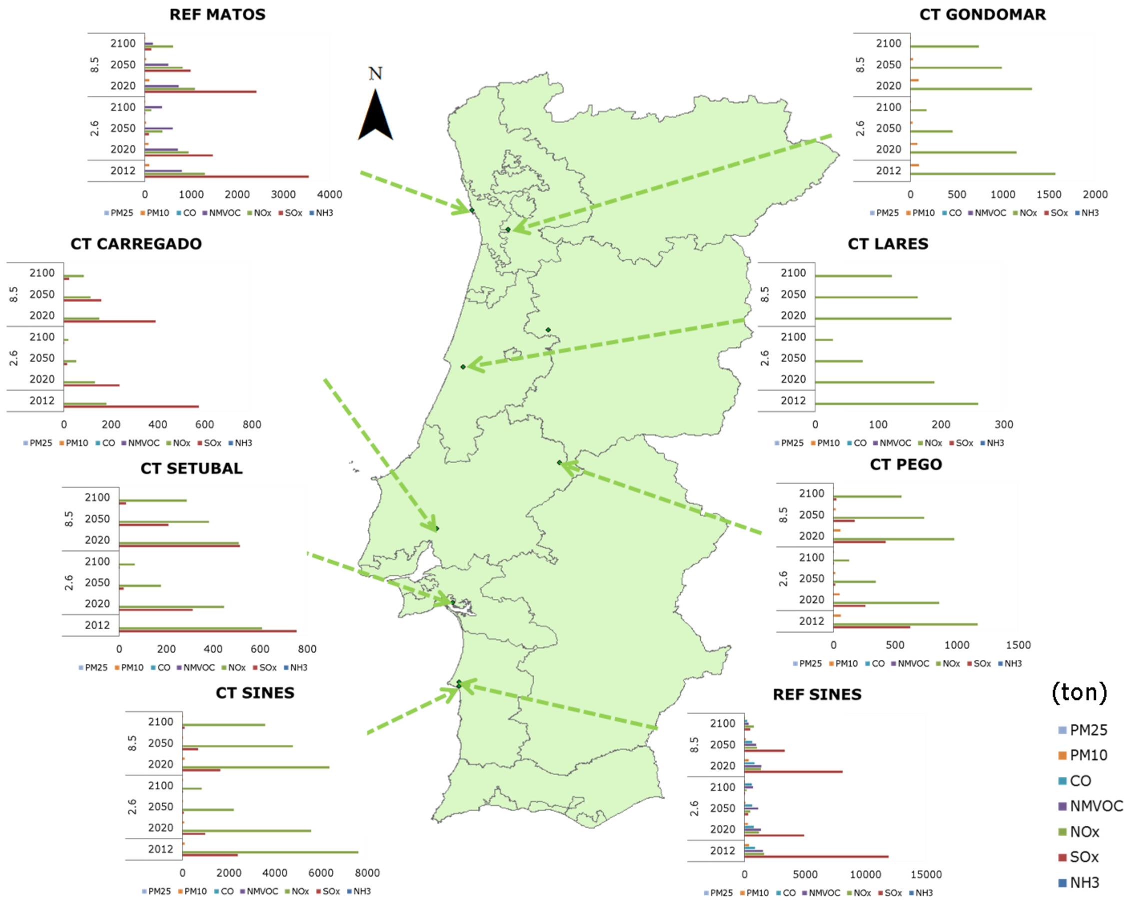


Figure 4.12– Comparison of 2012 point emissions (ton) for CO, PM2.5, PM10, SOx, NMVOC, NOx and NH_3 with 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

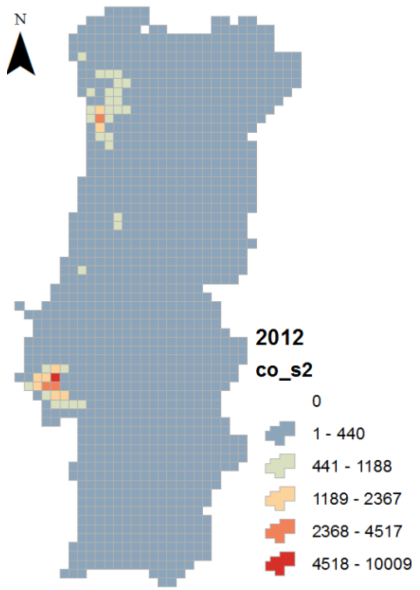
The pollutants considered in the national point sources are SOx, NOx, NMVOC, PM10 and CO, being the first two the ones with higher emission values. For all of these pollutants the decrease in the RCP2.6 scenario is higher than in the RCP8.5 scenario through the years until the year 2100, which is in agreement with the assumptions for each scenario.

Taking in consideration the SNAPs where each pollutant has the higher national emission values, Table 4.5 – Table 4.11 show the spatial distribution of the emissions for the years 2012, 2020, 2050 and 2100 for the RCP2.6 and the RCP8.5 scenarios (OECD90 region). The resolution of each grid cell is 9x9km2 and all the emissions are in ton/grid cell.

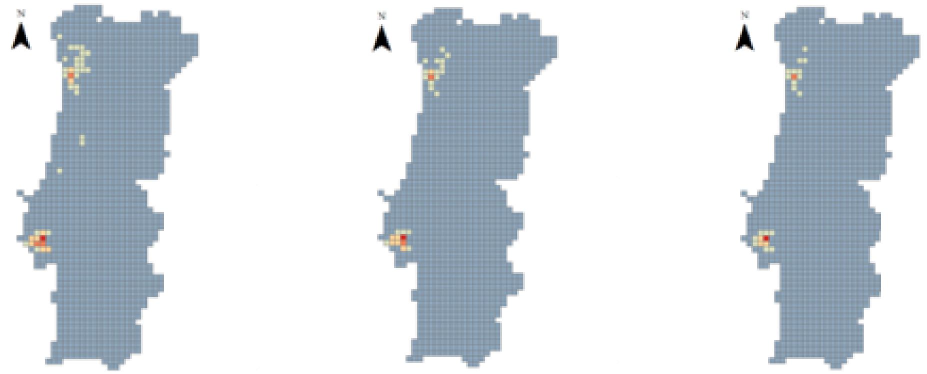
Table 4.5 - Comparison of 2012 SNAP emissions (ton) for CO with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

CO

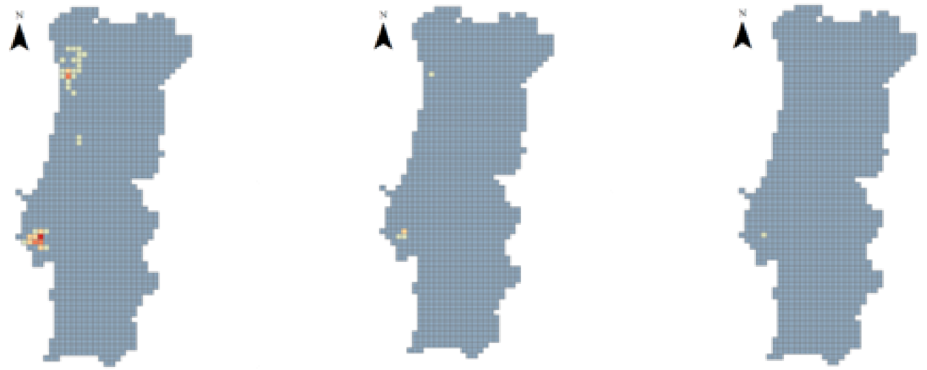
2012 | 2020 | 2050 | 2100



RCP2.6



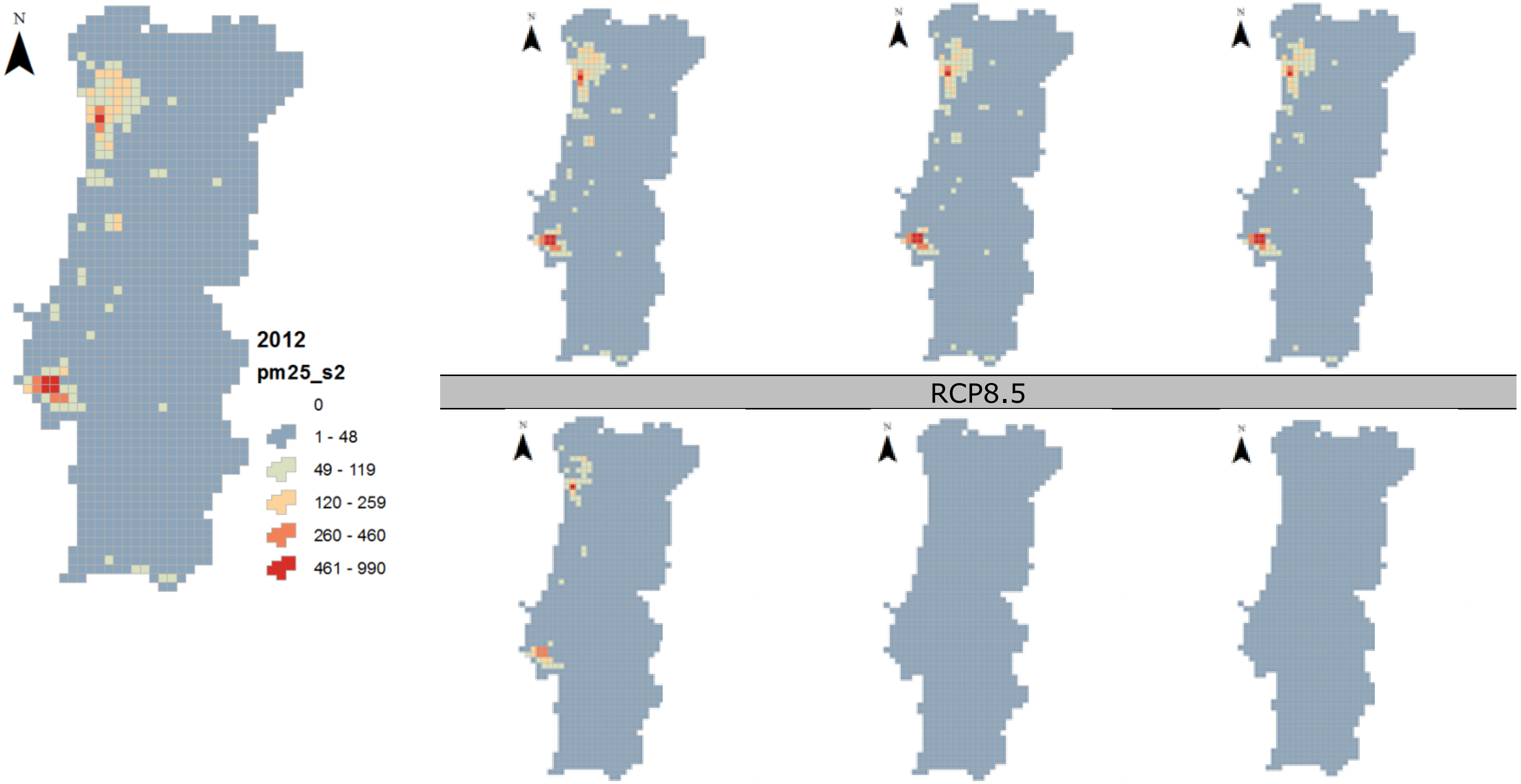
RCP8.5



Total emissions CO SNAP2 (ton)						
	RCP2.6	RCP8.5	RCP2.6	RCP8.5	RCP2.6	RCP8.5
2012	**2020**		**2050**		**2100**	
119 276	103 103	90 377	83 091	17 202	66 645	5 780

Table 4.6- Comparison of 2012 SNAP emissions (ton) for PM2.5 with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

PM2.5			
2012	2020	2050	2100
	RCP2.6		

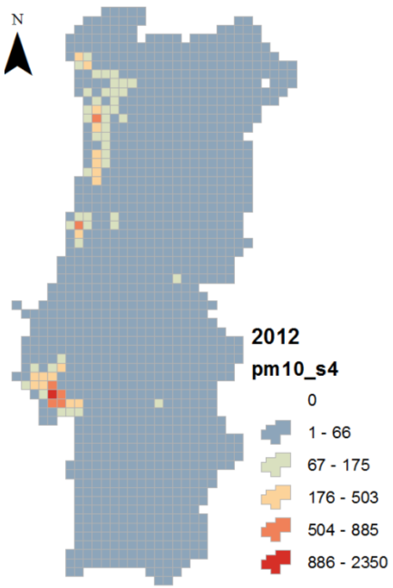


Total emissions PM2.5 SNAP2 (ton)						
	RCP2.6	**RCP8.5**	**RCP2.6**	**RCP8.5**	**RCP2.6**	**RCP8.5**
2012	**2020**		**2050**		**2100**	
24 286	23 091	11 814	20 290	517	18 518	209

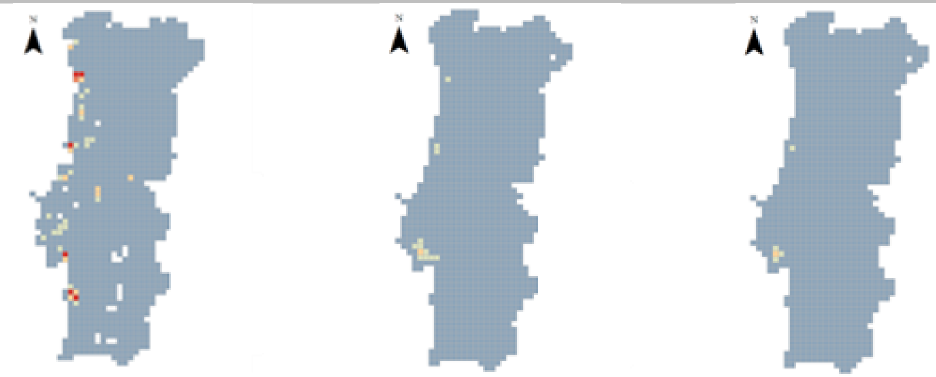
Table 4.7 - Comparison of 2012 SNAP emissions (ton) for PM10 with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

PM10

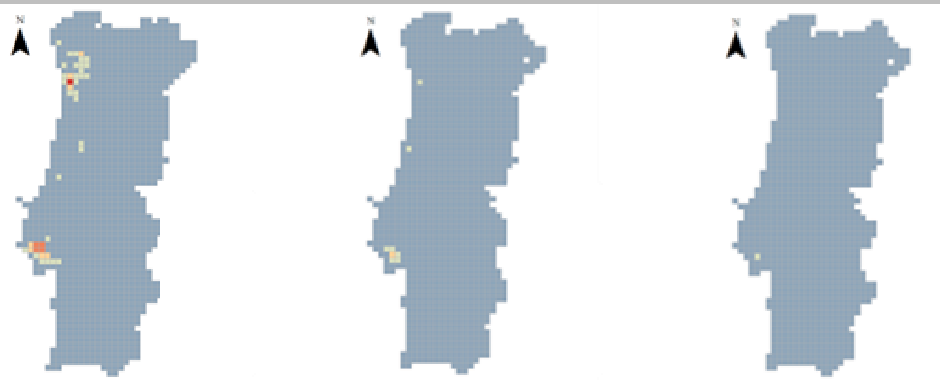
2012	2020	2050	2100



RCP2.6

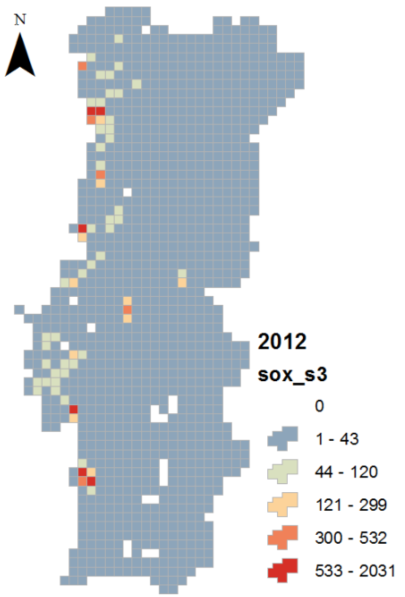


RCP8.5



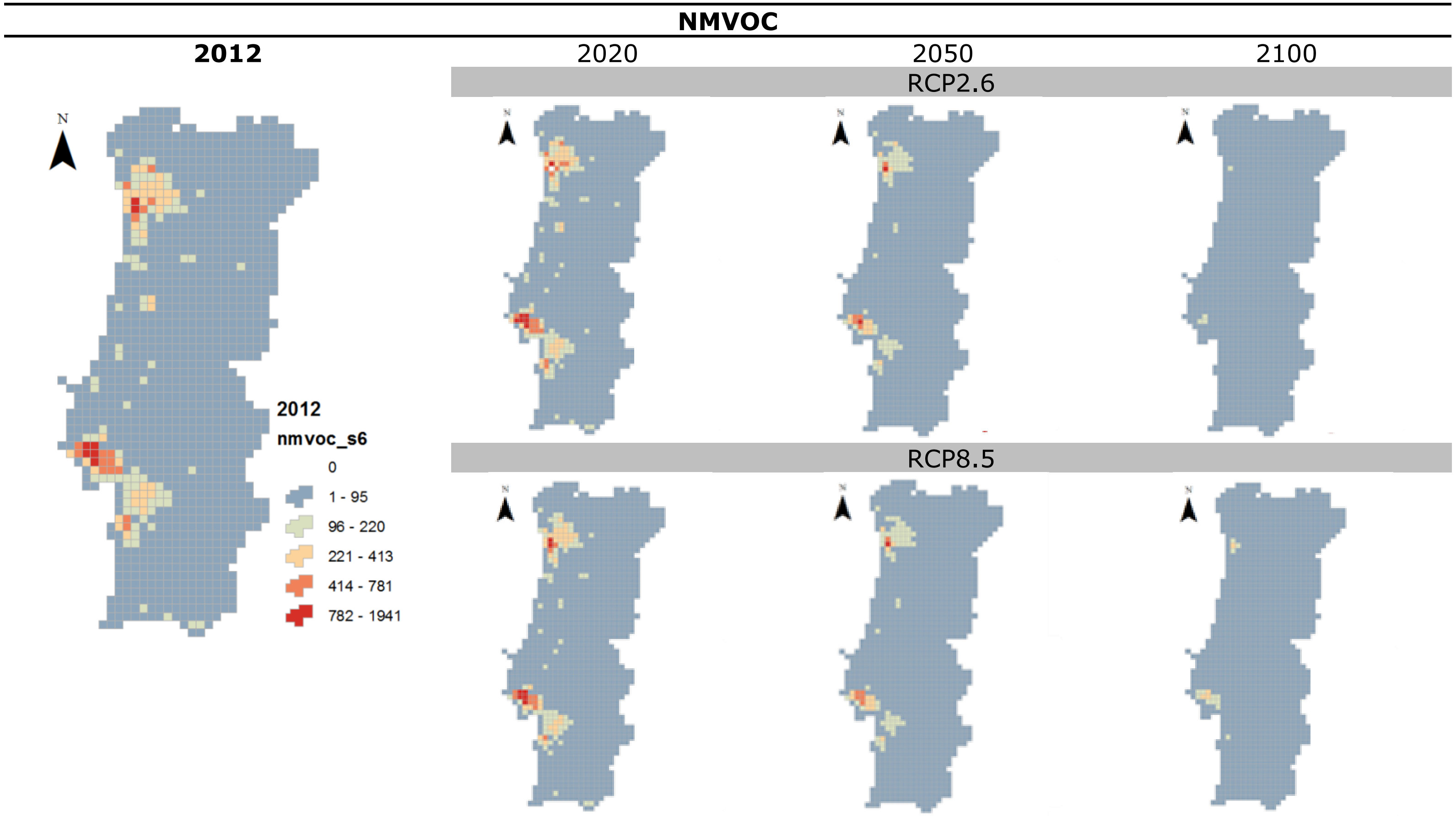
Total emissions PM10 SNAP4 (ton)						
	RCP2.6	**RCP8.5**	**RCP2.6**	**RCP8.5**	**RCP2.6**	**RCP8.5**
2012	**2020**		**2050**		**2100**	
26 680	19 737	14 603	4 613	3 721	2 855	783

Table 4.8 - Comparison of 2012 SNAP emissions (ton) for SOx with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

SOx			
2012	2020	2050	2100
	RCP2.6		
			
	RCP8.5		
			

Total emissions SOx SNAP3 (ton)						
	RCP2.6	**RCP8.5**	**RCP2.6**	**RCP8.5**	**RCP2.6**	**RCP8.5**
2012	**2020**		**2050**		**2100**	
20 066	13 676	13 846	3 110	3 119	2 201	608

Table 4.9 - Comparison of 2012 SNAP emissions (ton) for NMVOC with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

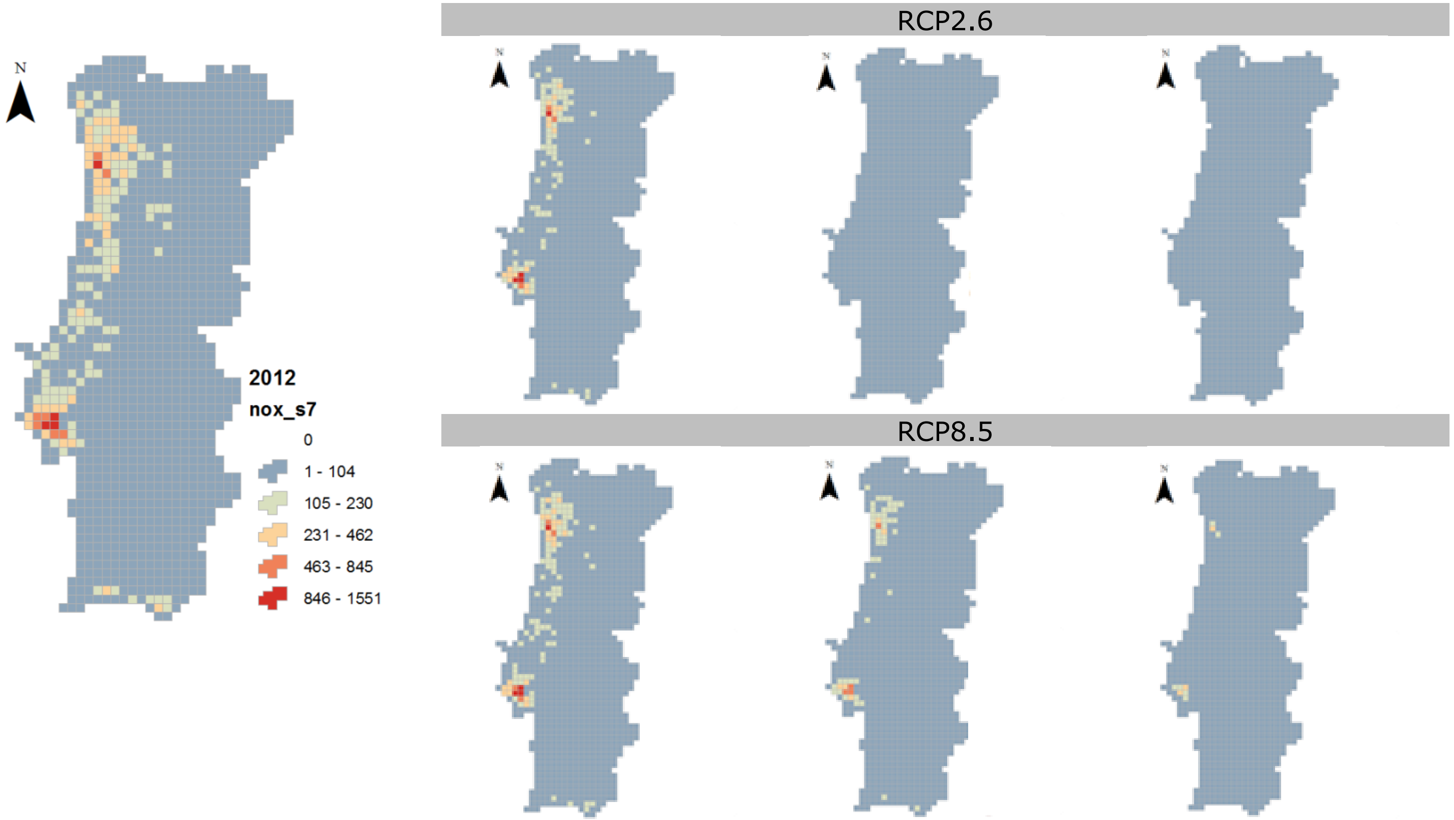


Total emissions NMVOC SNAP6 (ton)						
	RCP2.6	RCP8.5	RCP2.6	RCP8.5	RCP2.6	RCP8.5
2012	2020		2050		2100	
59 139	59 811	50 684	29 403	28 792	4 417	11 731

Table 4.10 - Comparison of 2012 SNAP emissions (ton) for NOx with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

NOx

2012	2020	2050	2100

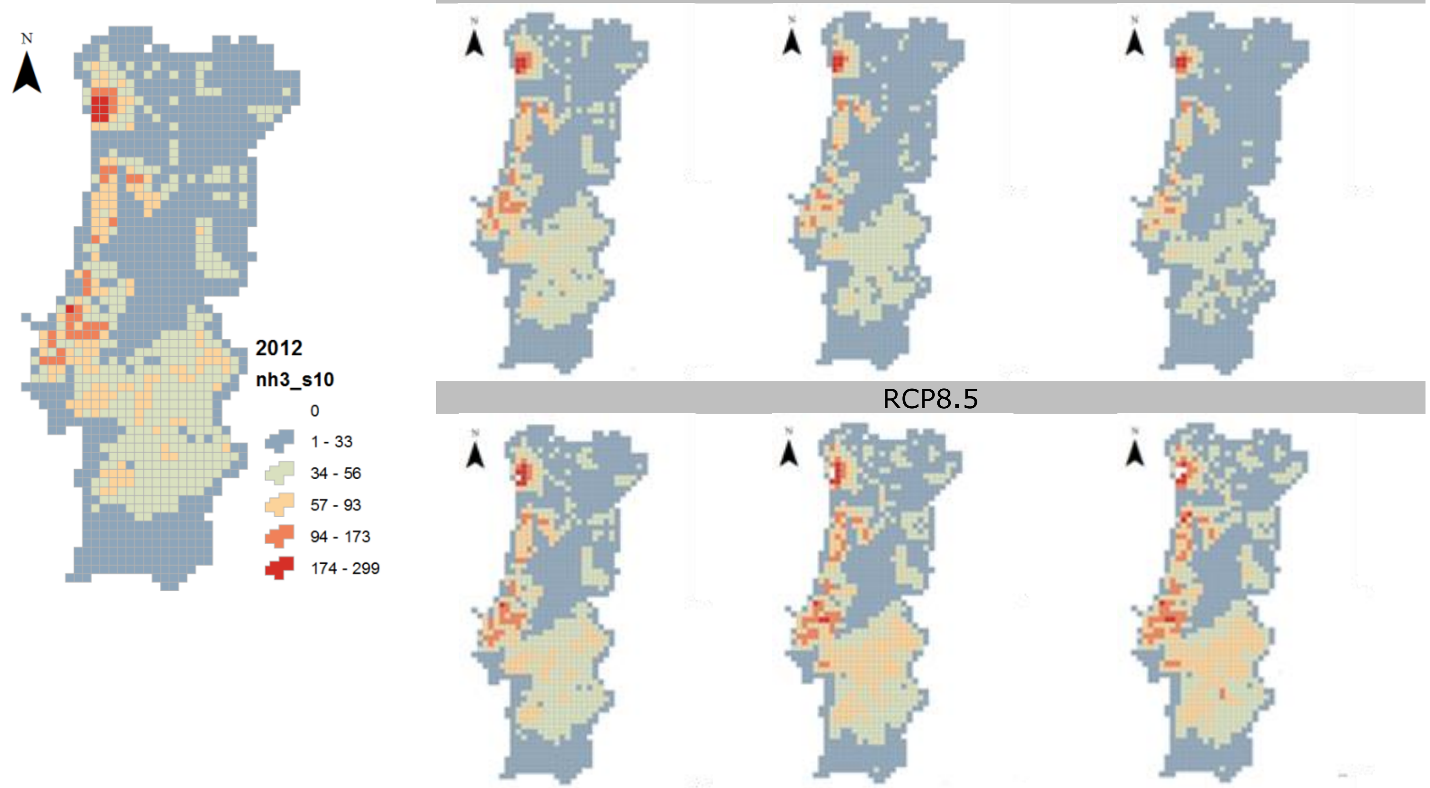


Total emissions NOx SNAP7 (ton)						
	RCP2.6	**RCP8.5**	**RCP2.6**	**RCP8.5**	**RCP2.6**	**RCP8.5**
2012	**2020**		**2050**		**2100**	
66 925	45 895	48 801	3 640	28 082	6	12 225

Table 4.11- Comparison of 2012 SNAP emissions (ton) for NH_3 with respectively 2020, 2050 and 2100 for RCP2.6 and RCP8.5 scenarios.

NH_3

2012	2020	2050	2100
	RCP2.6		



Total emissions NH_3 SNAP10 (ton)						
	RCP2.6	**RCP8.5**	**RCP2.6**	**RCP8.5**	**RCP2.6**	**RCP8.5**
2012	**2020**		**2050**		**2100**	
42 049	40 408	43 682	35 664	48 537	31 739	50 499

For CO, the RCP2.6 scenario shows a slightly progression of SNAP2 emissions. On the other hand, for the RCP8.5 scenario there is a significant reduction of the SNAP2 emissions. In both scenarios, the reduction is more evident in the urban areas of Porto and Lisbon, where the emission values are higher. This is in accordance with the values of the total emissions, which show a higher reduction in RCP8.5 than in RCP2.6.

The SNAP2 emissions of PM2.5 have the same behaviour as the CO emissions in both scenarios for the three years analysed. In the RCP8.5 since 2050 there is a significant reduction all over Portugal.

By analysing the spatial patterns of PM10, SOx and NMVOC emissions for SNAP4 (industrial processes), SNAP3 (industrial combustion) and SNAP6 (solvent use), respectively, it is possible to verify that most of the industries are located in the coastal zone of Portugal. Therefore, the higher differences between 2012 and the other years are in the coastal zone of Portugal, mainly in the Porto and Lisbon urban areas. For PM10 and SOx the emissions will be higher in 2100 with scenario RCP2.6 than with scenario RCP8.5. The PM10 values will be higher in the Lisbon urban area, than in the Porto urban area, and for SOx it will be the opposite. For RCP2.6 there is more difference in the emissions between 2020 and 2050 (76% for PM10, 77% for SOx) than between 2050 and 2100 (PM10 - 38%, SOx - 29%). On the other hand, for RCP8.5 the difference between 2020 and 2050 is almost the same between 2050 and 2100 (PM10 - 75%, SOx – 79%). In the case of NMVOC emissions, the highest values in 2100 will be found in RCP8.5 scenario and in the urban areas of Porto and Lisbon.

The 2012 emissions of NOx for SNAP7 are distributed along the coastal zone with higher values in the Porto and Lisbon urban areas. The reduction in RCP2.6 is extremely high, reaching almost zero in 2100 and in scenario RCP8.5 is more progressive through the years. Comparing the NOx emissions over the urban areas for both scenarios, they will be higher in scenario RCP8.5.

In terms of NH3 the emissions will increase in both scenarios through the years, being more evident for scenario RCP8.5. For this scenario there is an increase of the emission values in the coastal zone and in the southern inland of Portugal, which could be justified by the fact that the southern inland of Portugal is an intensive national rural zone.

For all the pollutants, the results obtained are consequence of the trend of the RCP emissions for each SNAP and for each pollutant.

4.4 Summary and Conclusions

The two main goals of this chapter are the estimation of current emissions for Portugal with a higher detailed disaggregation to improve the performance of the air quality simulations; and the estimation of projected emissions for Portugal based on the RCPs emissions scenarios in order to improve the results and keep the consistency of the climate change simulations.

In order to fulfil the first objective, a detailed methodology was applied to the national emission inventory, mainly to traffic sector, which allowed more accurate current emission estimations for Portugal. The results showed that the spatial distribution over Portugal of the SNAP7 emissions is improved when the traffic sector is considered as a line source. Therefore, there was a huge improvement of one of the inputs needed in air quality simulations and, consequently, in the accuracy of the results of the simulations.

The EmiPro-RCP model was developed to project the future emissions of GHG and common air pollutants, from the year 2020 until 2100. This model is only based on the RCP scenarios and does not consider socioeconomic and land-use changes of the study region, since there is no available data to characterize consistently the land-use changes in Portugal based on the RCPs scenarios. On the other hand, the fact that the model do not consider socioeconomic and land-use changes of the study region, allows keeping the initial trends defined in the RCP scenarios. As conclusion, the model permits a detailed analysis of the trends of the future national emissions based on the RCP scenarios.

By the application of EmiPro-RCP model, it was found that the emissions in Portugal tend to decrease in all the sectors, with the exception of NH_3 that tend to increase.

With this work an important and user-friendly tool was developed and can be used in upcoming studies, which focus on the climate change impacts on air quality. Also, it could be a helpful tool to assist decision-makers in analysing emission trends for the next decades.

Chapter 5

5 Air quality modelling sensitivity studies

The contents of this chapter were partially published in:

Sá, E., *Martins, H., Carvalho, A., Lopes, M., Miranda, A.I., Borrego, C. (2012), WRF-CAMx application over an urban area: parameterizations sensitivity study for air quality purposes. AIR QUALITY MANAGEMENT at URBAN, REGIONAL and GLOBAL SCALES, 4th International Symposium and IUAPPA Regional Conference, 10-13 September, Istanbul, Turkey. Eds: S. Incecik, C. Kahya. ISBN: 978-975-561-424-3.*

Coelho, M.C., Fontes, T., Bandeira, J., Pereira, S., Tchepel, O., ***Sá, E.,*** *Amorim, J., Borrego, C. (2014), Assessment of potential improvements on regional air quality modelling related with implementation of a detailed methodology for traffic emission estimation. Science of the Total Environment, 470–471, 127–137.*

Abstract

The main objective of this study is to evaluate the air quality modelling system WRF-CAMx over Portugal and Porto urban area. The performance of different combinations of physics parameterizations and a new land use (based in CLC2006) implemented in WRF was evaluated. Also, WRF-CAMx system was evaluated considering two different aspects: first, the CAMx flexi-nesting capability was assessed for meteorological variables; then, the impact of using two different emission data (from a top-down approach and from a combination of top-down and bottom-up approaches) were assessed. The obtained results were compared with available meteorological and air quality monitoring data. Results point out that the air quality system WRF-CAMx is an adequate tool for the analysis to be performed in the following chapter.

5.1 Introduction

In Portugal urban air pollution has become one of the main environmental concerns, namely in Porto, the second most important national urban area. PM10 annual and daily limit values and the NO_2 annual limit value have been surpassed in Porto urban area over the last decade (Borrego et al., 2012a, 2012b). Although, the analysis of monitoring data from the air quality stations of the Northern Region show that, as expected, ozone (O_3) concentrations are higher outside the urban centre of the region, O_3 information threshold is exceeded in the majority of the monitoring stations in Porto urban area, and often along a high number of hours per year. In this sense, together with PM10 and NO_2, O_3 is also considered a critical pollutant in the region, and also in Portugal.
The Directive on Ambient Air Quality and Cleaner Air for Europe (Directive 2008/50/EC) highlights that air quality models are a fundamental tool for the estimation of pollutants concentrations in the atmosphere, providing past, present and future estimations. Air quality models use mathematical and numerical techniques to simulate the physical and chemical processes that affect air pollutants as they disperse and react in the atmosphere. Based on inputs of meteorological data and source information like emission rates and stack height, these models are designed to characterize primary pollutants that are emitted directly into the atmosphere and, in some cases, secondary pollutants that are formed as a result of complex chemical reactions within the atmosphere. The most commonly used air quality models include three big groups of models: dispersion - usually used in the permitting process to estimate the concentration of pollutants at specified ground-level receptors surrounding an emissions source; photochemical - typically used in regulatory or policy assessments to simulate the impacts from all sources by estimating pollutant concentrations and deposition of both inert and chemically reactive pollutants over large spatial scales; and receptor – consists in observational techniques which use the chemical and physical characteristics of gases and particles measured at source and receptor to both identify the presence of and to quantify source contributions to receptor concentrations.

Photochemical are applied at multiple spatial scales from local, regional, national, and global. There are two types of photochemical air quality models commonly used in air quality assessments: the Lagrangian trajectory model that employs a moving frame of reference, and the Eulerian grid model that uses a fixed coordinate system with respect to the ground. Earlier generation modeling efforts often adopted the Lagrangian approach to simulate the pollutants formation because of its computational simplicity. The disadvantage of Lagrangian approach, however, is that the physical processes it can describe are somewhat incomplete. Most of the current operational photochemical air quality models have adopted the three-dimensional Eulerian grid modeling mainly because of its ability to characterize physical processes in the atmosphere in a comprehensive way and predict the species concentrations throughout the entire model domain. Nowadays there are several models that could simulate the chemistry of atmosphere at long-term and at regional scale. As example the American models CAM) and CMAQ (Tesche et al., 2006), the Australian model TAPM (Hurley et al., 2005) and the European models CHIMERE (Van Loon, 2004; Vautard et al., 2007) and LOTOS-EUROS (Schaap et al., 2008). In these air quality models, the chemical processes are treated independently of the meteorological model, i.e. "offline". This approach is computationally attractive when weather influences on the air chemistry is not the focus of a study, since it permits to carry sets of chemical transport simulations with a single meteorological dataset.

Air quality numerical modelling has become a fundamental tool on air quality management and meteorological models play an important role, since they provide representations of the weather processes for the air pollution simulations. The importance of meteorological inputs on regional air quality modelling has been clearly stated (e.g. Seaman, 2000) and consequently, the need to have a better insight on the sensitivity and performance of meteorological models. Parameterization of sub-grid scale phenomena persists as one of the most challenging problems in numerical modelling of the atmosphere.

The meteorological model selected to provide the meteorological fields required by the chemical-transport model, CAMx, is the WRF modelling system. The selection of this air quality modelling system (WRF-CAMx) is related with the fact that both models are freely available, and they have been extensively used and validated worldwide, being subject of constant improvement and update.
WRF model allows the application of different combination of physics parameterizations. Several studies (Borge, R. et al. 2008, Mercader, J. et al. 2010, García-Díez, M. et al., 2011) performed a comprehensive sensitivity analysis to WRF model for different regions and for different parameterizations, such as planetary boundary Layer, microphysics, land surface model and radiation; however an analysis of the WRF over the urban area of Porto has not been accomplished before. Moreover, WRF is a community model that is being increasingly used to study atmospheric dynamics and land-atmosphere interaction at various scales (Zhang et al. 2009, Rotunno et al. 2009, Catalano and Moeng 2010). One of the

most important inputs to the air quality models are the meteorological fields for all the domains. However, when the purpose is to study long-term periods, it becomes very expensive for the meteorological model to produce meteorological data for all the domains. In this sense, CAMx has the flexi-nesting capability, which is an algorithm that interpolates the missing fields from the parent grid (gridded surface emissions; land use and LAI distribution; height/pressure (defines the layer interface structure); horizontal wind components; temperatures; vertical diffusivities; water vapour; clouds and precipitation) into the other nesting grids. The use of this CAMx capability will reduce the time of simulation.

On the other hand, the complex process of atmospheric dispersion depends not only on topography, land use and meteorology, but also on the emissions (Seinfeld and Pandis, 2006). In the last decades, these data have been systematized allowing a general use of air quality models and an increasingly complex estimation of atmosphere concentrations (Zhang et al., 2012a; Zhang et al., 2012b). However, despite the complexity behind the development of air quality models, the accuracy and precision of their results are usually associated with the emission inventories used as input in those models (Taghavi et al., 2005). Atmospheric emissions inventories are usually quantified using one of two approaches: (i) top-down, based on the disaggregation process of total emissions from a certain area to smaller administrative units or a regular grid with higher resolution (Ossés de Eicker et al., 2008); and (ii) bottom-up, based on emissions estimation using detailed data of each emission source.

The top-down approach is very useful namely when local detailed information on main emissions generating activities is poor (Palacios et al., 2001). Tuia et al. (2007) verified that for a medium-sized city, the application of a top-down approach presents good results when compared with a bottom-up approach. Nevertheless, Wang et al. (2009) verified that the top-down approach overestimated the activities of Heavy Duty Vehicles (HDV) and Light Duty Vehicles (LDV) and underestimated the vehicle kilometres travelled for passenger car, taxi, shuttle bus, and bus. In fact, the top-down approach has some limitations. This is particularly true for the transportation sector in urban areas where this sector is responsible for 70% of emissions of local pollutants and 40% of carbon dioxide (CO_2) emissions (EC, 2007). Several studies with traffic emissions modelling using bottom-up approaches based on average speed models have been performed (such as Borge et al., 2012; Cai and Xie, 2011; Ho and Clappier, 2011; Wang et al., 2009). Wang et al. (2009) and Borrego et al. (2000) compared the top-down and bottom-up (using average speed models) approaches and verified that the top-down approach underestimated the emissions. In this sense, is relevant to assess how the different emission data provided by the different approaches could influence the air quality results.

The main aim of this chapter is to evaluate the air quality modelling system WRF-CAMx by performing: simulations using different combinations of physics parameterizations and a new land use (based in CLC2006); simulations using the flexi-nesting capability; and finally simulations using different emission data. A general model setup for Portugal will be defined to be used in air quality simulations in the next chapter.

5.2 Data and Methods

In all the different tests, the air quality modelling system, composed by the WRF and the CAMx, was applied to Portugal and Porto urban area. Both models are well-known and have been applied by several authors (Soares et al., 2012; Solazzo et al., 2012; Monteiro et al., 2013) however a detailed analysis of the WRF and CAMx model, over Portugal and Porto urban area, has not been accomplished before. The detailed analysis will focus on the modelling system configuration, options and inputs.

In the following section the modelling system (5.2.1), the input data (5.2.2) and the different methodologies (5.2.3) are presented.

5.2.1 The modelling system

5.2.1.1 WRF

WRF is a next-generation mesoscale numerical weather prediction system designed to serve both operational forecasting and atmospheric research needs. A detailed description of this model can be found on Skamarock et al. (2008). In Figure 5.1 is described the structure of WRF model. The first module, named WPS, aim to prepare input to WRF for real data simulations and include three programs: geogrid, ungrib and metgrid. The geogrid program defines the map projection, the geographic location and dimension of the domains. Also, interpolates static terrestrial data to each grid point of the 2D domain. The ungrib program extracts meteorological fields from the Global data, based on the information from the Vtable. The last program of the WPS, horizontally interpolate meteorological data (extracted by ungrib) to simulations domains (defined by metgrid). The outputs from this last program are used as input in the real program inside the WRF module. Additionally, to these inputs it is necessary to edit the namelist file, where all the inputs, parameterizations and outputs are defined. The real program provides the initial conditions files, which are used as inputs in wrf program. WRF module generates meteorological fields, which are one of the inputs required to the air quality model simulations.

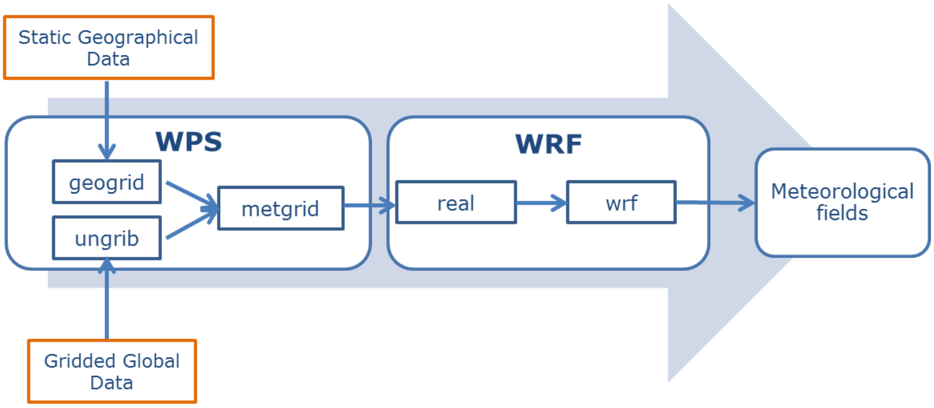


Figure 5.1 – WRF model structure.

The vertical structure of the WRF model includes 27 layers covering the whole troposphere. Topography, land use and land-water masks datasets were interpolated with the appropriate spatial resolution considering each domain.

5.2.1.2 CAMx

CAMx is an Eulerian photochemical dispersion model that considers the emission, dispersion, chemical reaction, and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species on a system of nested three-dimensional grids (Morris et al., 2004). In Figure 5.2 is described the structure of CAMx v5.2 model.

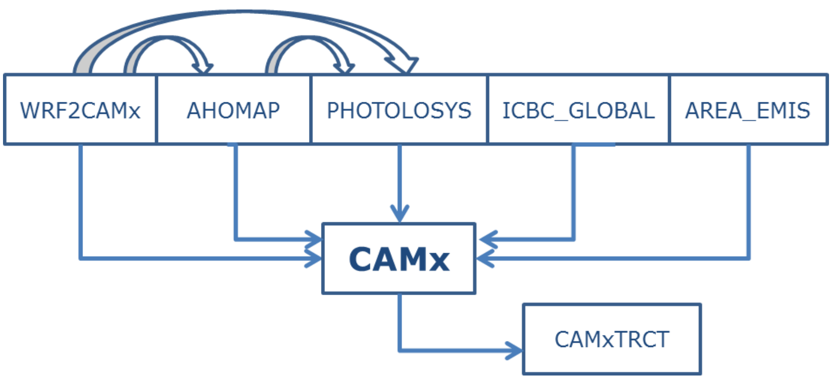


Figure 5.2 – CAMx v.5.2 model structure.

CAMx has five pre-processors: WRF2CAMx, AHOMAP. PHOTOLOSYS, ICBC_GLOBAL and AREA_EMIS. The WRF2CAMx provides CAMx meteorological input files from the WRF model. The land use grid together with ozone column data files from Ozone Monitoring Instrument (OMI)/AURA (URL5) constitutes the input for AHOMAP, which generates the albedo, haze and ozone column input files for PHOTOLOSYS. This pre-processor determines the photolosys rates for each grid cell. The ICBC_GLOBAL gives the initial and boundary conditions, based on the outputs from GOCART and INCA global models. At last, the AREA_EMIS prepares the emissions for all the domains. All, of these pre-processors generate inputs to the CAMx module and the outputs from this module could be processed by the pos-processor CAMxTRCT in order to help in the analysis of the results.

CAMx vertical structure includes 15 layers and in terms of chemical mechanism, the gas-phase photochemistry was resolved through the Carbon Bond (CB4) (Gery et al., 1989) and the model also contains detailed algorithms for the relevant processes, including aqueous chemistry (RADM-AQ), inorganic aerosol thermodynamics/partitioning (ISORROPIA), and secondary organic aerosol formation/partitioning (SOAP). Initial and boundary conditions for both gases and particulate species were, respectively, driven by the LMDZ-INCA global model (Hauglustaine et al., 2004) (for CO, PAN and O_3) and the GOCART global model (Ginoux et al., 2001) (for particulate sulphate (PSO_4), sodium (PNa) and chloride (PCl)).

5.2.1 Input Data

5.2.1.1 Meteorological data

The WRF simulations were driven by the ECMWF global analysis with 1° x 1° spatial resolution and temporal resolution of 6 h for surface and pressure levels. The data is collected from the website *http://data-portal.ecmwf.int/data/d/interim_daily/*, and the data is available from 1979, 1st January until nowadays. For each test in the website the parameters required to run WRF model were selected, taking in consideration each simulation period.

5.2.1.2 Emissions

A top-down methodology was applied to disaggregate the anthropogenic emissions, using the Portuguese national emission inventory (INERPA) (PEA, 2011). This inventory provides quantitative information for the main atmospheric pollutants: CO, CH_4, NOx, N_2O, SOx, NMVOC, NH_3, PM10 and PM2.5. The total national

emissions with NUT IV (municipalities) resolution are reported for 11 main source categories (SNAP) (Table 4.2).
The annual national emissions from anthropogenic area sources for each pollutant and activity sector are spatially disaggregated from the municipality level to the spatial resolution required for the selected simulation domains. The data are then converted to a regular grid, using GIS, for all the domains considered in order to be used as inputs to air quality modelling. The emissions year chosen for each test was selected taking in consideration the year of simulation.

In one of the test described in this chapter (Impacts on air quality by changing emissions), the emissions considered to perform the simulation also take in account emissions estimated by a bottom-up approach for four air pollutants: PM, NO_x, CO and HC. These emissions were estimated by TEMA team of University of Aveiro for 585 km of roads considered in the study domain. A more detailed description of these procedures can be found in Coelho et al. (2014).

5.2.2 Methodology

5.2.2.1 WRF parameterizations sensitivity study

In order to evaluate the impacts on air quality of different combinations of physics parameterizations and new land use in WRF, the WRF-CAMx modelling was applied to the domains shown in Figure 5.3. The meteorological simulation used the two-way nesting technique in the WRF model for three domains (Figure 5.3a): D1 with 25 km spatial resolution, centred at 41° N, 4°W. It covers the Northern part of Africa and an area of the Atlantic Ocean including the Azores and Canary Islands, and is intended to capture synoptic features and general circulation patterns. The first nested domain (D2), with a spatial resolution of 5 km, comprises Portugal and the innermost domain (D3) is centred over the Porto urban area and consists of 61 columns and 66 rows of 1 x 1 km^2 grid cells.
CAMx was applied for two domains (Figure 5.3b): D’1 with a spatial resolution of 5 km covering Portugal and the innermost domain (D’2) with a spatial resolution of 1 km over the Porto urban area. These domains are smaller than the WRF domains D2 and D3, respectively.

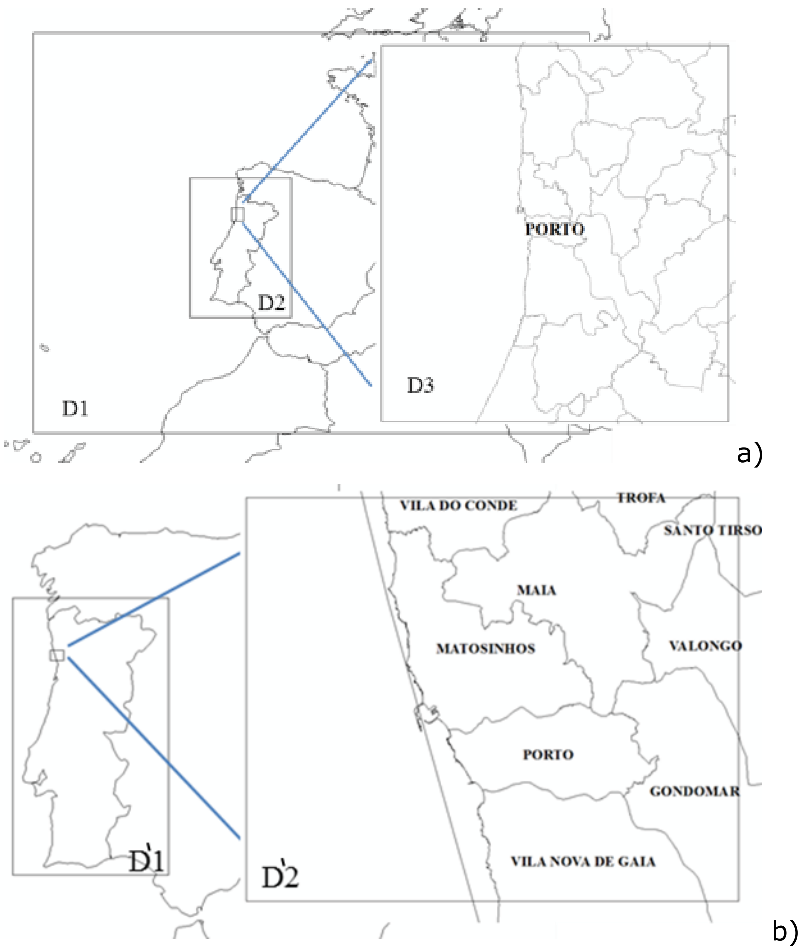


Figure 5.3 - WRF (a) and CAMx (b) simulation domains.

Simulations were conducted for the month of December of the year 2010. The first six hours of the period were considered as spin-up, while the remaining days are used for the analyses, i.e. 744 h per episode. The selection of the temporal domain to develop the sensitivity runs is based on the analysis of air pollution records across the area of interest (Porto, Matosinhos and Maia municipalities – D2 CAMx) in the air quality stations: urban - Antas (ANT), Ermesinde (ERM), Senhora da Hora (SHOR) and Vermoim (VER); and suburban - Vila Nova da Telha (VNT) and Custóias (CUS). Generalized high concentration values of PM10 and NO_2 were observed in Porto urban area during the winter period (Figure 5.4).

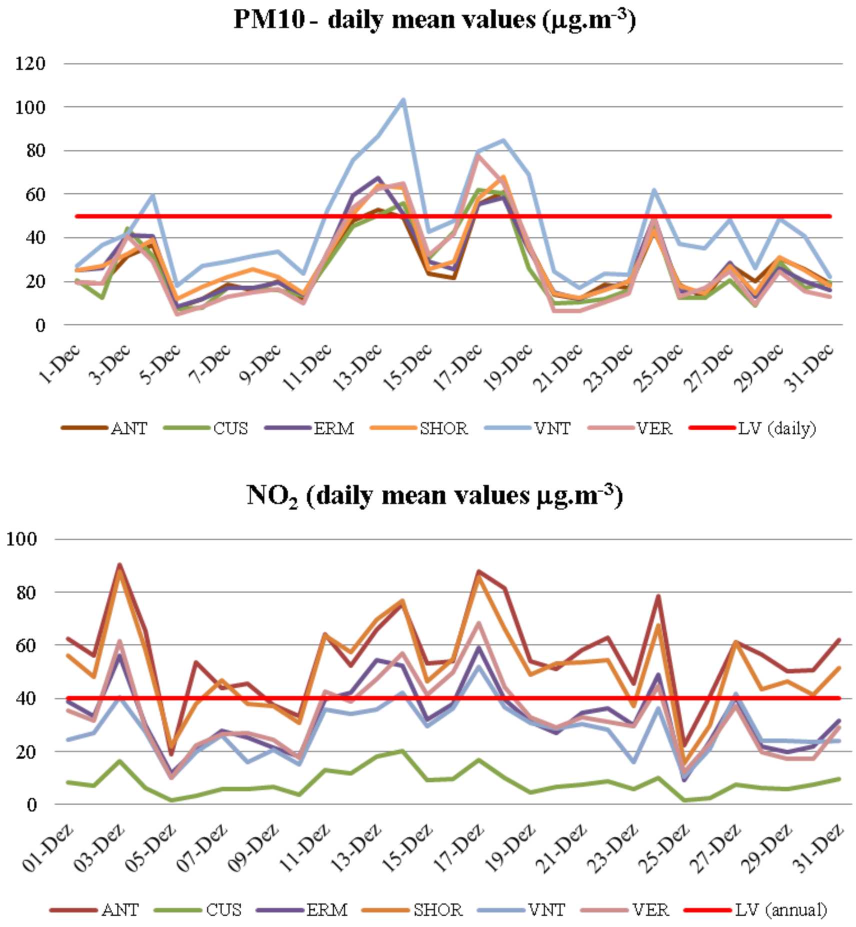


Figure 5.4 - PM10 and NO_2 concentrations over the Porto urban area (December 2010).

WRF model has the possibility to choose the physics parameterizations for each simulation and in this work is intend to performed a sensitive analyses of the different options for land-surface models (LSMs) and solar radiation for the study area and its impacts on air quality simulations.

LSMs combine atmospheric information from the surface layer scheme with land surface properties (dependent on land uses) to evaluate the vertical transport done in the PBL schemes, which has a direct influence on the estimation of the PBL height (PBLH) (Han et al., 2008). The three options currently available in WRF are: 5-layer thermal diffusion LSM (Dudhia, 1996), Noah LSM (Chen and Dudhia, 2001) and Rapid Update Cycle (RUC) Model (Smirnova et al., 2000), and only the first two were tested.

Solar radiation is the primary driver to PBL dynamics. The radiation schemes in WRF provide atmospheric heating due to radiative flux divergence and surface downward longwave (LW) and shortwave (SW) radiation for the ground heat budget. The LW schemes available in WRF V3.3.1 are the Rapid Radiative Transfer Model (RRTM) (Mlawer et al., 1997), the Eta Geophysical Fluid Dynamics Laboratory (GFDL) scheme (Fels and Schwarzkopf, 1981), the NCAR Community Atmospheric

Model (CAM) scheme (Collins et al., 2004), the Rapid Radiative Transfer Model (RRTMG) scheme (Iacono et al., 2008), the new Goddard scheme (Chou and Suarez, 2001).

The schemes included to represent SW processes are the Dudhia scheme (Dudhia, 1989), the Goddard scheme (Chou and Suarez, 1994), the NCAR Community Atmospheric Model (CAM) scheme (Collins et al., 2004), the Rapid Radiative Transfer Model (RRTMG) scheme (Iacono et al., 2008), the new Goddard scheme (Chou and Suarez, 2001) and the Eta Geophysical Fluid Dynamics Laboratory (GFDL) scheme (Fels and Schwarzkopf, 1981).

Additionally, WRF has the possibility to choose different land use/land cover classification: USGS 24-category and IGBP-Modified 20-category or also introduce new input data set. So, a Corine Land Cover 2006 (CLC2006) dataset for Portugal and, more specifically for the Porto urban area was introduced as input data set.

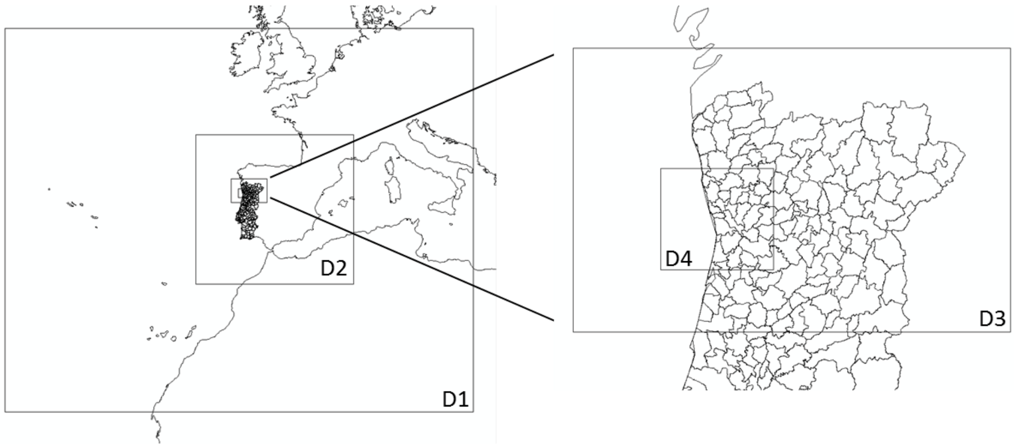
A combination of the model simulations was performed has described in Table 5.1.

Table 5.1 - WRF options considered in each WRF simulation.

Test	Microphysics	LW radiation	SW radiation	PBL scheme	Cumulus scheme	Land surface model	Land use input data set
1	WSM5	CAM	CAM	YSU	Kain-Fritsch	NOAH	USGS-24
2	WSM5	RRTMG	RRTMG	YSU	Kain-Fritsch	NOAH	USGS-24
3	WSM5	RRTMG	RRTMG	YSU	Kain-Fritsch	5-layer	CLC06

5.2.2.2 Assessment of CAMx flexi-nesting capability

In order to assess the CAMx flexi-nesting capability, two WRF-CAMx simulations were performed over the study domain. The domains selected for this analysis are represented in Figure 5.5. There were four WRF domains: D1 with a 27 km spatial resolution, D2 with a 9 km spatial resolution, D3 with a 3 km spatial resolution and D4 with a 1 km spatial resolution. For the air quality simulation there were only three domains: D1 with a 9 km spatial resolution, D2 with a 3 km spatial resolution and D3 with a 1 km spatial resolution.



a)

b)

Figure 5.5 - WRF (a) and CAMx (b) domains.

The first WRF-CAMx simulation (designated as N) was conducted using for all the CAMx domains the input from the WRF model, i.e, CAMx uses the meteorological fields from WRF D2, D3 and D4 for CAMx D'1, D'2 and D'3, respectively. In the other simulation (designated as FN) the only considered meteorological field was for the parent grid (CAMx D'1), the meteorological fields for the nested CAMx domains were obtained by the algorithm Flexi-nest.

5.2.2.3 Impacts on air quality by changing emissions

In order to evaluate the impacts on air quality by changing emissions, two emissions scenarios are considered as input in CAMx. S1 includes the emissions from top-down approach (INERPA emissions of 2009) and S2 includes the emissions resulting from integration of top-down and bottom-up approaches. Emissions obtained from bottom-up approach provide detailed information on spatial variability of the data. However, their use in air quality modelling is limited due to incompleteness of emission data since there is a significant lack of source-specific information. On the other hand, top-down methodology may provide better overview on the data in terms of total emissions but their spatial allocation is an important source of uncertainty.

In this context, a harmonized methodology was developed in this study in order to analyse the difference between bottom-up and top-down approaches to traffic emissions estimation and possible improvements in air quality modelling. For this purpose, the regular grid of bottom-up traffic emissions was spatially integrated with the top-down emissions on a horizontal resolution of 1x1 km2 (Figure 5.6).

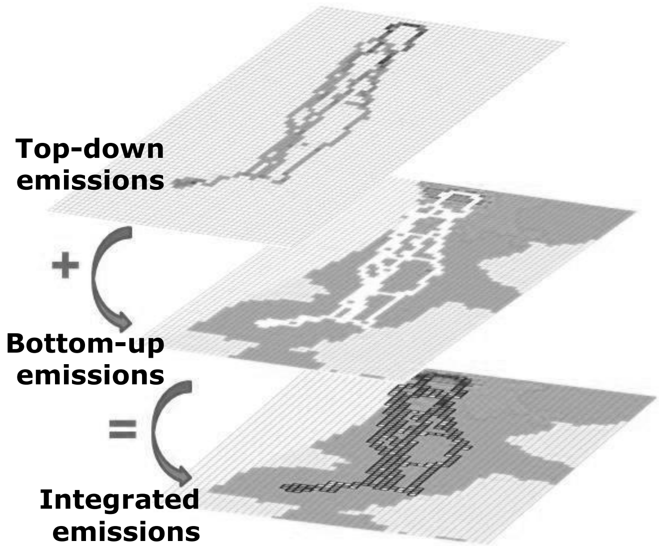


Figure 5.6 - Schematic representation of the conceptual approach to integrate traffic emissions.

The bottom-up emissions consider only the cells values corresponding to the roads selected in this research. Thus, in order to consider the local variation of traffic emissions obtained by bottom-up approach and emissions obtained from other road segments of the study domain, the top-down emissions are combined with the bottom-up emissions. Based on this approach a regular grid with traffic emissions was obtained and used as an input for air quality modelling. Simulations were conducted for March 2011 data.

The WRF-CAMx modelling domains are shown in Figure 5.7. For the meteorological simulation the two-way nesting technique has been used in the WRF model for three domains (Figure 5.7a): D1 with 25 km spatial resolution, centered at 41° N, 4°W. It covers the Northern part of Africa and an area of the Atlantic Ocean including the Azores and Canary Islands, and is intended to capture synoptic features and general circulation patterns. The first nested domain (D2), with a spatial resolution of 5 km, comprises Portugal and the innermost domain (D3) is centred over the selected study domain and consists of 73 columns and 118 rows of 1 x 1 km^2 grid cells.

CAMx was applied for two domains (Figure 5.7b): D’1 with a spatial resolution of 5 km covering Portugal and the innermost domain (D’2) with a spatial resolution of 1 km over the study domain. These domains are smaller than the WRF domains D2 and D3, respectively.

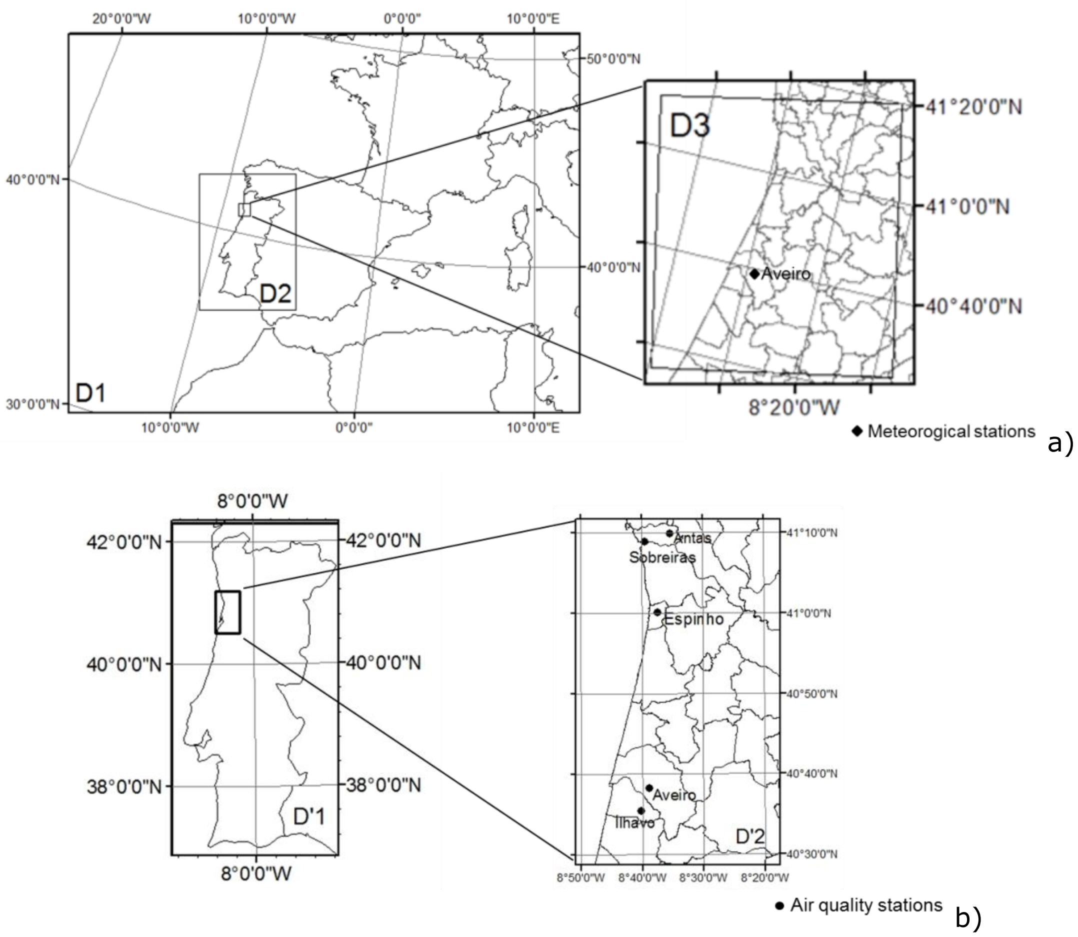


Figure 5.7 - Simulation domains for WRF (a) and CAMx (b).

5.3 Results and Discussion

5.3.1 WRF parameterizations sensitivity study

To evaluate the air quality modelling system results, a comparison with measured data in the selected meteorological and air quality stations was performed, using quality indicators (Table 5.2): BIAS and r. BIAS indicates if the model follows the variations of the observed values: if positive, the model overestimates the results, and if negative, the model underestimates the results. A good correlation between model results and measurements is considered when the Pearson Correlation Coefficient (r) value is nearly 1.

Table 5.2- Quality indicators applied in the WRF-CAMx validation.

Indicator	BIAS	Pearson Correlation Coefficient (r)
Equation	$BIAS = \frac{\sum_{i=1}^{N}(Oi - Pi)}{N}$	$r = \frac{N\left(\sum_{i=1}^{N} O_i P_i\right) - \left(\sum_{i=1}^{N} O_i\right)\left(\sum_{i=1}^{N} P_i\right)}{\sqrt{\left[N\left(\sum_{i=1}^{N} O_i^2\right) - \left(\sum_{i=1}^{N} O_i\right)^2\right]\left[N\left(\sum_{i=1}^{N} P_i^2\right) - \left(\sum_{i=1}^{N} P_i\right)^2\right]}}$
Ideal value	0	1

O_i – observed values, P_i – predicted values; N – Number of values

Statistical results from the evaluation of WRF outputs for each test, are presented in this section (Table 5.3). The meteorological stations selected are Aveiro (AVR), Porto (POR), Lisboa (LIS), all situated in Atlantic coastal zone of Portugal and Viseu (VIS) that is approximately 50 km east of the Atlantic ocean, located in Portugal northern-centre.

WRF application with both LW and SW radiation scheme (CAM and RRTMG) shows similar model performance for temperature, wind speed and wind direction. The use of CLC06 as input land use data set improve the model performance, however not significantly, therefore the UGS24 data set represent well Portugal land use, at least for WRF applications at this resolution (> 1 km).

Table 5.3 - WRF Statistical analysis for T (ºC), WSPEED ($m.s^{-1}$) and WDIR (º).

	Statistic		BIAS			r		
	Test		1	2	3	1	2	3
	Variable				T			
Monitoring Network	AVR	D1	0.68	0.23	0.07	0.85	0.85	0.92
		D2	0.69	0.26	-0.05	0.84	0.84	0.89
	POR	D1	-2.04	-2.33	-2.13	0.91	0.91	0.93
		D2	0.03	-0.45	-0.85	0.88	0.88	0.91
		D3	0.01	-0.45	-0.95	0.87	0.88	0.91
	LIS	D1	0.46	0.28	0.27	0.84	0.82	0.82
		D2	-0.03	-0.62	0.1	0.91	0.89	0.86
	VIS	D1	-0.58	-0.92	-1.43	0.83	0.83	0.85
		D2	0.2	-0.07	-0.49	0.85	0.84	0.87
	Variable				WSPEED			
Monitoring Network	AVR	D1	-3.03	-2.87	-2.53	0.56	0.57	0.59
		D2	-2.93	-2.83	-2.12	0.57	0.58	0.54
	POR	D1	-5.52	-5.48	-5.11	0.45	0.4	0.42
		D2	-4.22	-4.22	-2.59	0.68	0.62	0.65
		D3	-4.24	-4.28	-2.58	0.65	0.6	0.61
	LIS	D1	-3.1	-3.05	-2.28	0.61	0.6	0.63
		D2	-0.92	-1.05	-0.18	0.67	0.65	0.7
	VIS	D1	0.15	0.15	-0.21	0.65	0.65	0.67
		D2	-0.09	-0.09	-0.23	0.72	0.72	0.75
	Variable				WDIR			
Monitoring Network	AVR	D1	-14.55	-17.78	-9.99	0.12	0.19	0.15
		D2	-20.99	-23.31	-11.13	0.28	0.25	0.23
	POR	D1	-94.89	-94.32	-92.18	0.24	0.16	0.17
		D2	-92.89	-94.16	-89.96	0.16	0.16	0.17
		D3	-92.71	-94.67	-88.68	0.17	0.16	0.15
	LIS	D1	-25.69	-27.77	-27.73	0.57	0.54	0.62
		D2	-21.21	-24.38	-25.68	0.61	0.6	0.69
	VIS	D1	-15.84	-15.84	-12.96	0.5	0.5	0.53
		D2	-16.9	-16.9	-12.27	0.54	0.54	0.52

CAMx results of the three tests were compared with air quality monitoring data for PM10 and NO_2 at the Porto urban area (Table 5.4).

Table 5.4 - CAMx statistical analysis for PM10 and NO_2 concentration levels ($\mu g.m^{-3}$).

	Statistic		BIAS			r		
	Test		1	2	3	1	2	3
	Variable				**PM10**			
Monitoring Network	ANT	D1	8.13	12.89	7.88	0.21	0.38	0.43
		D2	11.83	10.73	6.74	0.3	0.16	0.37
	CUS	D1	4.43	11.33	5.51	0.09	0.36	0.56
		D2	9.94	9.04	5.7	0.27	0.26	0.48
	ERM	D1	11.13	19.16	15.43	0.07	0.27	0.42
		D2	18.76	18.5	15.78	0.22	0.17	0.41
	SHOR	D1	9	14.71	9.76	0.12	0.36	0.44
		D2	12.56	12.27	9.3	0.23	0.2	0.4
	VNT	D1	24.55	34.11	28.7	0	0.25	0.27
		D2	33.12	33.72	29.5	0.11	0.21	0.26
	VER	D1	6.79	16.58	11.33	-0.01	0.28	0.48
		D2	15.49	15.44	11.89	0.17	0.23	0.43
	Variable				**NO_2**			
Monitoring Network	ANT	D1	28.42	28.33	-5.5	0.6	0.6	0.27
		D2	28.93	24.54	-7.87	0.6	0.47	0.23
	CUS	D1	-25.45	-25.76	-16.17	0.42	0.57	0.45
		D2	-29.52	-31.31	-19.21	0.5	0.44	0.39
	ERM	D1	12.58	15.38	6.23	0.55	0.5	0.18
		D2	16.69	14.84	7.3	0.48	0.42	0.14
	SHOR	D1	20.56	20.15	-8.75	0.58	0.66	0.28
		D2	17.9	14.88	-12.09	0.6	0.48	0.24
	VNT	D1	-1.53	2.16	14.06	0.45	0.48	0.12
		D2	3.28	3.44	15.27	0.43	0.45	0.09
	VER	D1	3.35	8.54	-3.58	0.43	0.43	0.28
		D2	8.43	8.2	-3.22	0.44	0.47	0.21

On the contrary of WRF, CAMx statistical analysis shows different model performances for each test and for each pollutant. For PM10 test 3 shows better correlations for all the air quality stations, however, in terms of NO_2, this test shows weak correlations. Test 1 and test 2 demonstrate similar behaviour for NO_2, having correlations ranging between 0.4 and 0.7. On the other hand for PM10 test 2 shows significantly better results than test 1.

5.3.2 Assessment of CAMx flexi-nesting capability

The results from both simulations were statistical and spatial analysed. The air quality results were compared with the measured data from the selected air quality stations. The statistical analysis was performed, using quality indicators such as Correlation Coefficient (*r*), systematic error (BIAS) and Root Mean Square Error (RMSE). Table 5.5 shows the mean value of the statistical indicators for all the air quality stations for O_3, PM10 and NO_2, obtained in both simulations (N and FN).

Table 5.5 - CAMx statistical analysis for O_3, PM10 and NO_2.

			R	**BIAS**	**RMSE**
O_3	D’2	N	0.58	0.27	3.47
		FN	0.56	1.73	3.27
	D’3	N	0.54	-0.25	3.58
		FN	0.55	0.50	3.47
PM10	D’2	N	0.42	-33.45	5.73
		FN	0.41	-32.70	5.64
	D’3	N	0.34	-32.27	5.62
		FN	0.32	-31.80	5.55
NO_2	D’2	N	0.33	-20.20	4.19
		FN	0.28	-19.38	3.99
	D’3	N	0.31	-21.25	4.28
		FN	0.28	-20.18	4.07

From this analysis the difference between both simulations is not significantly relevant. Even more if is taking in account the time and cost of a simulation of WRF for all the domains.

In order to perform a more exhaustive comparison between both simulations was made a spatial comparison of the concentration fields for domain D’3. Figure 5.8, Figure 5.9 and Figure 5.10 show the results for the simulation N (a), the simulation FN (b) and the differences between both simulations for O_3, PM10 and NO_2.

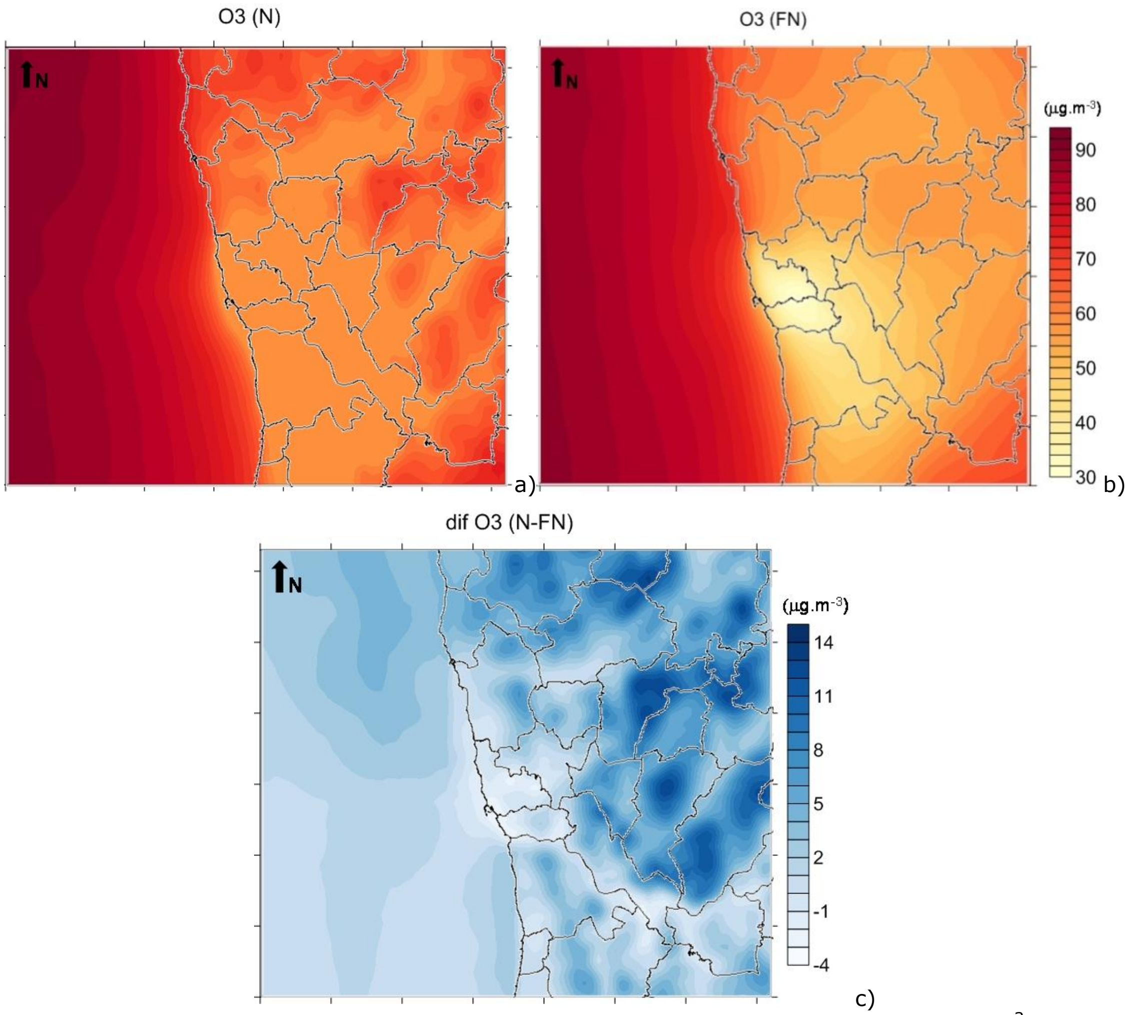


Figure 5.8 - a) Spatial distribution of O_3 monthly average concentrations ($\mu g.m^{-3}$) for the normal simulation (a), flexi-nest simulation (b) and the differences between both simulations (N-FN) (c).

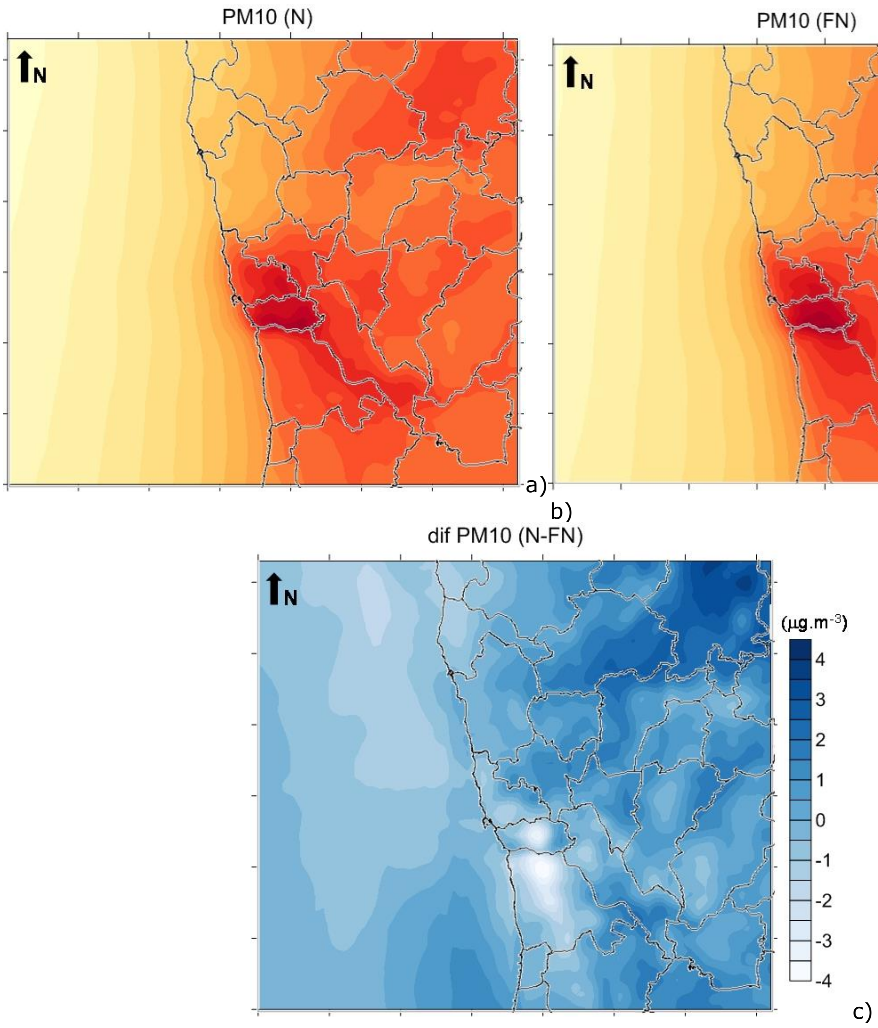


Figure 5.9 - a) Spatial distribution of PM10 monthly average concentrations (µg.m^{-3}) for the normal simulation (a), flexi-nest simulation (b) and the differences between both simulations (N-FN) (c).

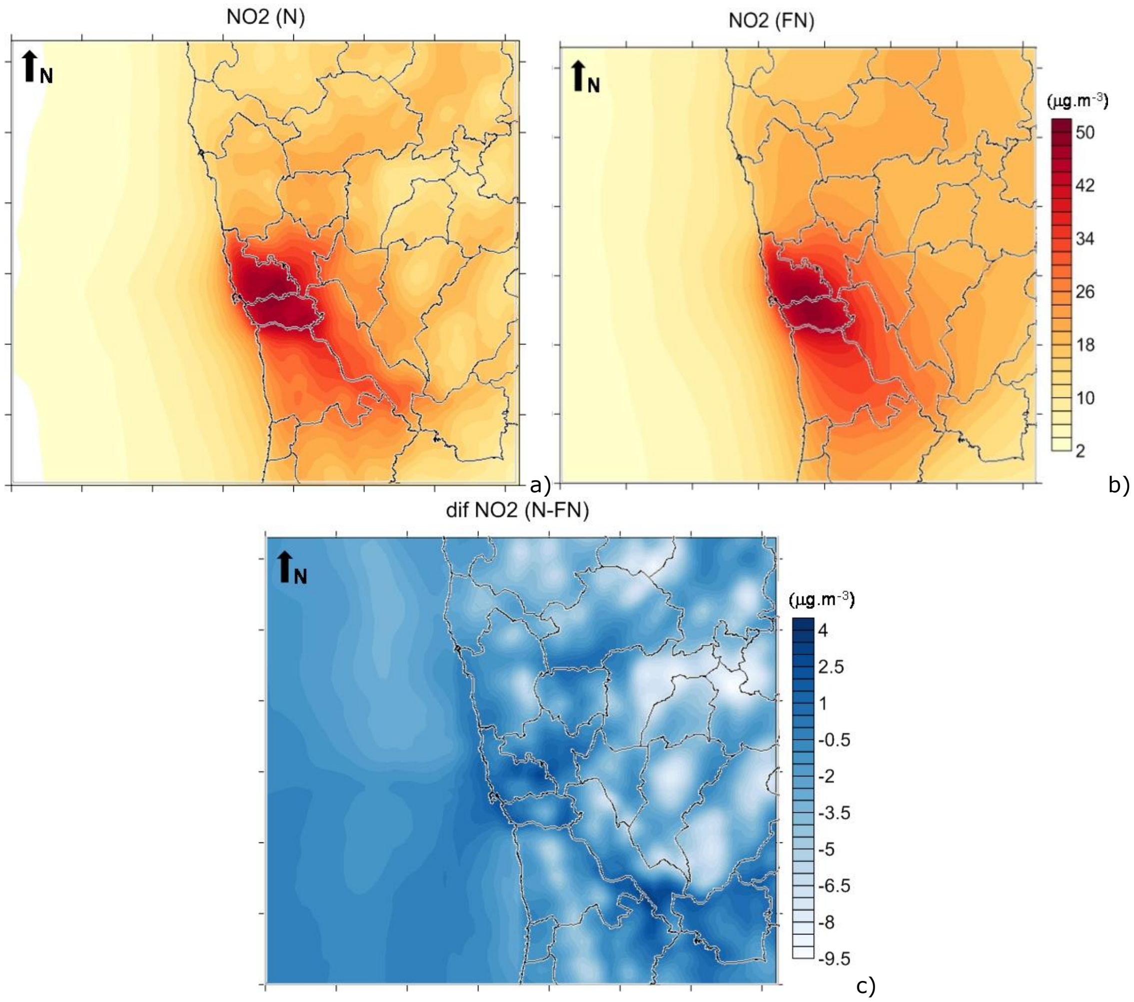


Figure 5.10- a) Spatial distribution of NO_2 monthly average concentrations (µg.m^{-3}) for the normal simulation (a), flexi-nest simulation (b) and the differences between both simulations (N-FN) (c).

From the analysis of Figure 5.8, Figure 5.9 and Figure 5.10 is possible to conclude that the higher differences between the simulations are in the O_3 concentration fields. In the FN simulation the obtained results are higher than the results obtained in the CAMx normal simulation for the municipalities were the air quality stations are located. As the results of the model are often underestimated compared with the measured values in the air quality stations for O_3, with the FN simulation CAMx shows a better performance for this pollutant.

5.3.3 Impacts on air quality by changing emissions

The results obtained from the air quality system are analyzed in terms of spatial and temporal variations. To evaluate the air quality modeling system results, a comparison with the measured data from the selected meteorological and air quality stations was performed, using quality indicators such as systematic error (BIAS), Root Mean Square Error (RMSE) and Correlation Coefficient (*r*).

The Aveiro meteorological station, located in Atlantic coastal zone of Portugal (Figure 5.7), was selected, taking into account the modeling domain and data availability, as an example for the validation of WRF results. The results obtained with the meteorological model for the inner-most domain were compared with the observed values for temperature (TEMP) and for the wind components (U and V) (Table 5.6). Also, the WRF skill was evaluated through the application of the quantitative analysis for the same meteorological parameters.

Table 5.6 - Statistical analysis of WRF model results for Aveiro meteorological station data considering the temperature and wind components for March 2011.

Statistical parameter	Optimal values	***Aveiro***		
		TEMP (ºC)	**U ($m.s^{-1}$)**	**V ($m.s^{-1}$)**
BIAS	≈0	2.85	-0.10	-0.08
RMSE	≈0	3.35	2.09	2.47
r	≈1	0.83	0.62	0.51

Table 5.6 indicates an agreement between the model results and the measurements, having better performance for temperature than for the wind components. Overestimation of air temperature is obtained for Aveiro meteorological station, while for the wind components underestimation is presented in the modeling results. In general, WRF shows a good performance simulating the meteorological data for the study domain, which is in agreement with other studies where the WRF model was applied for Portugal (Soares et al., 2012).

In order to assess the performance of the CAMx model, the model results for D2 were compared with the available monitoring data from the air quality stations located in the study domain (Figure 5.7b). Five air quality stations were selected, taking into account the collection efficiency: three stations are "urban traffic" type - Antas (ANT), Aveiro (AVR) and Espinho (ESP); one "urban background" - Ílhavo (ILH); and one "suburban background" - Sobreiras (SOB). The pollutants considered in this analysis are PM10, NO_2, CO and O_3.

The results of CAMx for PM10 concentration were compared with the measurements in terms of daily average values. For this purpose time series from all the selected air quality stations for March 2011 were considered. For the other three pollutants the hourly data from CAMx results were compared with hourly measurements from all the air quality stations. Table 5.7 shows the statistical analysis of the results for S1 and S2.

Table 5.7 - Statistics of CAMx performance for PM10 daily data and NO_2, CO and O_3 hourly data ($\mu g.m^{-3}$).

Pollutant	**Statistical parameter**	**Optimal values**	**Air quality stations**	
			S1	S2
PM10	BIAS	≈0	-10.59	-10.26
	RMSE	≈0	16.50	16.33
	r	≈1	0.38	0.37
CO	BIAS	≈0	-17.80	-13.54
	RMSE	≈0	193.51	189.84
	r	≈1	0.40	0.41
NO_2	BIAS	≈0	4.37	5.16
	RMSE	≈0	18.14	18.21
	r	≈1	0.55	0.53
O_3	BIAS	≈0	-27.51	-28.45
	RMSE	≈0	40.61	41.07
	r	≈1	0.40	0.47

Better results between S1 and S2.

Analysis of the modelling results and their comparison with the time series obtained from air quality measurements demonstrates that the model is underestimating the concentration of pollutants during the study period with the only exception for NO2 that presents positive bias. A comparison of the two scenarios shows that S2 scenario based on the improved emission data demonstrates a better model performance in terms of BIAS and RMSE for PM10 and CO concentrations. Oppositely to these primary pollutants, NO_2 and O_3 that are strongly involved in the photochemical processes do not improve in S2 and present higher BIAS and RMSE in the comparison with S1. Nevertheless, the correlation between the modelling results and measurements for O_3 is better when bottom-up methodology for traffic emissions is applied (S2) which suggests a better distribution of emissions. Additionally, variations in HC emissions considered in these two scenarios will be important to explain the behaviour obtained for the secondary pollutants. However, as we explained previously natural emissions from biogenic emissions (SNAP 11) as isoprene and monoterpenes are not considered in this study. In fact, these emissions develop an important role on O_3 chemical reactions (Pinto et al., 2007) which can help to explain these results.

The simulation results were also analysed and interpreted in terms of spatial distribution. The analysis was focused on the difference between the CAMx concentration outputs for S1 and S2, namely for PM10, NO2, CO and O_3. As an example, Figure 5.11 illustrates the spatial distribution of the differences between both scenarios for the 1st of March 2011 at 12 AM.

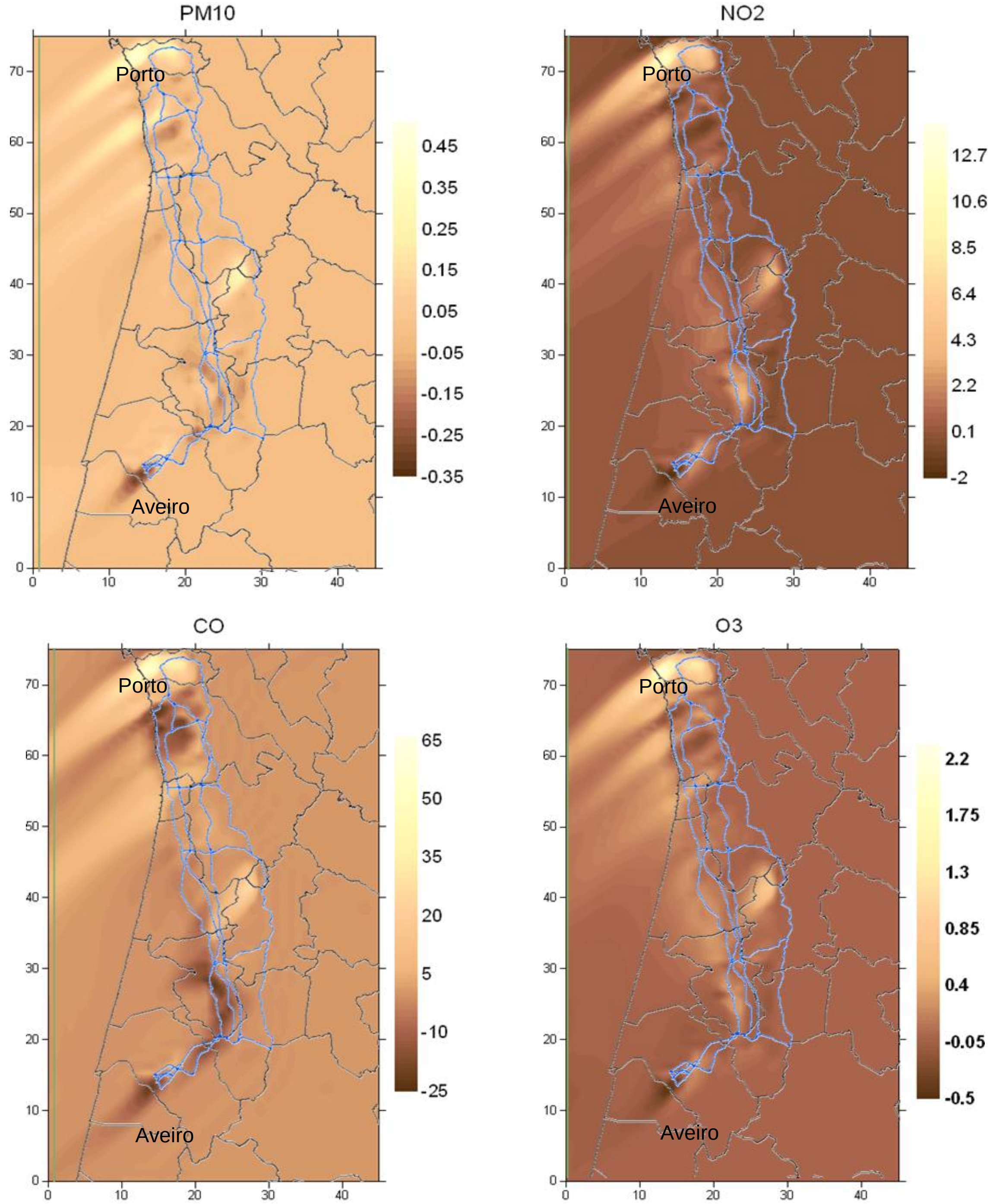


Figure 5.11 - Difference between scenario 1 and scenario 2 results from CAMx (concentration in $\mu g.m^{-3}$) - 1st March 2011 at 12 AM.

The spatial distribution of the differences between both scenarios in terms of pollutant concentrations is in agreement with the differences between the spatial distribution of the emissions, since in S1 the emissions were higher in Porto municipality than in S2, and the opposite occurred in the emissions of Aveiro municipality. For this reason there are negative differences for the four pollutants in Aveiro municipality and positive differences in Porto municipality

5.4 Summary and Conclusions

The main purpose of this chapter was to select and test the best air quality modelling system to be used as the main tool for the purposed work. In this sense, several sensitivity tests were performed, in which were analysed different capabilities of the air quality modelling system. Based on an exhaust review of the state of the art models that are being implement worldwide, the air quality modelling system selected for these tests was the WRF-CAMx because was the system that offered the necessary options for the main study.

The main conclusions from the results of the diverse tests are:

- WRF had a good performance at all the resolutions used, even at higher spatial resolutions (at least 1000m);
- For each pollutant the meteorology has a significant contribution for the simulated air quality concentrations, in this sense is extremely important to select the correct options in the WRF model, taking in consideration the study region;
- The Flexi-nesting capability of CAMx is an important option when long-term simulations are performed, as it can reduce the duration of the WRF model simulation and thus expedite the simulations;
- The use of a more detailed emission inventory is a key input to a better air quality simulation. Due to lack of available data, the top-down approach is the most used in regional simulations. However, the emissions from the traffic sector may be included through a bottom-up approach in order to improve the detail of the emission inventory and, as consequence, the results from the air quality simulations.

In general WRF-CAMx is an air quality modelling system that demonstrates a good performance when is applied over Portugal and over Porto urban area and for this reason is a fundamental and helpful tool to be use in air quality studies.

Chapter 6

6 Linking air quality with climate change at national and urban scale

The contents of this chapter will be published in:

Sá, E., *Martins, H., Ferreira, J., Almeida, M., Rocha, A., Carvalho, A., Borrego, C. (submitted). Climate change and pollutant emissions impacts on regional and urban air quality in 2050. Atmospheric Environment.*

Abstract

Changes in climate and air pollutant emissions will affect future air quality from global to urban scale. In this work, regional air quality simulations for historical and future periods are conducted with CAMx version 6.0 to investigate the impacts of future climate and anthropogenic emission projections on air quality over Portugal and Porto metropolitan area. The climate and the emission projections were derived from the Representative Concentrations Pathways (RCP8.5) scenario. By itself, changes in atmospheric patterns due to climate change impact NO_2, PM10 and O_3 concentrations in the atmosphere in Portugal. The NO_2 and PM10 annual mean will increase in Portugal and in Porto municipality. In the case of O_3, the maximum 8-hr daily value will increase in suburban areas (around 5%) and decrease in urban areas (around 2%), such as Porto municipality. When considering climate change and projected anthropogenic emissions, the NO_2 annual mean decreases (around 50%); PM10 annual mean will increase in Portugal and decrease in Porto municipality (around 13%); however PM10 and O_3 levels increase and extremes occur more often, surpassing the currently legislated annual parameters and showing a higher frequency of daily exceedances. In terms of PM10 and O_3 concentration levels, the Porto urban area was better characterized with the results from the air quality simulation at higher resolution. The results show the need for Portuguese authorities and policy-makers to design and implement air quality management strategies that take into account climate change impacts.

6.1 Introduction

Changes in climate are firmly expected over the 21st century (IPCC, 2013b). Climate change will result in an impact on the general weather patterns, in particular, wind climatology, temperature, sunshine hours and rainfall patterns. This in turn may result in a change in the processes that govern chemical transformations in the atmosphere, which will cause air quality changes (AQEG, 2007).

Future projections of air quality must account for changes in both future emissions and climate due to their closely-coupled impacts on air quality (Penrod et al., 2014). Major pollutants, such as O_3 and PM, are sensitive to changes in climate, namely to perturbed ventilation rates (wind speed, mixing depth, convention and frontal passages), precipitation scavenging, dry deposition chemical production and loss rates, natural emissions and background concentrations (Jacob and Winner, 2009). Also, the influence of climate change on the pollution levels must be studied by using an adequate set of emissions scenarios, since the changes in emissions of primary air pollutants and the precursors of secondary pollutants will lead to changes in air quality (Zlatev and Moseholm, 2008 and Penrod et al., 2014).

Numerical modelling represents a powerful tool to assess the influence of future climate scenarios on the air pollutant concentrations and consequently in air quality management. There are three main approaches to study air quality under future climate based on air quality modelling systems:

- the studies that only consider the effect of climate change, keeping anthropogenic emissions at constant values (Langner et al., 2005; Meleux et al., 2007; Racherla and Adams, 2008;Zhang et al., 2008; Fiore et al., 2011; Carvalho et al., 2010; Tai et al., 2012; Manders et al.; 2012);
- the studies that keep the meteorology conditions constant (same as the historical year) in future scenarios and only change the pollutant emission scenarios (Dentener et al., 2005; Zhang et al., 2010);
- the studies that consider the effect of climate change along with the modification of the anthropogenic emissions in the future scenarios (Unger et al., 2006; Dentener et al., 2006; Stevenson et al., 2006; Tagaris et al., 2007; Nolte et al., 2008; Lei et al., 2012; Coleman et al., 2013; Doherty et al., 2013; Gao et al., 2013; Jiang et al., 2013; Colette et al., 2013; Penrod et al., 2014; Trail et al., 2014; Lacressonnière et al., 2014).

In terms of emission scenarios, most of the studies conducted so far perform the future-year simulations based on the IPCC scenarios developed in the last years: IPCC IS92 (Langner et al., 2005); IPCC SRES scenarios (Tagaris et al.,2007 ; Nolte et al., 2008; Manders et al., 2012; Doherty et al., 2013; Jiang et al.,2013; Penrod et al., 2014; Trail et al., 2014); and IPCC RCP scenarios (Gao et al., 2013; Coleman et al., 2013; Lacressonnière et al., 2014). Also, there are studies that, besides the IPCC scenarios, also considered the IIASA emission scenarios (MFR and CLE) and performed a comparison of the obtained results (Dentener et al, 2005; Stevenson et al., 2006).

The vast majority of the studies assess the impacts of climate change and pollutant emissions on ozone (O_3). Climate change alone will affect the factors that influence O_3 patterns, such as temperature, water vapour, cloud cover, mixing height and precipitation, and that contribute to variations in O_3 levels (Hogrefe et al., 2004; Lam et al., 2011; Carvalho et al., 2010; Coleman et al., 2013; Gao et al., 2013). To evaluate the combined impact of climate change and anthropogenic emissions on O_3, Lacressonnière et al. (2014) simulated changes in future air quality in Europe for the 2030s and 2050s, under the RCP8.5 scenario. These simulations showed an increase in surface ozone in north-western Europe and a decrease in southern areas. Over Europe, average O_3 levels steadily increase at a rate of around 3 $\mu g.m^{-3}$ per decade in Summer time. The tropospheric ozone budget was found to be dominated by enhanced stratosphere-troposphere exchanges in future climate while the chemical budget is significantly reduced. Also, the results point out that a NOx-limited chemical regime will stretch over most of Europe, including especially Western France in the future. Over Portugal, Carvalho et al. (2010) concluded that O_3 monthly mean levels in the atmosphere may increase almost 20 $\mu g.m^{-3}$ in July when only climate change forcing was considered.

In the last years, besides assessing the impacts on O_3, studies have also focused on the evaluation of the impacts on PM2.5 (Tagaris et al., 2007; Racherla and Adams, 2008; Zhang et al, 2010; Lam et al., 2011; Tai et al., 2012; Jiang et al., 2013; Colette et al. 2013; Penrod et al., 2014; Trail et al., 2014;) and PM10 (Carvalho et al., 2010; Manders et al., 2012). The studies which have examined the impact of climate change on surface PM concentrations showed some differences on the conclusions, including the sign of the effect and the identification of the most important driver of PM sensitivity to climate change. Carvalho et al. (2010) found that, throughout Portugal, the maximum increases are foreseen for the northern coastal region in September reaching nearly 30 $\mu g.m^{-3}$.

The interaction between climate change, pollutant emissions and atmospheric concentrations is still of great debate and much has still to be explored in order to understand and accurately predict the changes in pollutant levels under future climatic conditions and at different spatial scales. In this sense, the objective of this study is to evaluate air quality over mainland Portugal and over Porto metropolitan area, which is the second major urban area in Portugal and is, nowadays, facing air quality problems, under the IPCC RCP8.5 scenario. This work is distinguished from previous studies that have included the impacts of both climate and emission changes as it uses one of the latest Comprehensive Air quality Model with Extensions (CAMx) (Tesche et al., 2006) released versions (version 6.0) that contains updated model treatments for several processes, and it was tested to the study region showing a good behaviour. Also, most of the studies focused only on O_3 and PM (mainly PM2.5) while in this work the three critical pollutants in Portugal (O_3, PM10 and NO_2) will be analysed. This study considered a 5-year model simulation in terms of meteorological and chemical predictions, instead of the single or ≤3 years (or seasonal/monthly) evaluation. Finally, there are no studies with this detail for Portugal or for an urban settlement like Porto metropolitan area.

This chapter is structured as follows. Section 6.2 describes the modelling system and the pollutant emissions scenario. The modelling results and discussion are provided in Section 6.3. The conclusions are given in Section 6.4.

6.2 Data and Methods

This section presents the input data considered in the air quality modelling simulations, namely climatic data and emission projection scenarios adopted. The air quality modelling system characteristics and setup for the current application are also described.

6.2.1 Input Data

6.2.1.1 Climatic data

The WRF model (regional meteorological model) has been used to downscale global climate simulations previously performed by the Earth Systems Model MPI-ESM-LR, which has been shown to be one of the most robust models to simulate European climate (Brands et al., 2013).

Three domains, online nested, were used covering part of the North Atlantic and Europe, with a resolution of 81 km (D-1), of 27 km (D-2) and reaching 9 km in the innermost domain (D-3) which covers the Iberian Peninsula (Figure 6.1). Two simulation periods were studied: (i) historical (1986 to 2005) and (ii) medium term future (2046 to 2065). For the future simulations, the IPCC greenhouse gas concentration scenario RCP8.5 has been adopted. For validation purposes, an additional simulation for the year 2011, forced by ERA-INTERIM data was performed. For a detailed description of this downscaling approach and modelling configuration and validation see Marta-Almeida et al., 2015.

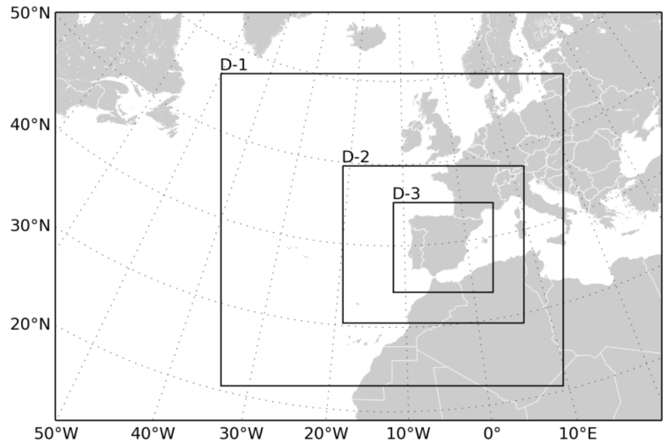


Figure 6.1 – WRF domains used in the simulations: D-1 covering part of the North Atlantic and Europe, with a resolution of 81 km, D-2 with a of resolution 27 km and reaching 9 km in the innermost domain (D-3) which covers the Iberian Peninsula.

6.2.1.2 Atmospheric pollutant emissions projections

For the current emissions, a top-down methodology was applied to disaggregate the anthropogenic emissions, using the Portuguese national emission inventory (INERPA) (PEA, 2014). This inventory provides quantitative information for the main atmospheric pollutants: CO, NOx, N_2O, SOx, NMVOC, NH_3, PM10 and PM2.5. The total national emissions (country resolution) are reported for 11 main source categories (SNAP) (Table 4.2).

The annual national emissions from anthropogenic area sources for each pollutant and activity sector were spatially disaggregated to the sub-municipality level for SNAP2, SNAP3, SNAP4, SNAP6, SNAP9, SNAP10 and to the municipality level for SNAP5, SNAP7 and SNAP8. The data are then converted to a regular grid, using GIS, for the spatial resolution required for the selected simulation domains.

Also, for the traffic sector (SNAP7) a bottom-up methodology combined with the top-down methodology was applied to this sector (described in Chapter 4).

For the future emissions, the EmiPro-RCP model (described in Chapter 4) was applied using the following options:

- Base year: 2010
- Projection year: 2050
- RCP scenario: 8.5
- Study region: OECD90
- Analysis: SNAP

The scenario RCP8.5, corresponding to the pathway with the highest greenhouse gas (GHG) emissions, was selected to perform the future simulations. This scenario combines assumptions about high population and relatively slow income growth with modest rates of technological change and energy intensity improvements, which will lead to a high energy demand and GHG emissions (Riahi et al., 2011). The total emissions of the main anthropogenic pollutants are given in Table 6.1.

Table 6.1 – Total annual anthropogenic emissions of the main air pollutants (Gg) for Portugal.

	2011	**RCP 8.5 - 2050**
NH_3	47.47	59.71
SO_2	43.37	10.99
NOx	161.22	80.11
VOC	168.50	95.73
CO	305.51	140.35
PM10	73.53	43.03
PM25	55.98	32.76

6.2.2 The air quality modelling system

The meteorological outputs for D3 (9 km resolution, Figure 6.1) from the WRF application with ERA-INTERIM or MPI-ESM-LR are used as inputs in CAMxv6.0. In this study, the focus will be the CAMx model application. CAMx is an Eulerian photochemical dispersion model that considers the emission, dispersion, chemical reaction, and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species on a system of nested three-dimensional grids (Morris et al., 2004). CAMx vertical structure includes 15 layers and in terms of chemical mechanism, the gas-phase photochemistry was resolved through the Carbon Bond (CB5) (Yarwood et al., 2005) and the model also contains detailed algorithms for the relevant processes, including aqueous chemistry (RADM-AQ), inorganic aerosol thermodynamics/partitioning (ISORROPIA), and secondary organic aerosol formation/partitioning (SOAP). Initial and boundary conditions for both gases and particulate species were driven by the MOZART4 model (using GEOS met) to be included in the pre-processor MOZART2CAMx. For the future simulations, the MOZART4 initial and boundary conditions were estimated based on the results of the LMDZ-INCA model for 2006 and 2050 (Szopa et al., 2013). The preparation of initial and boundary conditions for the air quality simulations for 2050 involved a considerable amount of work, including the preparation of a set of Fortran and Python programs to convert the LMDZ-INCA model results for 2006 and 2050 to the necessary format. Data from the Ozone Monitoring Instrument (OMI)/AURA (URL5) was used as input to run the CAMx pre-processor (O3MAP) to estimate the ozone column. The ozone column, in the future simulations, was estimated based on the ozone database in support of CMIP5 simulations (Cionni, I. et al., 2011). Figure 6.2 illustrates the air quality system WRF-CAMx flowchart.

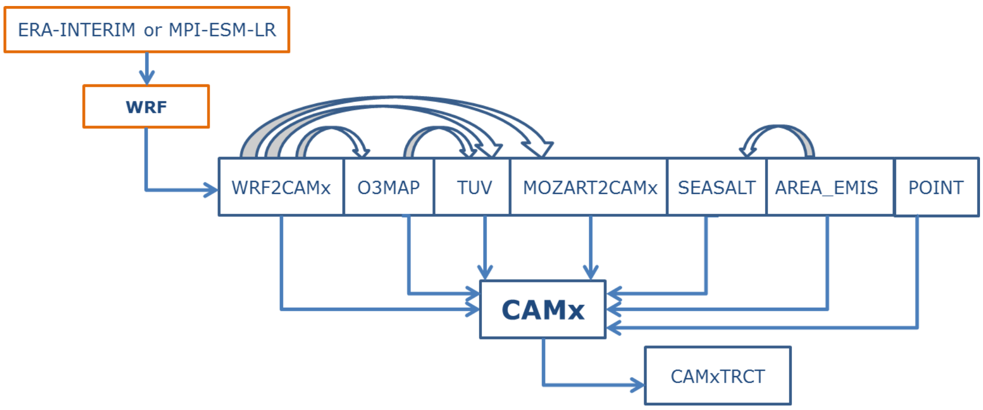


Figure 6.2 – Air quality modelling system WRF-CAMx flowchart.

CAMx was applied with the flexi-nesting capability enabled, which is an algorithm that interpolates the missing fields from the parent grid, in this case, meteorological fields into the other nesting grids. The use of this CAMx capability reduces the time of simulation.

The air quality simulations considered three domains: D1 with a 9 km spatial resolution, covering Portugal, D2 with a 3 km spatial resolution, covering the Northern Region and D3 with a 1 km spatial resolution, covering the Porto metropolitan area (Figure 6.3).

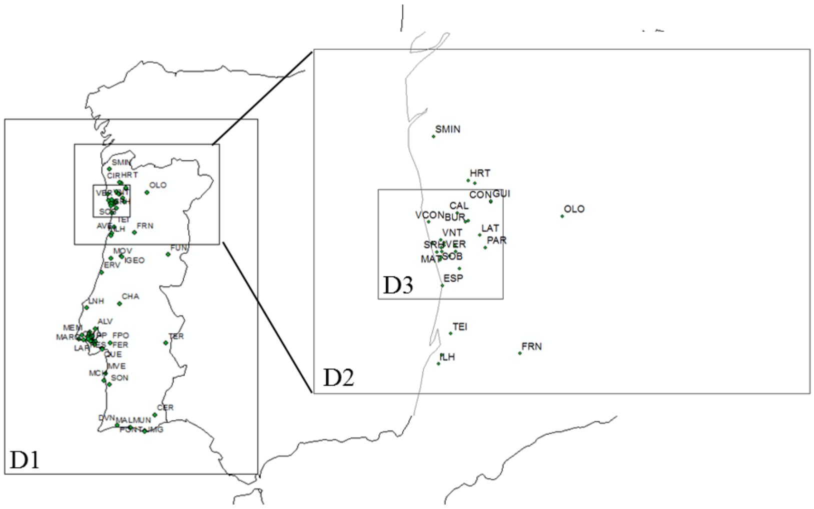


Figure 6.3 - Simulation domains used by CAMx air quality model and the location of air quality stations used for model validation and analysis of results.

Four different simulations were performed with CAMx (Table 6.2).

Table 6.2 – Characteristics of the simulations considered in this study.

Simulations	Current	CLIM	S2050_clim	S2050_emiss
Periods	2011	5 yr of historical climate	5 yr of 2050 period	5 yr of 2050 period
Meteorological forcings	ERA-INTERIM Re-analysis	MPI CLIM	MPI RCP8.5 – 2050	MPI RCP8.5 – 2050
Anthropogenic Emissions	Current emissions	Current emissions	Current emissions	Projected emissions under the RCP8.5 for 2050
BC/IC	MOZART for 2011	MOZART for 2011	MOZART for 2011	MOZART for 2050
Ozone column	OMI data for 2011	OMI data for 2011	OMI data for 2011	OMI data for 2050

6.3 Results and Discussion

In this section results from the four air quality modelling system simulations were explored. In section 6.3.1 the results from the Current simulation were compared with the measured data from the national air quality monitoring network. The two simulations for 2050 were compared with the CLIM simulation and the analysis is discussed in section 6.3.2.

6.3.1 Air quality modelling system validation

In order to evaluate the performance of the air quality modelling system for the three domains, the results were compared with the measured data in 2011, using the DELTA tool framework. The DELTA tool (Thunis et al., 2012) is an IDL-based evaluation software tested in a set of studies which addresses model applications for the air quality Framework Directive (FD) (Directive 2008/50/EC), mainly assessment. The pollutants (O_3, PM, and NO_2) and temporal timespans (hourly and yearly frequency for PM and NO_2; and 8-hr daily maximum frequency for O_3) analysed within DELTA tool are those relevant to the FD and critical in Portugal. The DELTA tool produces the necessary information needed for the accurate validation of the modelling system namely the target plot and the summary report. The target plot represents the distance between the measured and modelled data, combining several statistical parameters, such as BIAS, correlation (R) or standard deviation (SD). The percentage value of air quality stations that fulfil the target criteria is identified on the diagram and should be higher than 90%. The summary report summarizes the statistical analysis (BIAS; R; SD and spatial correlation), for the several air quality stations considered (Figure 6.3). The other plots from the application of Delta tool are in Appendix C.

The performance results for daily average NO_2 concentrations, for 8-hr daily maximum O_3 concentrations and for daily average PM10 concentrations are exhibited in Figure 6.4 to Figure 6.6 for each domain.

D1		
NO_2	O_3	PM10

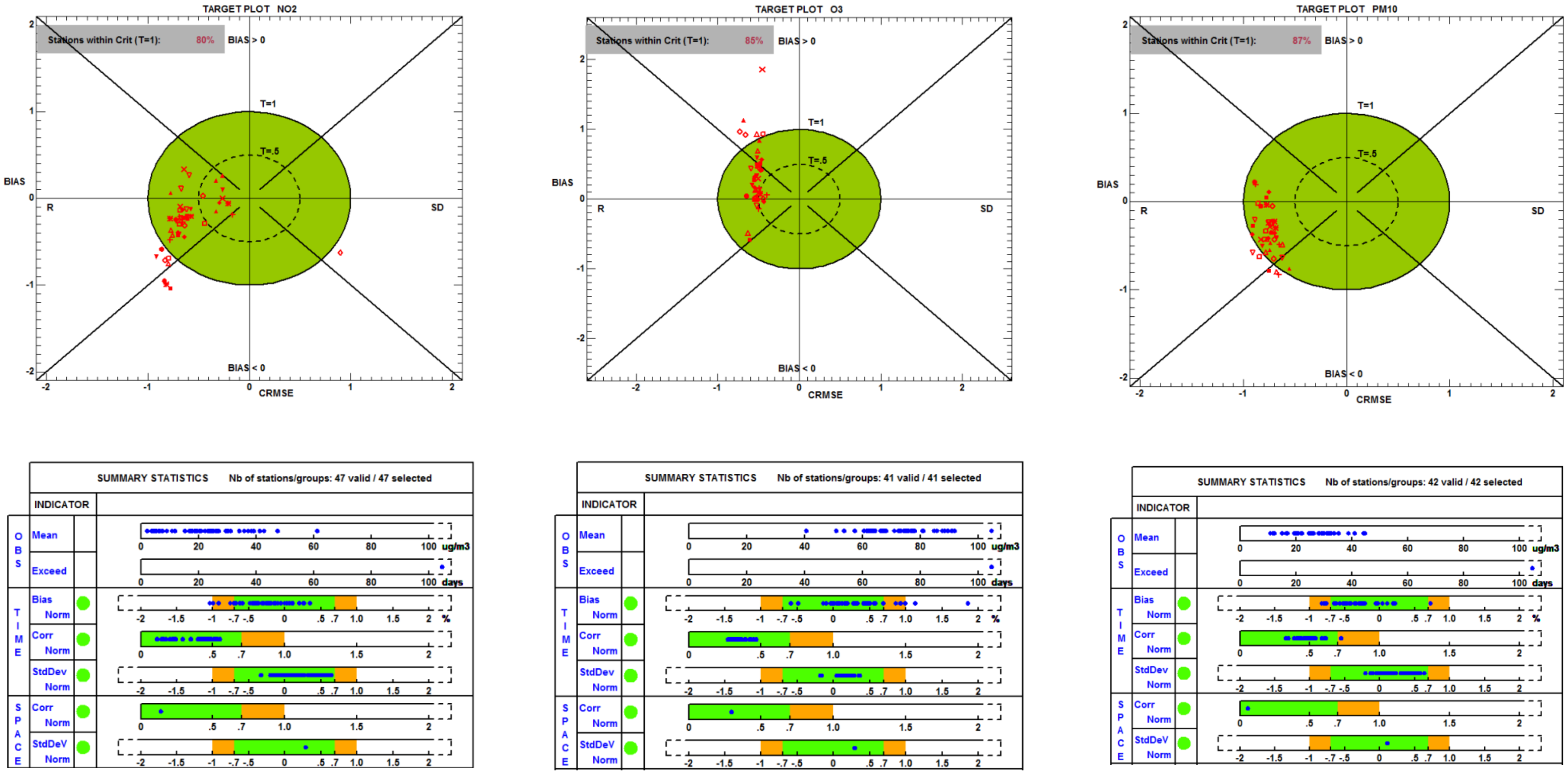


Figure 6.4 – Delta tool plots for CAMx D1 for NO_2, O_3 and PM10.

D2		
NO_2	O_3	PM10

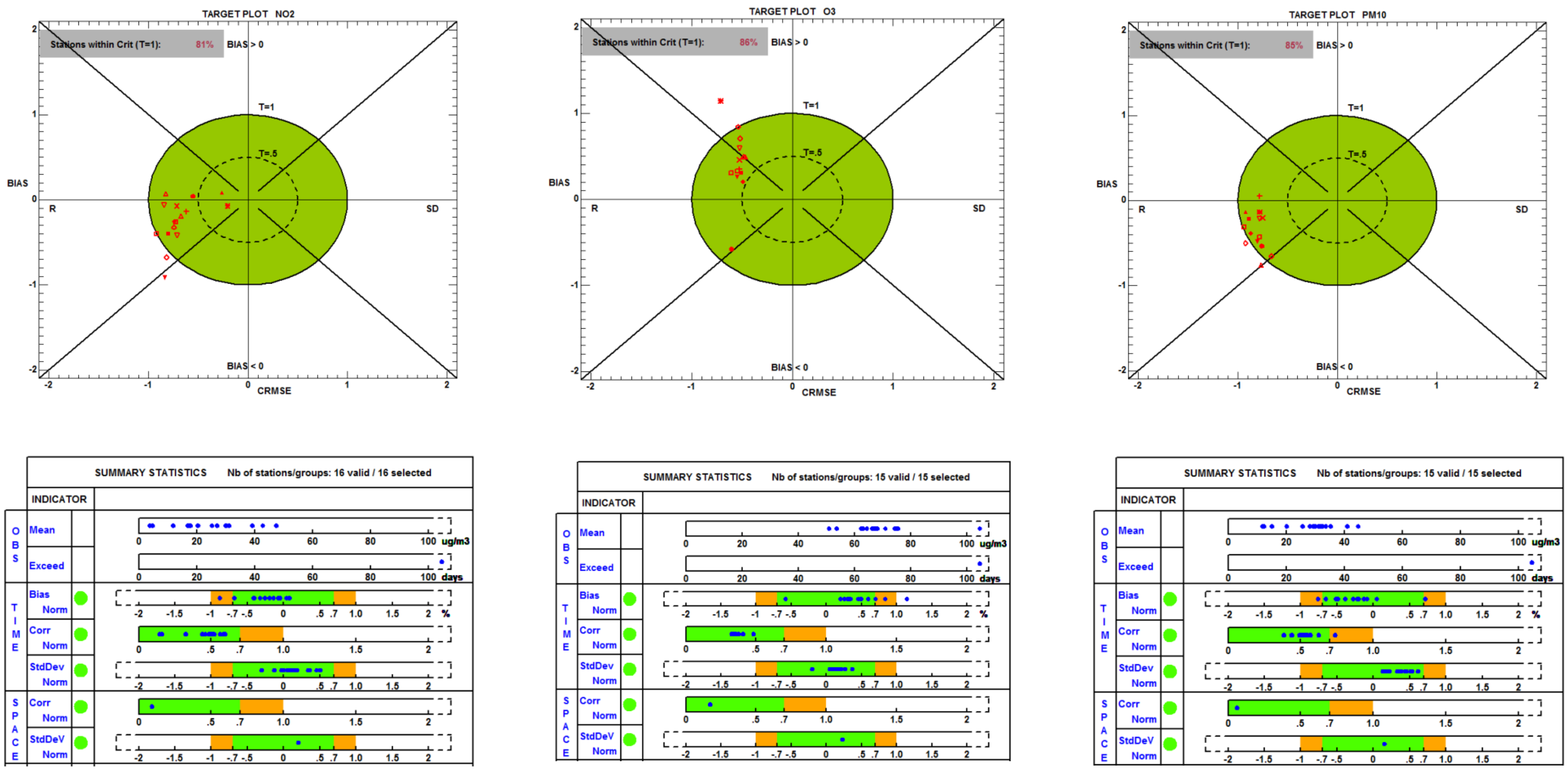


Figure 6.5 – Delta tool plots for CAMx D2 for NO_2, O_3 and PM10.

D3		
NO_2	O_3	PM10

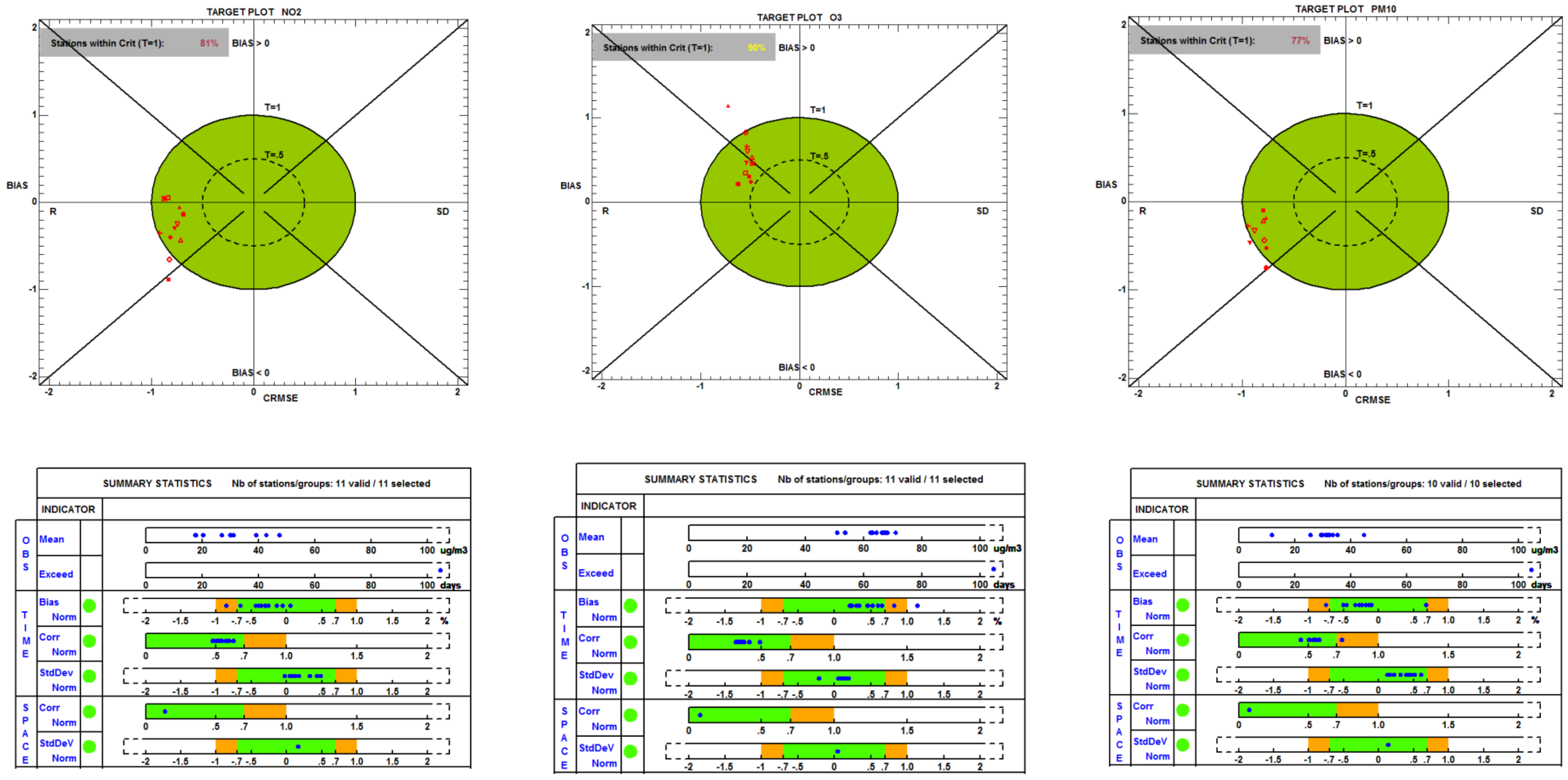


Figure 6.6 – Delta tool plots for CAMx D3 for NO_2, O_3 and PM10.

For D1 the air quality modelling system shows a good performance for all the three atmospheric pollutants. Taking in consideration the information from the target plot, PM10 have the highest value for the percentage of air quality stations that fulfil the criteria (T=1); however the lowest value is for NO_2 and is 80%, which demonstrates that a higher percentage of air quality stations fulfil the criteria for this pollutant. For NO_2 and PM10 the BIAS is predominately negative, which indicates that the model underestimates the concentration for these two atmospheric pollutants. On the other hand, for O_3 the model overestimates the concentration (BIAS predominately positive). For all the three pollutants the correlation is higher than 0.5, representing a good performance of the air quality modelling system for D1.

For the other two domains the percentage of air quality stations that fulfil the criteria increases for NO_2 and O_3, with this last one achieving 90% in D3. For PM10, the percentage of air quality stations that fulfil the criteria decreases to 77% in the innermost domain, due to a reduced number of air quality stations (9) located in the innermost domain from which two of them are not fulfilling the criteria. In D3 the model is again underestimating the NO_2 and PM10 concentrations, and overestimating the O_3 concentrations. Most of the air quality stations for all the three pollutants show correlation factors above 0.5 for both studied domains.

Globally, the summary statistics highlight that the model presents a good performance in simulating concentrations of NO_2, O_3 and PM10, presenting a positive behaviour for all parameters within the DELTA tool, both in terms of time or space.

6.3.1 Air quality assessment under future climate

The air pollutant concentrations under future climate conditions are discussed in this section. The analysis concerns the comparison between the results from the future climate simulations (S2050_clim and S2050_emis) against the historical simulation (CLIM). Firstly, a spatial analysis of NO_2 and PM10 annual mean and O_3 26th maximum daily eight hour mean is performed for D1 – mainland Portugal (Figure 6.7-Figure 6.9) and D3 – Porto urban area (Figure 6.10-Figure 6.12).

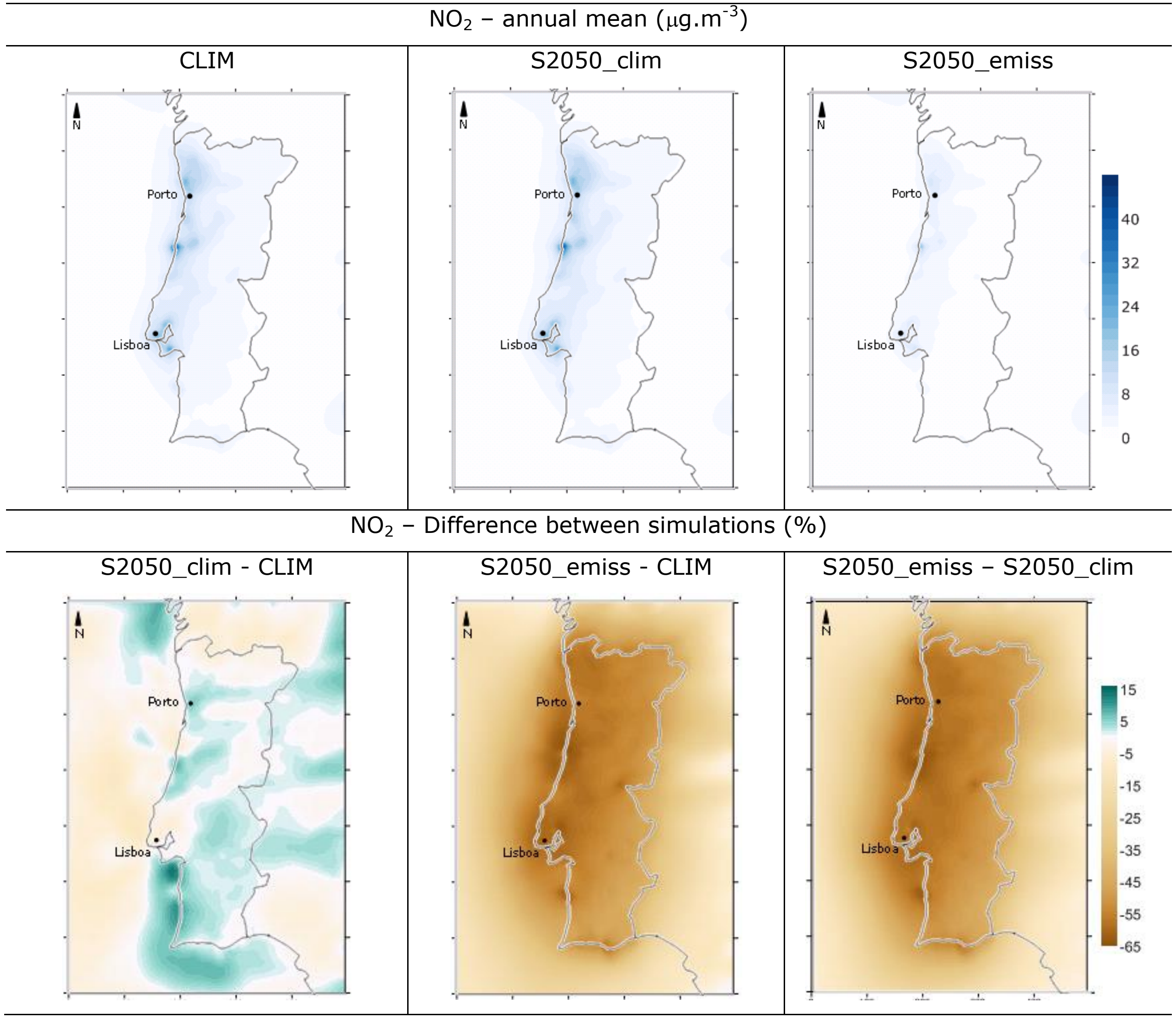


Figure 6.7 – NO_2 annual mean in each simulation and the difference in percentage between simulations (D1).

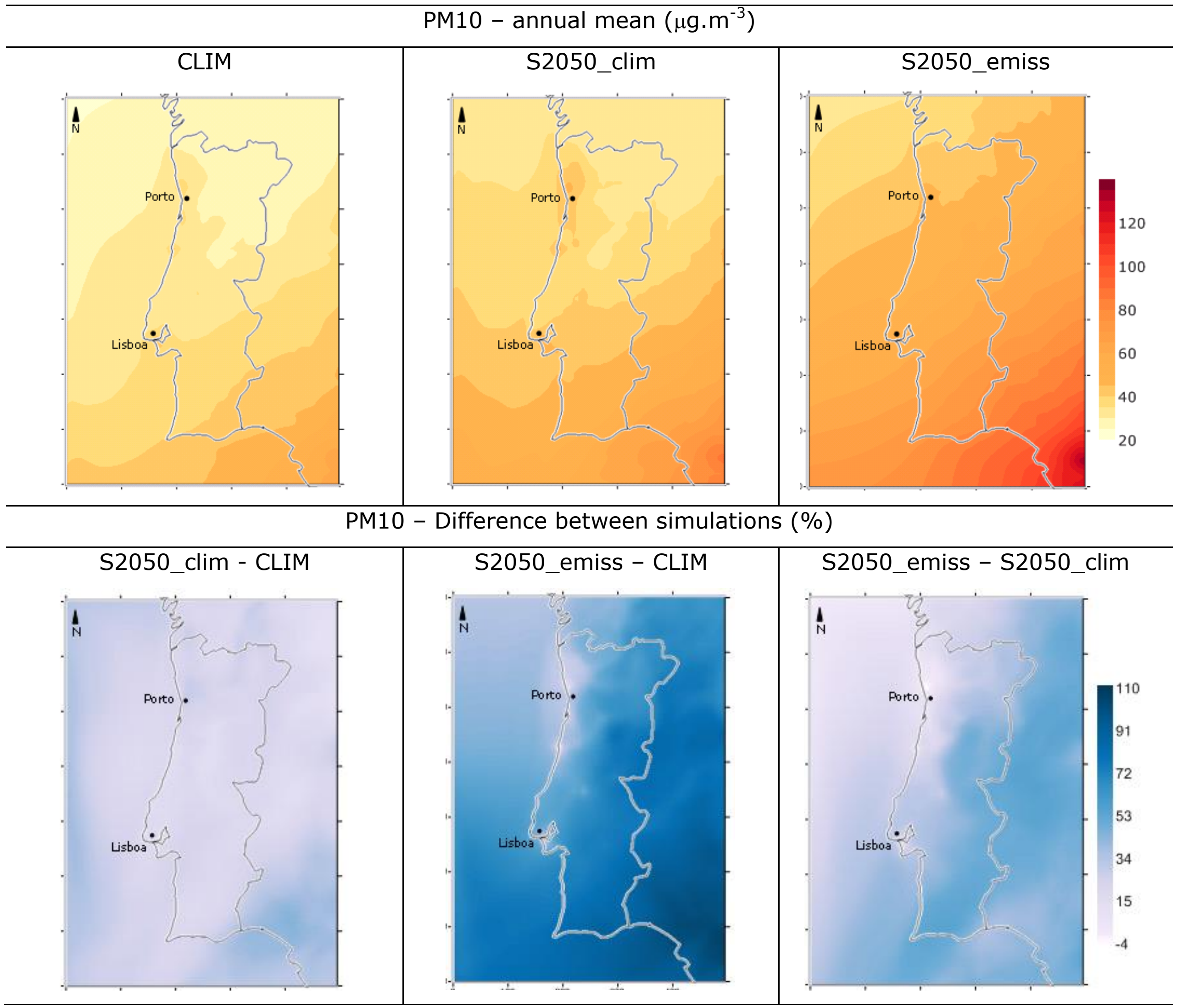


Figure 6.8 – PM10 annual mean in each simulation and the difference in percentage between simulations (D1).

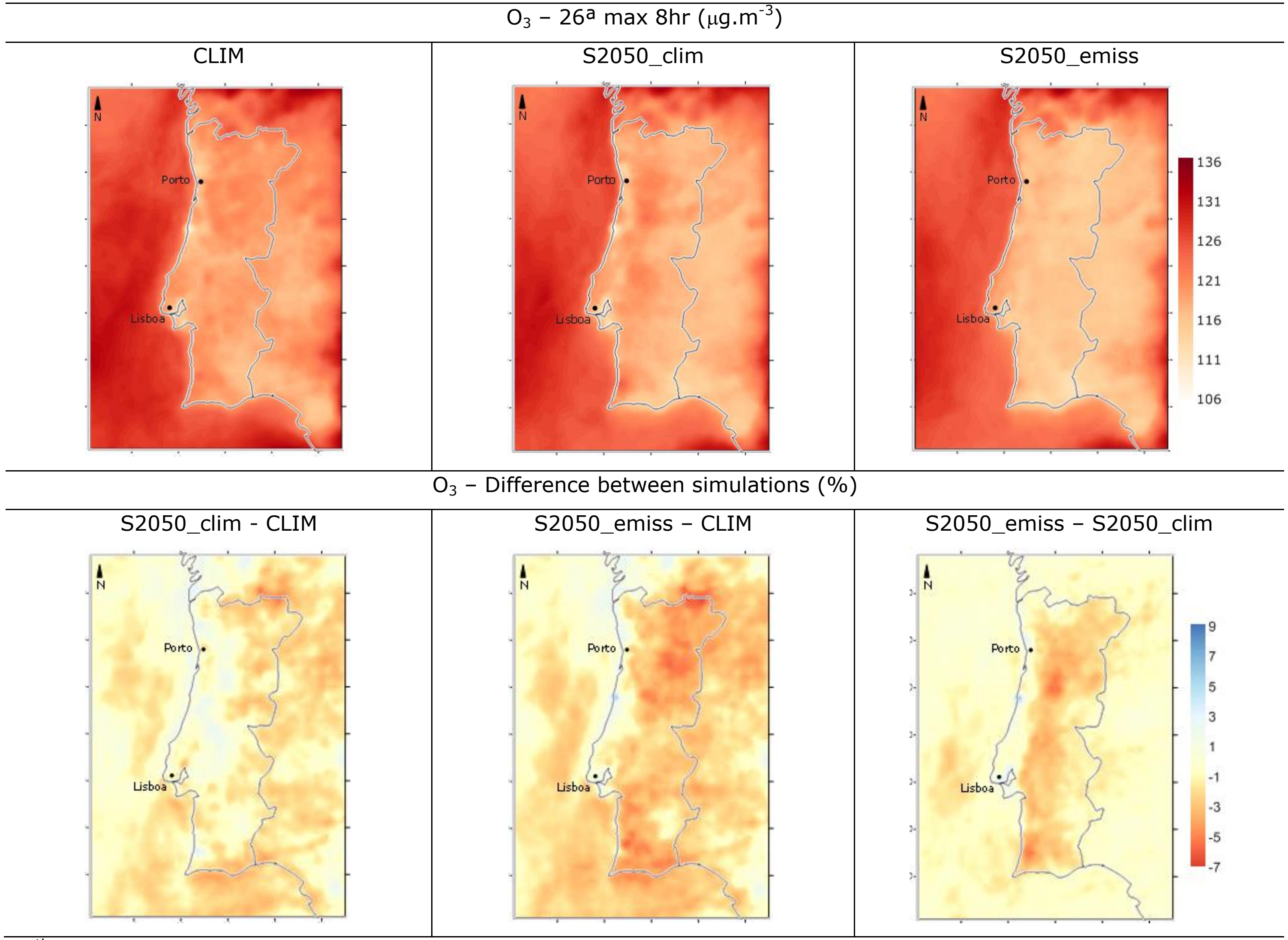


Figure 6.9 – O_3 26th highest maximum 8hr daily value in each simulation and the difference in percentage between simulations (D1).

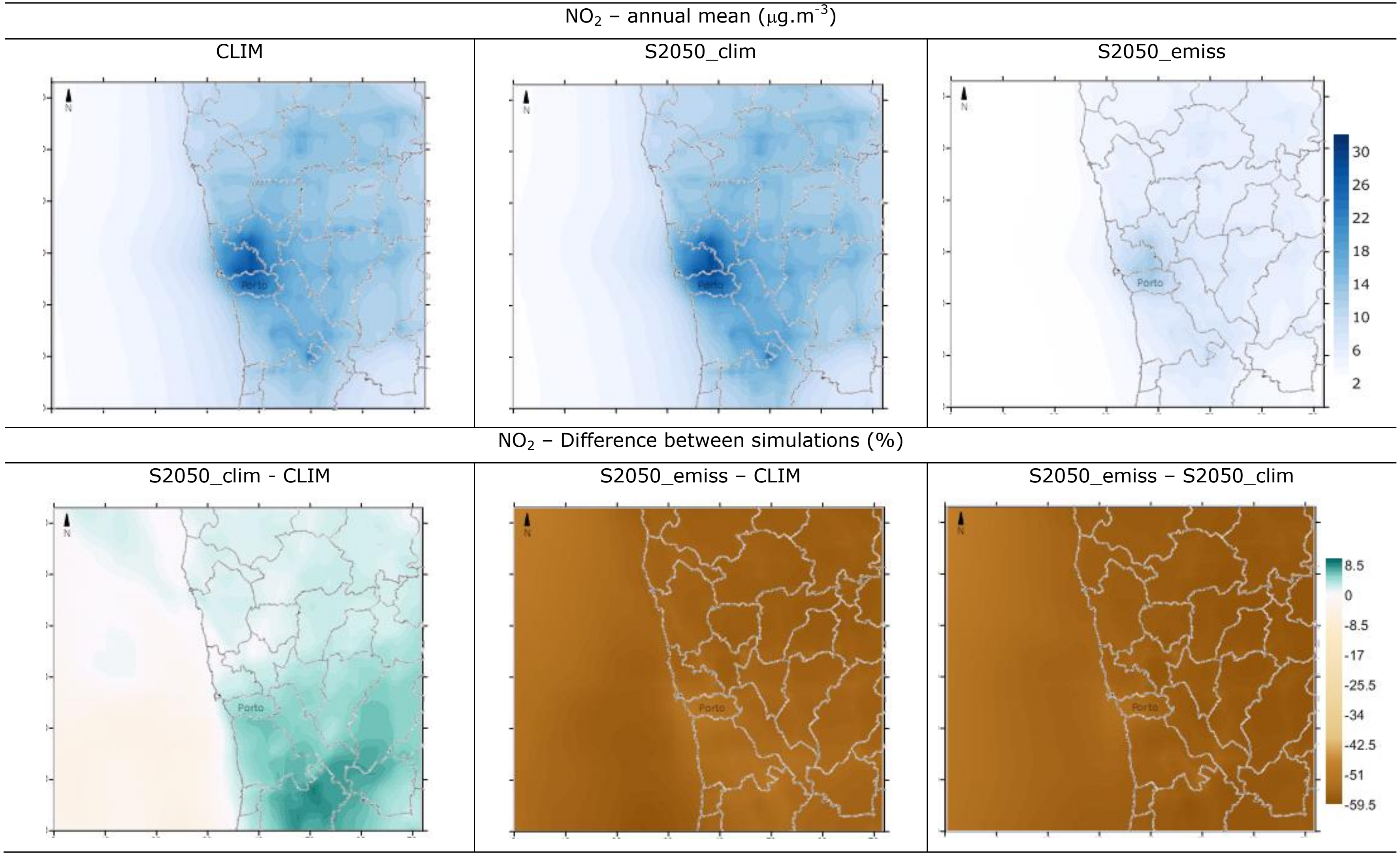


Figure 6.10 - NO_2 annual mean in each simulation and the difference in percentage between simulations (D3).

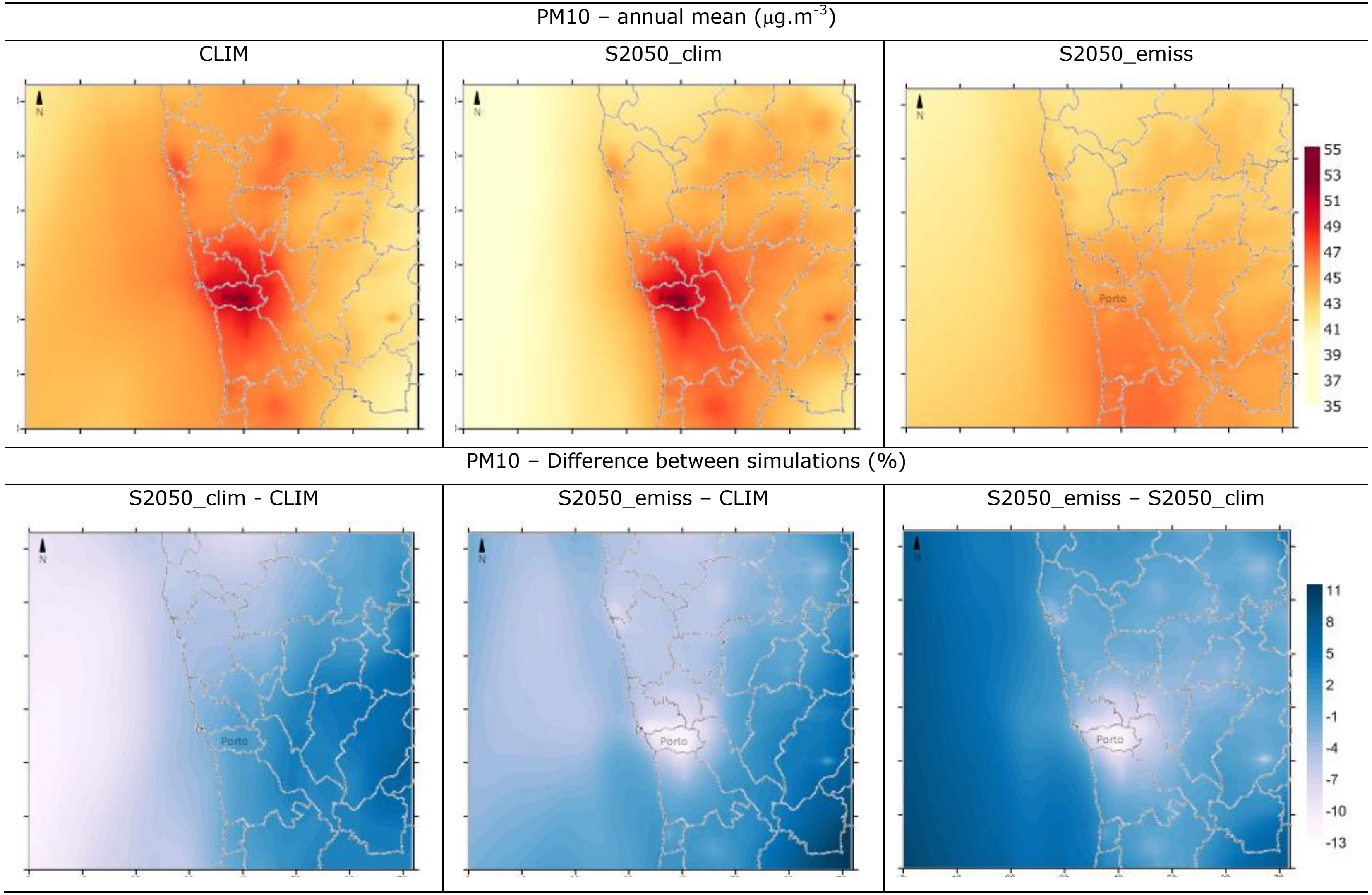


Figure 6.11 – PM10 annual mean in each simulation and the difference in percentage between simulations (D3).

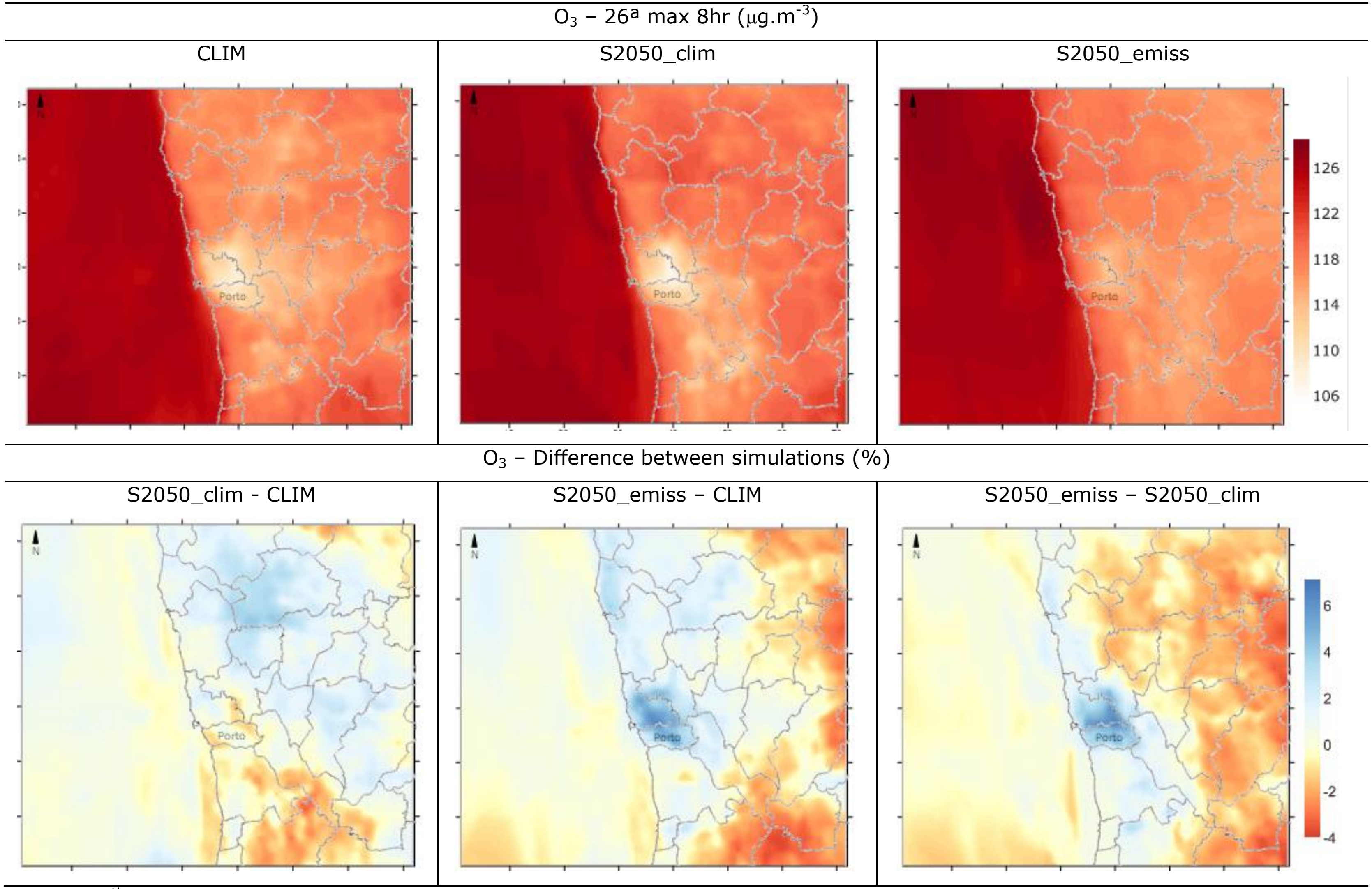


Figure 6.12 - O_3 26th highest maximum 8hrour daily value in each simulation and the difference in percentage between simulations (D3).

From the S2050_clim simulation, which just considers the climate change effect, the NO_2 annual mean will increase in most of the regions of Portugal and in Porto urban area; in addition, PM10 annual mean will also increase over Portugal and in Porto urban area; O_3 26th highest maximum 8-hr daily value will also increase over Portugal and in the surroundings of Porto urban area. The increase of NO_2 annual mean level will be of almost 15% in Portugal and between 2 and 9% in Porto urban area. In the case of PM10 the annual mean increases will not be so high over mainland Portugal (around 8%), but will reach 11% over Porto. O_3 will decrease almost 5% in the inland of Portugal and will increase almost 3% in coastal areas. In the innermost domain, the municipalities located north of Porto will register a 5% O_3 increase and will have a decrease of 2% over Porto municipality. The relative changes, in terms of percentage, are not very high; however, nowadays, these pollutants are already showing exceedances to the limit values and, consequently, these positive changes will potentiate the occurrence of acute air quality problems.

The comparison between the S2050_emiss and the other two simulations is not so linear among the three pollutants analysed. For NO_2 annual mean concentration, there is a reduction between 45 and 55% across Portugal, with higher values in the coastal regions. Also, in D3 the reduction reaches 50%, mainly in Porto municipality. The decrease of NO2 annual mean level is explained by the reduction of the projected emissions, and not by climate, with the concentration differences between CLIM and S2050_emiss very similar to the map of the differences between S2050_clim and S2050_emiss.

When considering climate change effects along with the projected emissions (S2050_emiss), PM10 annual mean level will increases in Portugal (D1) and decreases in the urban area (D3), reaching 13% reduction in Porto municipality. In the case of D1, the differences between the comparison of CLIM and S2050_emiss and the comparison of S2050_clim and S2050_emiss allow to understand that the increase is not only due to the projected emissions, but also due to the climate change effect. Furthermore, D1 is strongly affected by the boundary conditions of MOZART model, as can be seen from the maps of the annual mean; and the projections of RCP8.5 for 2050 indicate an increase of MOZART concentrations in terms of dust, which could support this increase in PM10 concentrations.

For O_3, in the S2050_emiss simulation the highest reductions in D1 are located in inland of Portugal and the highest increases are over urban areas, such as Porto and Lisboa, reaching 4% of O_3 increase. In D3 there is an O_3 increase of almost 6% over Porto municipality and in the surrounding municipalities. Comparing both future simulations, the increase on O_3 concentrations registered in the inland of

Portugal is higher in S2050_clim than in S2050_emiss, which could be justified by the reduction of ozone precursors in the 2050 emission projections.

Next, the analysis will focus on the innermost domain, Porto urban area, starting with the study of the air quality data in each air quality station located in this domain. Figure 6.13 shows the NO_2 and PM10 annual mean and the O_3 maximum daily 8-hr value at the air quality stations locations.

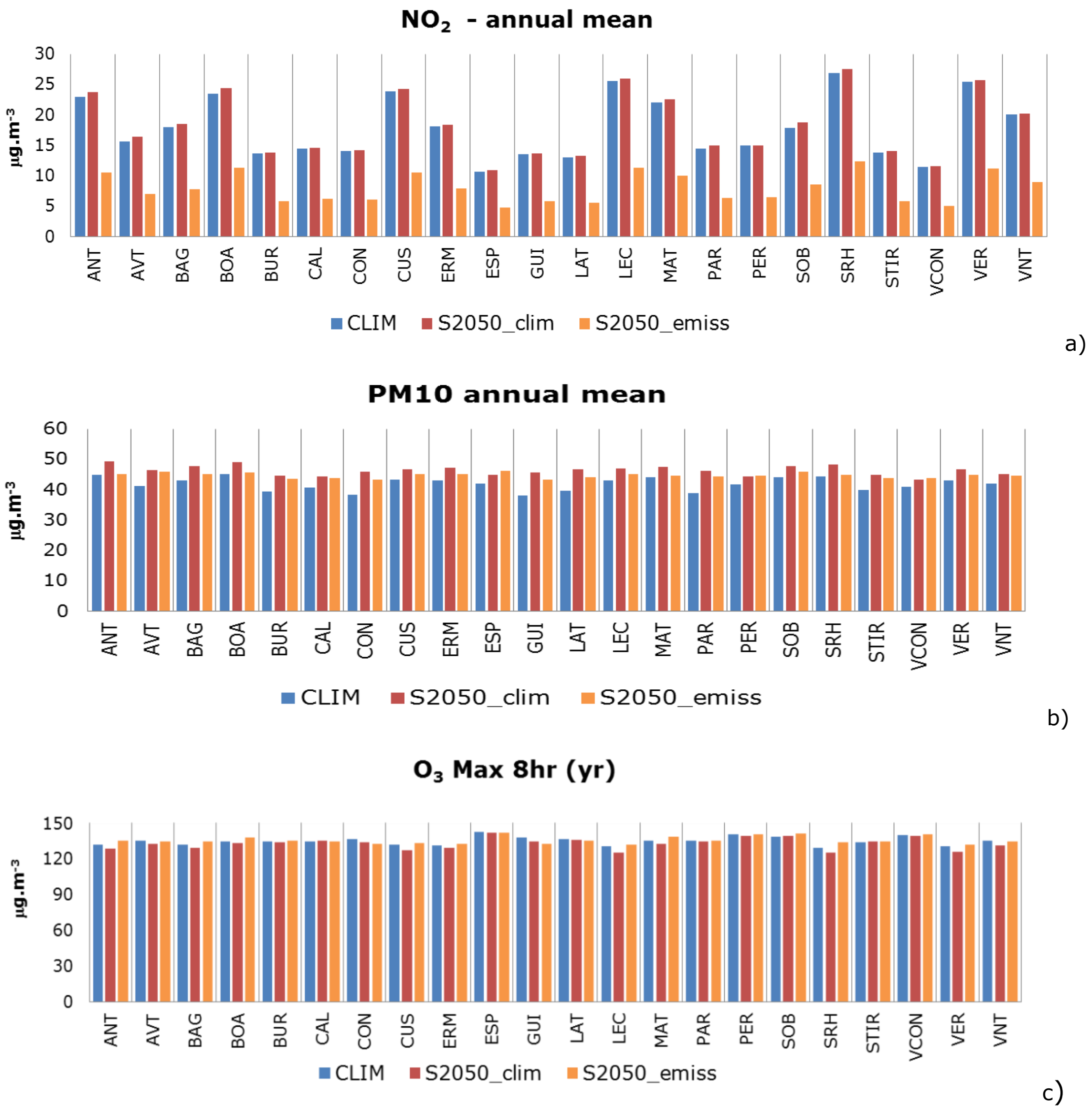


Figure 6.13 – NO_2 and PM10 annual mean and 8hr maximum O_3 values at the air quality stations located in the Porto urban area (D3).

For the analysed air quality stations, the annual mean values for NO_2 and PM10 are higher in the S2050_clim scenario and lower in the S2050_emiss. The NO_2 annual mean concentration values, in S2050_emiss scenario, are lower than in the CLIM scenario; on the other hand, the PM10 annual mean concentration values are higher in both 2050 scenarios than in the CLIM scenario. The maximum of daily 8-hr mean O_3 concentrations decreases in S2050_clim and increases in S2050_emiss. This could be justified by the fact that all of the air quality stations in D3 are in an urban environment. In the case of PM10 and O_3, in 2050, all the air quality stations are not fulfilling the current air quality standards (PM10 - annual mean higher than 40 $\mu g.m^{-3}$; O_3 – maximum daily 8-hr mean higher than 120 $\mu g.m^{-3}$), for both scenarios.

In order to obtain a detailed analysis of the air quality data distribution for each air quality station located in Porto urban area, a statistical analysis was performed for the different simulation scenarios (Figure 6.14 - Figure 6.16), including five statistical parameters: median, 5th percentile, 25th percentile, 75th percentile and 90th percentile.

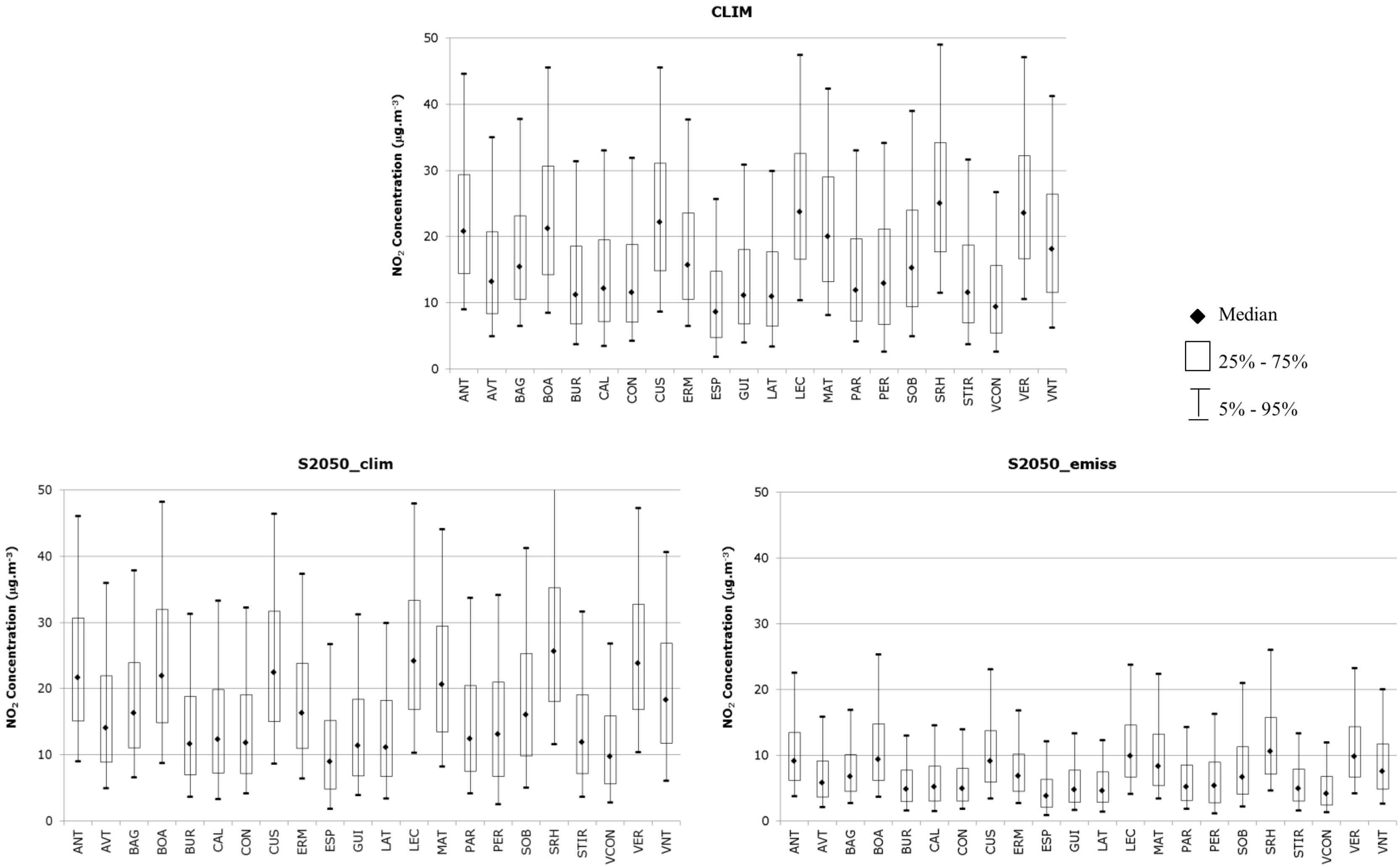


Figure 6.14 – Statistical analysis of the NO_2 data in each air quality station of D3 for all the simulations.

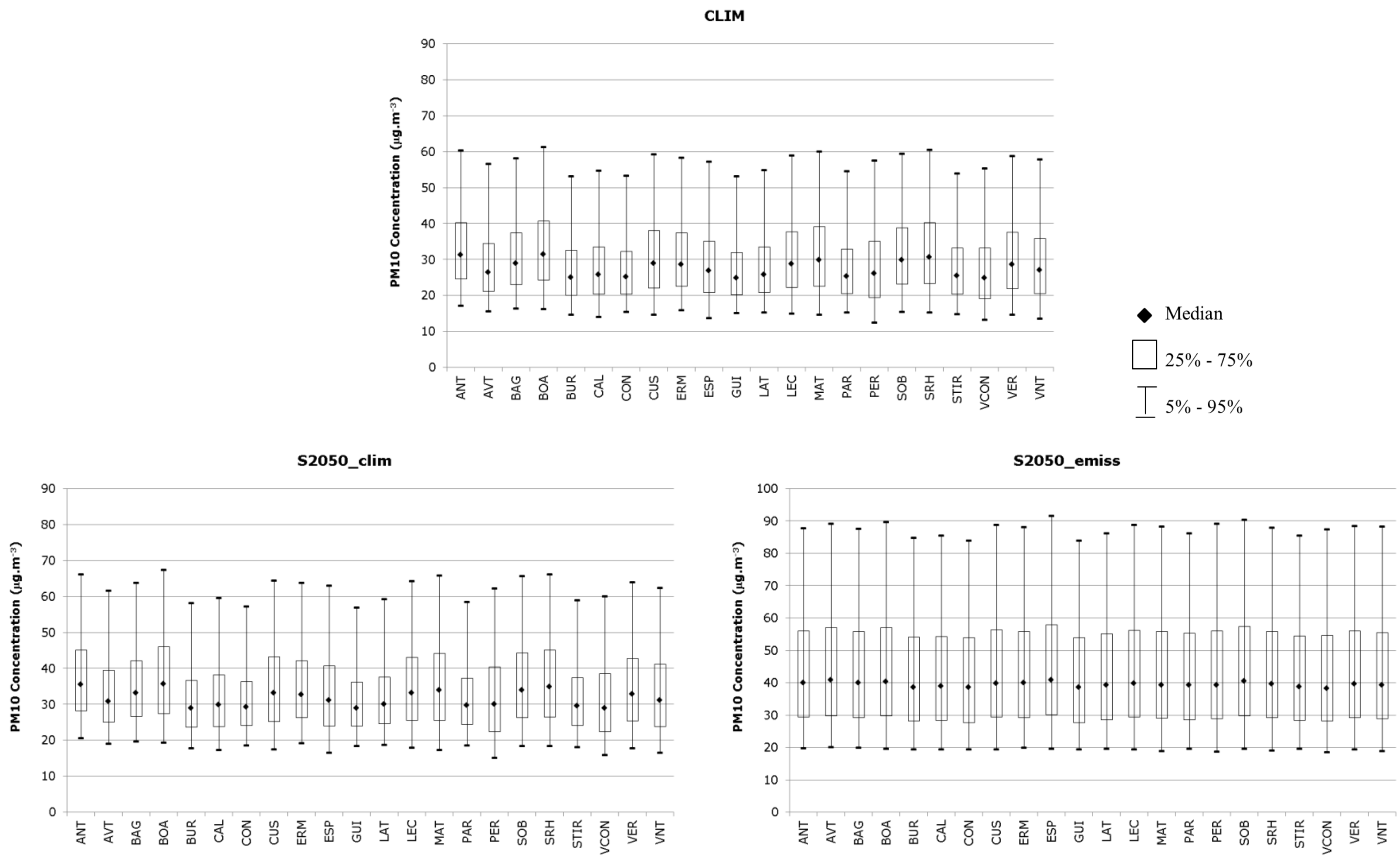


Figure 6.15 - Statistical analysis of the PM10 data in each air quality station of D3 for all the simulations.

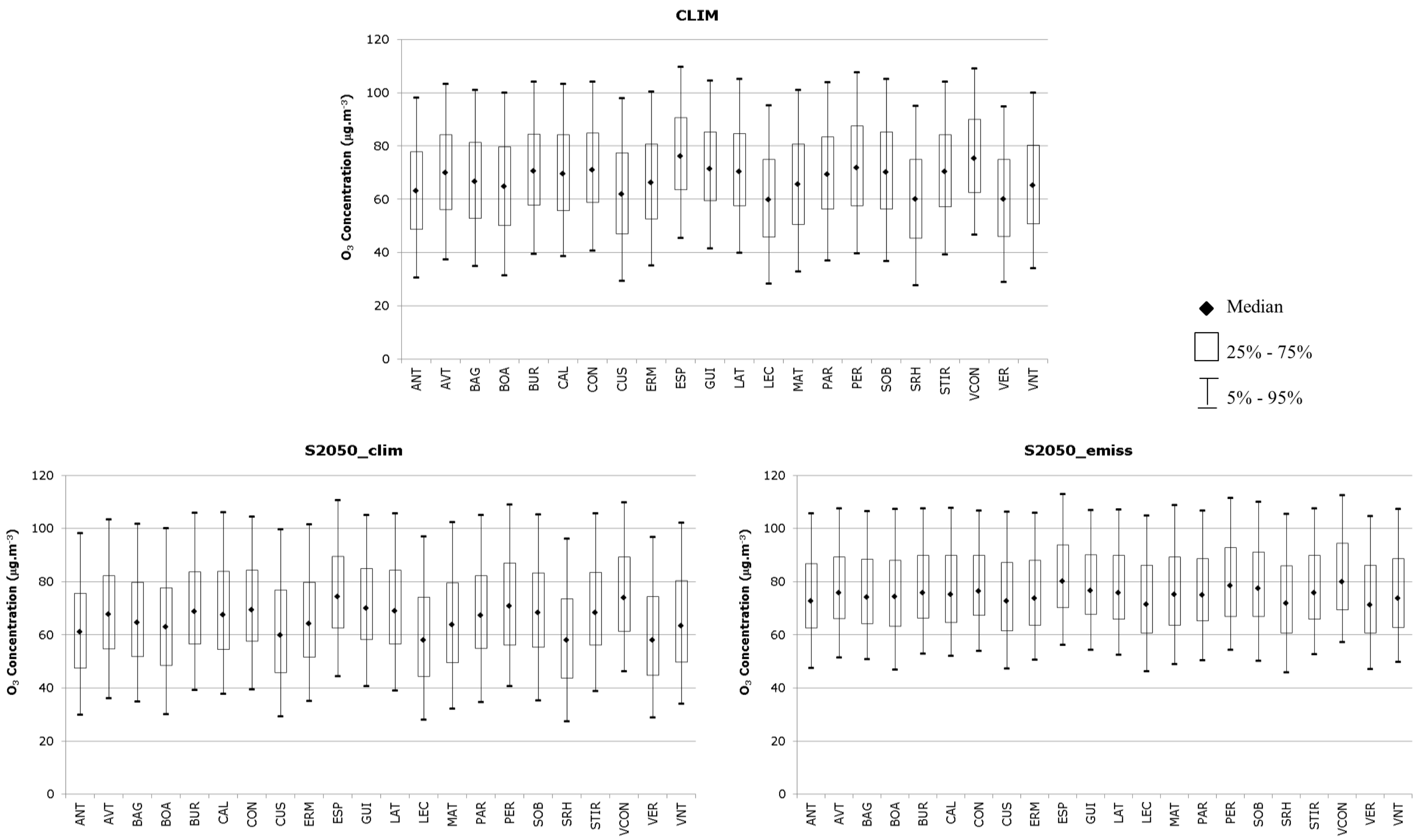


Figure 6.16 - Statistical analysis of the O_3 data in each air quality station of D3 for all the simulations.

In Figure 6.14 it is possible to verify that, besides the fact of the detected NO_2 annual mean increases in S2050_clim (Figure 6.13), the median NO_2 values and the other statistical parameters remain almost constant. This highlights that in 2050, considering only the effect of climate change, the hourly NO_2 levels will be constant; however the NO_2 annual mean will surpass the NO_2 annual mean limit value. As expected in S2050_emiss all the statistical parameters decrease.

For PM10, all the statistical parameters increase in both simulations for 2050; however in S2050_emiss the 75th percentile is above the PM10 daily LV (50 $\mu g.m^{-3}$), which will originate PM10 daily exceedances.

In S2050_clim simulation, the O_3 percentiles will increase in 2050, but will not be significantly. However, in S2050_emiss simulation, all the statistical parameters analysed will increase significantly. The results indicate that the O_3 concentrations will increase comparing with nowadays, namely the general/background O_3 concentrations will increase and therefore higher values will occur more often. This is a very important feature that highlights that important changes can be detected at the background O_3 levels and this may have medium to long term impacts that are not yet explored.

A detailed analysis of the Porto urban area in terms of air quality in 2050 was conducted (Figure 6.17).

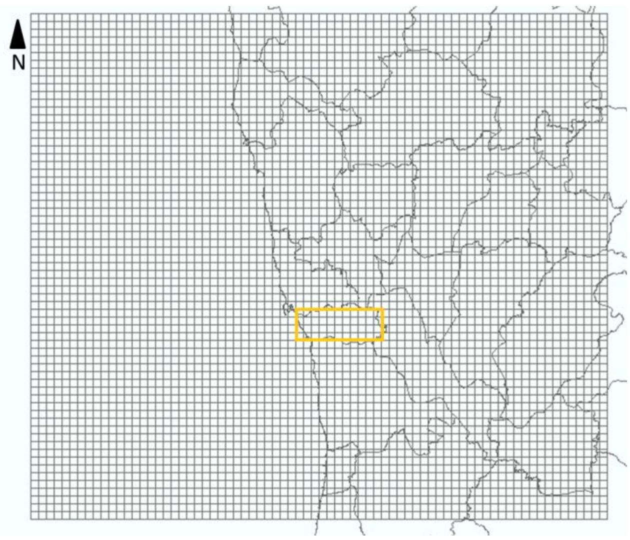
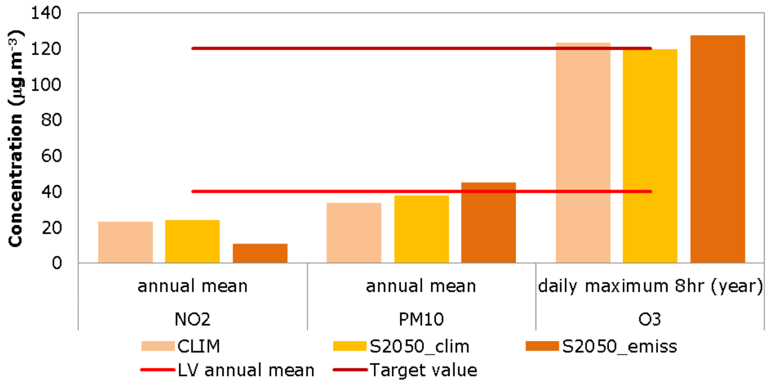


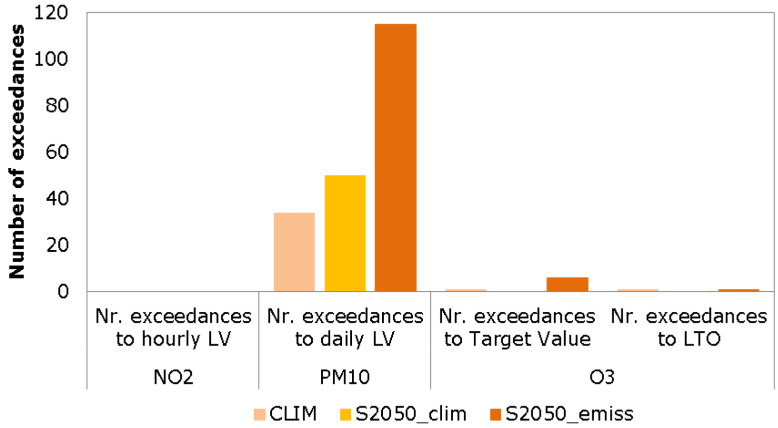
Figure 6.17 – Area selected from D3 to characterize the Porto urban area.

In order to deepen explore the main chances that may occur at Porto urban area, a restricted area over the innermost simulation domain has been delimited for a detailed analysis. This area was selected based on the statistical analysis of the representative time-series of each area, as defined and described in the Supporting material. The area, which only includes the Porto municipality, was selected to represent the Porto urban area, because when the area of influence was enlarged the concentrations were diluted, mainly for NO_2 and O_3 (Appendix D). Based on the air quality time series obtained within the delimited area a new time-series has been built based on the individual data. The synthetic hourly data, representative of the Porto urban area, was estimated based on the average of the hourly data of all the cells included in the restricted area.

Figure 6.18 presents the NO_2 and PM10 annual means and the O_3 daily maximum 8-hr for one year (Figure 6.18a) and the number of exceedances for the three pollutants considering the legislated parameters (Figure 6.18b).



a)



b)

Figure 6.18 – NO_2, PM10 annual mean and the O_3 daily maximum 8 hour (a) and the number of exceedances of NO_2, PM10 and O_3 (b).

From Figure 6.18 it is possible to conclude that in 2050 the Porto urban area will have air quality problems related with PM10 and O_3, where despite the fact that the annual mean and the daily maximum 8-hr, respectively, will not have a significant increase, the daily exceedances will increase in both pollutants.

The cumulative distribution of the NO_2 hourly data, PM10 daily data and O_3 daily maximum 8-hr data has been estimated for the Porto urban area (Figure 6.19).

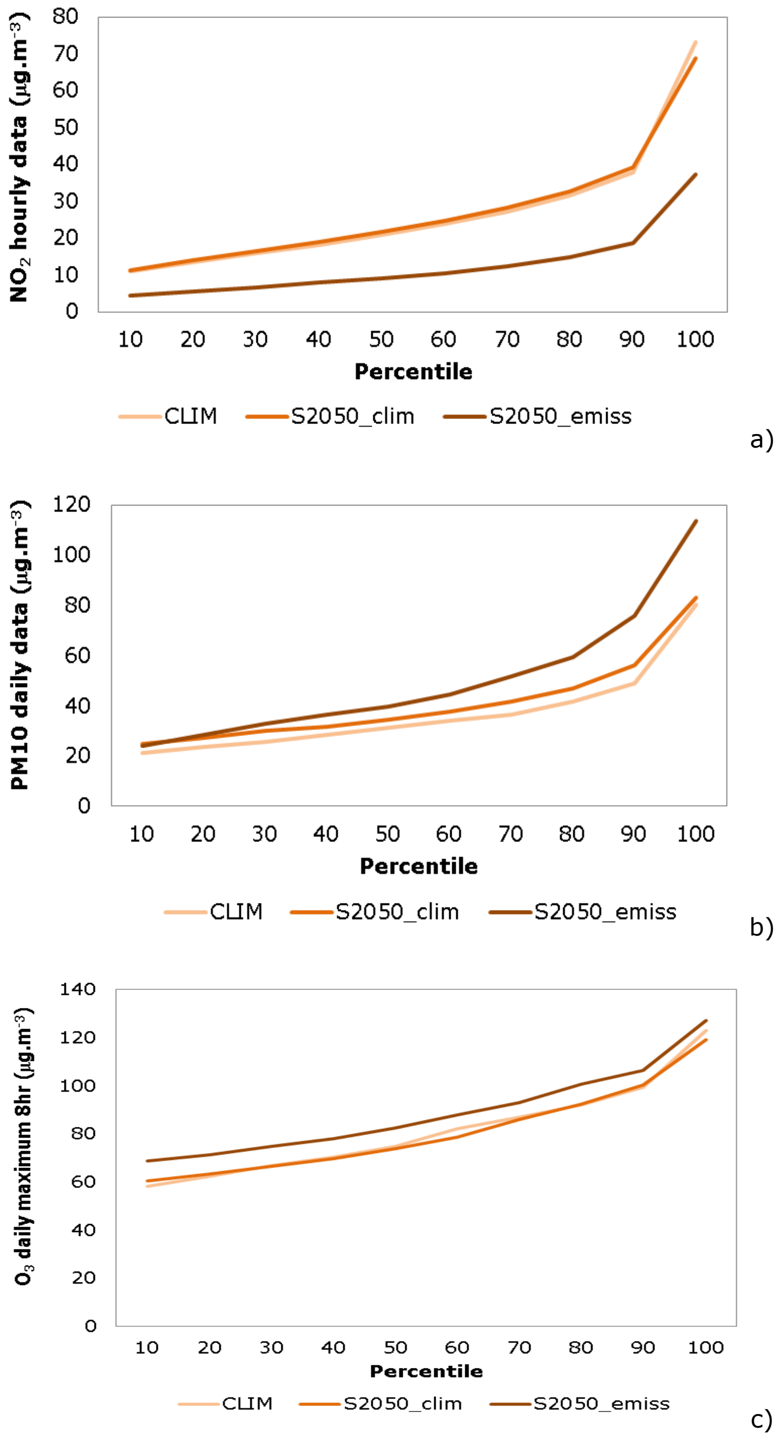


Figure 6.19 - Distribution of NO_2, PM10 and O_3 percentiles computed over Porto urban area for CLIM, S2050_clim and S2050_emiss.

Figure 6.19 supports the evidence about the increase of extreme values (minimum and maximum) in 2050 for O_3 and PM10. The results also point out that in Porto urban area the S2050_clim simulation has a similar behaviour to the CLIM simulation, which allows concluding that the behaviour of S2050_emiss is caused by the projected anthropogenic emissions. For NO_2, the difference between CLIM and S2050_clim is below 2 $\mu g.m^{-3}$. On the other hand, the difference between CLIM and S2050_emiss is always increasing, from 10th percentile to 100th percentile, with the highest change found between the 90th and 100th percentiles.

In the case of PM10, the differences between CLIM and S2050_clim are higher between the 70th and 90th percentiles (5 μg.m^{-3}) than between the 10th and 60th percentiles (3 μg.m^{-3}). Comparing CLIM and S2050_emiss, the differences reach 7 μg.m-3 between 20th and 50th percentiles, and 33 μg.m^{-3} between 70th and 100th percentiles. These results point out that under future climate there will be an increase in the extreme values.

As in NO_2, in O_3 the difference between CLIM and S2050_clim does not reach 2 μg.m^{-3}. On the other hand, the differences between CLIM and S2050_emiss start with 10 μg.m^{-3} in 10th percentile and decreases until 5 μg.m^{-3} in 60th percentile and then increase until 8 μg.m^{-3} in 80th percentile, decreasing again until the 100th percentile.

Also, an analysis of the extent and the intensity of the PM10 and O_3 exceedances, in S2050_clim and S2050_emiss, was performed to understand the behaviour of the synthetic air quality data of Porto urban area in 2050. Figure 6.20 shows the periods and duration of PM10 exceedances to the daily LV and Figure 6.21 shows the intensity of each exceedance compared with the values of the CLIM synthetic air quality data. Figure 6.22 shows the periods and duration of O_3 exceedances to the maximum daily 8-hr mean and Figure 6.23 shows the intensity of each exceedance compared with the values of the CLIM synthetic air quality data.

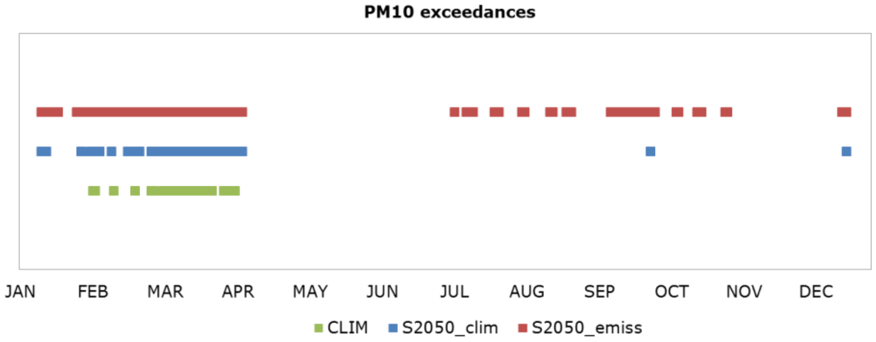


Figure 6.20 – PM10 exceedances to the daily LV in the synthetic data for CLIM, S2050_clim and S2050_emiss.

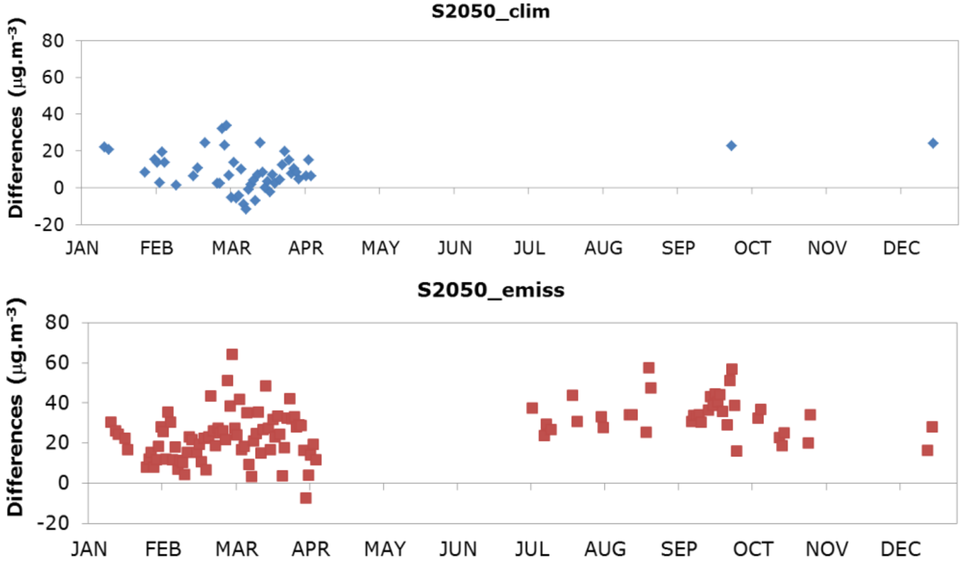


Figure 6.21 – PM10 differences between the exceedances from S2050_clim and S2050_emiss with the same values in CLIM ($\mu g.m^{-3}$).

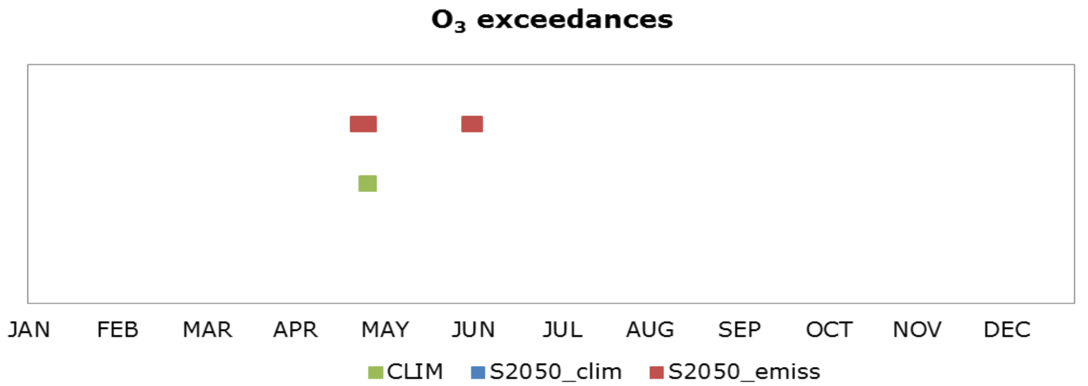


Figure 6.22 – O_3 exceedances to the maximum daily 8hr mean in the synthetic data for CLIM, S2050_clim and S2050_emiss.

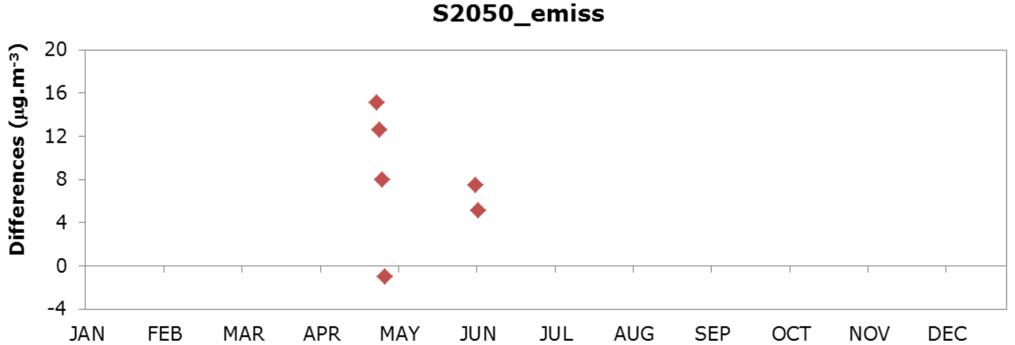


Figure 6.23 – O3 differences between the exceedances from S2050_emiss with the same values in CLIM (µg.m-3).

In 2050 the duration of the PM10 exceedances to the daily LV in the winter months will increase significantly, considering just the climate change effect (S2050_clim). Additionally, the concentration values of these exceedances will increase, reaching a 35 $\mu g.m^{-3}$ difference when compared to the historical simulation (CLIM). Adding the effects of the projected anthropogenic emissions for 2050, the duration of the PM10 exceedances to the daily LV in the winter months will increase, however the large modifications are related to the existance and duration of PM10 exceedances in the period from July to November. The intensity of the PM10 exceedances in S2050_emiss will increase significantly in comparison with CLIM, for both periods of the year, reaching almost 70 $\mu g.m^{-3}$ in the winter months and 60 $\mu g.m^{-3}$ from July to November.

Considering only the climate change effetcs, there will be no exceedances to the O_3 maximum daily eight hour mean. However, when the climate change effects is combined with the projected anthropogenic emissions, the only exceedance to the maximum daily eight hour mean that occurs in CLIM simulation (April) is increased to an exceedance with a duration of 4 days in 2050. Furthermore, a new period of exceedances to the O_3 maximum daily eight hour mean occurs in 2050 in July. The intensity of the exceedance in April do not increase comparing with historical data, still the intensity of the exceedances in the previous days increases, reaching almost 16 $\mu g.m^{-3}$.

As a final remark we may stress that not only the number and duration of PM10 and O_3 daily exceedances will increase in 2050, but also the intensity of the exceedances.

6.4 Summary and Conclusions

The main objective of this work is to understand how air quality will be in 2050 under the IPCC RCP8.5 climate change scenario in Portugal, and more specifically over Porto urban area. To achieve this goal the CAMx air quality model was applied to a 5-year historical (S2050_clim) period and to a 5-year future period (S2050_clim and S2050_emiss) for three domains. All these simulations were driven by WRF outputs for the corresponding periods.

The modelling system was evaluated for the year 2011 using the DELTA tool framework and the results point out that the model has a good performance in simulating concentrations of NO_2, O_3 and PM10, presenting a positive feedback/behaviour for all parameters in DELTA tool, both in terms of time or space.

Under future climate, considering the climate change effects alone (S2050_clim), the NO_2 and PM10 annual mean will increase in Portugal and in Porto municipality. In the case of O_3, the concentrations will increase in suburban areas and decrease in urban areas, such as Porto municipality. When, along with climate change effects, projected anthropogenic emissions are also considered (S2050_emiss) the NO_2 annual mean decreases significantly in Portugal and Porto urban area (around 50%), caused by the reduction of the projected emissions. The PM10 annual mean will increases over Portugal, probably because of the boundary conditions driven by MOZART, and will decreases in Porto urban area. The O_3 results are mainly caused by the reduction of ozone precursors, with higher reductions in urban areas and increases in the surrounding areas.

At the air quality stations located in the innermost domain, the NO_2 and PM10 annual mean are higher in S2050_clim scenario and lower in S2050_emiss. On the contrary, the maximum of daily 8-hr mean O_3 concentrations decreases in S2050_clim and increases in S2050_emiss.

All the analysis performed for both simulations for Porto urban area support that, for PM10 and O_3, there will be an increase in the occurrence, duration and intensity of extreme values, surpassing the annual legislated parameters and registering a higher number of daily exceedances.

These conclusions demonstrated the added value of studying the impacts of climate change on air quality at an urban scale. In terms of PM10 and O_3 concentration levels, the Porto urban area was better characterized with the results from the air quality simulation at a higher resolution.

The understanding of climate change impacts on air quality is essential to better assess the inter-relation between these topics. The Portuguese authorities and policy-makers should design and implement mitigation plans to improve air quality management that take into account climate change impacts.

Conclusions

7 Conclusions

The main goal of this work was to assess the impacts of climate change on air quality over Portugal in 2050, taking in consideration the RCP8.5 scenario. The work was organized in seven chapters, starting with the general introduction, which focus on the principal topics, climate change, air quality, their relationships and air quality modelling under climate change.

As was understood, despite the improvements that the EU is building over the last decades, air pollution continues to be a problematic environmental area. In Portugal eight air pollutants were analysed continuously (PM10, PM2.5, CO, O_3, NO_2, VOCs, SO_2 and C_6H_6) in the national air quality network. Three of these pollutants (NO_2, PM10 and O_3) are considered critical in terms of air quality, having exceedances every year, since 2002.

Air quality plans have been already developed for NO_2 and PM10, and a new AQP for O_3 is being prepared. From the two AQPs for the Northern region it is possible to verify that in terms of sources contribution, for NO_2, was evident the weight of traffic (around 32%). For PM10, several sectors have contributed to the concentration levels registered in the air quality stations. Based on this information the most appropriate measures to reduce the concentration values of these pollutants were selected. Some measures are already implemented for PM10 and are defined for NO_2, but it will be necessary to define and to implement new measures for the O_3 precursors.

However, air pollution results from the combination of high emissions and unfavorable weather conditions, where the strong linkage between weather conditions and pollutant levels can obscure the effects of changing emission levels over time. By the application of a KZ filter to the air quality (NO_2, O_3 and PM10) and meteorological datasets (T, Td, U and V components) for the period 2002-2012 was verified how the air pollutant concentrations were affected by meteorological variables. The conclusion allows understanding that future air pollutants concentrations will be affected by climate change. From the obtained results, NO_2 and PM10 were not as weather-dependent as O_3 for the different analysed components. However, the method is adequate for determining the meteorological variables that mostly influence NO_2 and PM10. The analysis showed that O_3 concentration has a statistically significant relationship to temperature for most of the components. For NO_2 and PM10, a meteorological variable with higher influence was not identified; nonetheless all the meteorological variables have some influence in the air quality values, depending on the urban area analysed.

The fact that there are critical pollutants in Portugal and that they are affected by some meteorological variables, strengthen the importance to study the impacts of climate change on air quality in Portugal and in Porto urban area. In this sense, the air quality modelling system (WRF-CAMx) was applied to evaluate air quality in Portugal and in Porto metropolitan area under the IPCC RCP8.5 scenario.

One of the main WRF-CAMx required inputs are the emissions for the current period and for 2050. The current emissions were estimated for Portugal with a higher detailed disaggregation to improve the performance of the air quality simulations. The results showed that the national spatial distribution of the SNAP7 emissions is improved when the traffic sector is considered as a line source. This fact improves the accuracy of the air quality simulations, since traffic is one of the major sources for the national air quality concentrations. In order to estimate the emissions for 2050 a new model was developed (EmiPro-RCP) under the RCPs scenarios. This model is only based on the RCP scenarios and do not consider socioeconomic and land-use changes of the study region. However, allows a detailed analysis of the trends and of the spatial patterns of the future national emissions based on the RCP scenarios. One of the main innovations of this thesis is the estimation of the emissions under the RCP scenarios because allows keeping the consistency of the air quality simulations under a climate change scenario, in terms of anthropogenic emissions. By the application of EmiPro-RCP model, it was found that the emissions in Portugal tend to decrease in all the sectors, with the exception of NH_3 that tend to increase. Besides the fact that this model estimated the emissions to be used in the air quality simulations under a climate change scenario, it is also an important and user-friendly tool that can be used in upcoming studies of this type.

A sensitivity study to the air quality modelling system (WRF-CAMx) was accomplished by performing: simulations using different combinations of physics parameterizations and a new land use (based in CLC2006); simulations using the flexi-nesting capability; and finally simulations using different emission data. From this analysis was defined a general model setup for Portugal and Porto urban area to be used in air quality simulations under climate change. This analysis revealed that WRF-CAMx is an air quality modelling system that demonstrates a good performance when is applied over Portugal and over Porto urban area and for this reason is a fundamental and helpful tool to be use in air quality studies.

The validation of the WRF-CAMx modelling system with the model setup selected for Portugal and Porto urban area, was performed for the year 2011. In general, the results from this simulation showed that the model had a good performance in simulating concentrations of NO_2, O_3 and PM10, presenting a positive feedback for all parameters in DELTA tool, both in terms of time or space. Hereupon, this air quality modelling system is an available tool to achieve the main purpose of this work, assess the impact of climate change on air quality over Portugal and Porto urban area.

Three simulations with WRF-CAMx modelling system driven by the MPI-ESM-LR model were performed to achieve the main outcome of this work. The first simulation was for the historical period and the other two for 2050 future period. The difference between these two last simulations is related with the use of current and projected emissions. The simulations were performed for a 5 years period, since is a period long enough to obtain a climate signal. The results point out that if the anthropogenic emissions are kept the same in 2050, the annual mean concentrations of NO_2 and PM10 will increase in Portugal. When is considered the projected anthropogenic emissions, besides the climate change effects, the annual mean concentrations of NO_2 decrease significantly in Portugal and Porto urban area; on the contrary, the annual mean PM10 concentrations increases in Portugal and decrease in Porto urban area. In both simulations the O_3 results are mainly caused by the reduction of ozone precursors, getting the higher reductions in urban areas and increases in the surrounding areas. All the analysis performed for both simulations for Porto urban area support that, for PM10 and O_3, there will be an increase in the occurrence of extreme values, surpassing the annual legislated parameters and having more daily exceedances. In terms of PM10 and O_3 concentration levels, the Porto urban area was better characterized with the results from the air quality simulation at a higher resolution, which explain the importance of simulating air quality under a climate change scenario at a higher resolution. The main outcome of this work is that the 2050 air quality in Portugal and in Porto urban area will be affected by climate change and also by the projected anthropogenic emissions. The combination of these two factors will lead, in 2050, to and improvement of the NO_2 national air quality levels, but deterioration of the PM10 and O_3 national air quality levels.

WRF-CAMX air quality modelling system application has proved to be fundamental to investigate the impacts of climate change and projected air pollutant emissions on air quality levels. However, for simulations under climate change scenarios should consider other pollutant emissions (GHGs) to improve the interaction of these pollutants with common air pollutants.

The application of more than one climatic scenario allows a better characterization of the range of possible changes that can be detected in future. An ensemble of the results from the several possible scenarios may give important information about uncertainty analysis and promote a better characterization of the air quality in Portugal.

Also, it will be important to include in future scientific studies the human exposure in order to understand how the new air pollutant concentrations will influence human health.

Finally, this work represents an important effort to assess air quality at regional and urban scales in Portugal in 2050, being the main conclusion that in 2050 the air quality problems in Portugal and in Porto urban area, based on RCP8.5 scenario, will persist, namely for PM10 and O_3. This conclusion is a strong advice to Portuguese authorities and policy-makers to design and implement mitigation plans to improve air quality management in Portugal.

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Appendices

Appendices

Appendix A - Classification of the air quality stations in Mainland Portugal (short name, type of surrounding environment and influence).

Appendix B – Spatial distribution of the emission value by municipality and sub-municipality for all the air pollutants analysed

Appendix C - Delta tool plots for CAMx (D1, D2 and D3) for NO_2, O_3 and PM10.

Appendix D – Statistical analysis of air quality data in each selected area.

Appendix A

Appendix A – Classification of the air quality stations in Mainland Portugal (short name, type of surrounding environment and influence).

Table A.1 presents the short name of the air quality stations in Mainland Portugal and classify each one by type of surrounding environment and influence.

Table A.1 – Air quality stations classification in Mainland Portugal.

Air quality station	Short name	Environment	Influence
Afonso III	AFSIII	urban	traffic
Águas Santas	AST	urban	traffic
Alfragide/Amadora	ALF	urban	background
Alto Seixalinho	SEIX	urban	traffic
Alverca	ALV	urban	background
Antas	ANT	urban	traffic
Arcos	ARC	urban	background
Av. Casal Ribeiro	ACR	urban	traffic
Av. da Liberdade	AVL	urban	traffic
Av. Fernão Magalhães	COI-FM	urban	traffic
Av.24-Espinho	ESP	urban	traffic
Aveiro	AVE	urban	traffic
Baguim	BAG	urban	traffic
Beato	BEA	urban	background
Benfica	BEN	urban	traffic
Boavista	BOA	urban	traffic
Burgães	BUR	urban	background
Calendário	CAL	suburban	background
Camarinha	CAM	urban	background
Cascais-Mercado	CASC	urban	traffic
Cent.Lacticínios	CM	urban	background
Cerro	CER	rural	background
Chamusca	CHA	rural	background
Chelas	CHE	urban	background
Circular Sul	CSUL	urban	traffic
Custóias	CUS	suburban	background
David Neto	DVN	urban	traffic
Entrecampos	ENT	urban	traffic
Ermesinde	ERM	urban	background
Ervedeira	ERV	rural	background
Escavadeira	ESC	urban	industrial
Espinho	ESP	urban	traffic
Estarreja/Avanca	EST	rural	background
Estarreja/Teixugueira	TEI	suburban	industrial
Fernando Pó	FPO	rural	background
Fidalguinhos	FID	urban	background

Formosa	FOR	urban	traffic
Fornelo do Monte	FRN	rural	background
Fundão	FUN	rural	background
Guimarães-Centro	GUI	urban	traffic
Horto	HRT	suburban	background
Hospital Velho	LNH	urban	traffic
Ílhavo	ILH	suburban	background
Inst. Geof. de Coimbra	IGEO	urban	background
Joaquim Magalhães	JMG	urban	background
Lamas de Olo	OLO	rural	background
Laranjeiro	LAR	urban	background
Lavradio	LAV	urban	industrial
Leça do Balio	LEC	suburban	background
Loures-Centro	LOU	urban	background
Lourinhã	MVE	rural	background
Malpique	MAL	urban	background
Matosinhos	MAT	urban	traffic
Mem Martins	MEM	urban	background
mindelo	MCH	suburban	background
Monte Chãos	MARQ	suburban	industrial
Monte Velho	MVE	rural	background
Montemor-o-Velho	MOV	rural	background
Município	MUN	urban	traffic
Odivelas-Ramada	ODI	urban	traffic
Olivais	OLI	urban	background
Paio Pires	PP	suburban	background
Paredes-Centro	PAR	urban	traffic
Perafita	PER	suburban	industrial
Pontal	PONT	urban	background
Quebedo	QUE	urban	traffic
Quinta do Marquês	QMAR	urban	background
Reboleira	REB	urban	background
Restelo	RES	urban	background
Rua da Prata	RPRA	urban	traffic
Senhora da Hora	SRH	urban	traffic
Senhora do Minho	SMIN	rural	background
Sobreiras	SOB	urban	background
Sonega	SON	rural	industrial
St. Cruz de Benfica	BEN	urban	traffic
St. Tirso	STIR	urban	background
Terena	TER	rural	background
Vermoim	VER	urban	traffic
Vila do Conde	VCON	urban	traffic
Vila Nova da Telha	VNT	suburban	background

Appendix B

Appendix B - Spatial distribution of the emission value by municipality and sub-municipality for all the air pollutants analysed.

Figure A.1 to Figure A.8 present the spatial distribution of the emission values of all the pollutants for all the SNAPs.

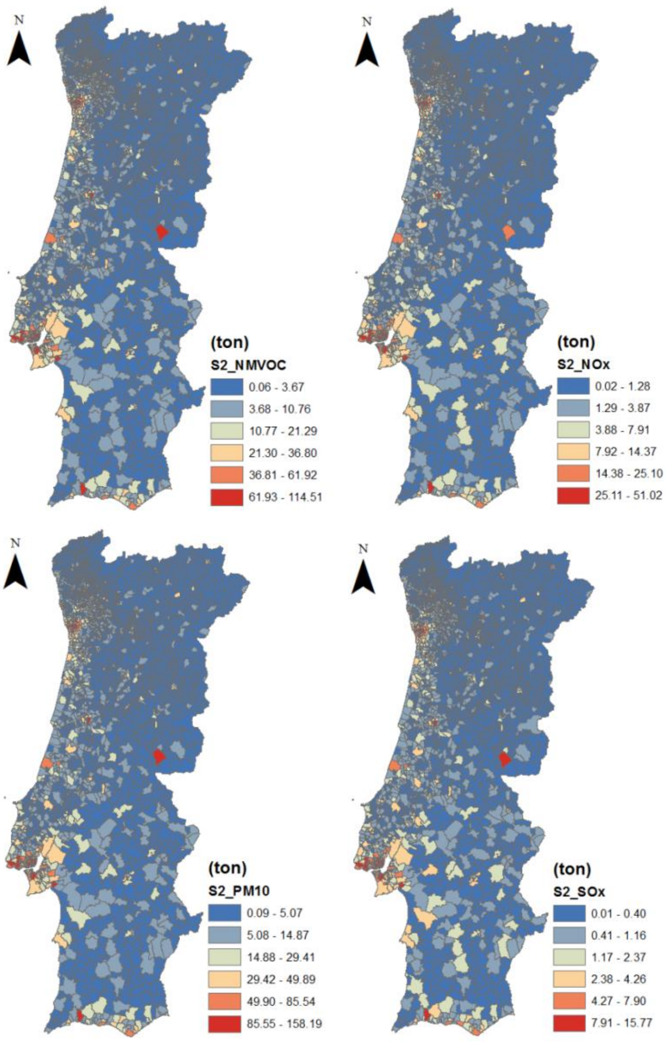


Figure A.1 – Spatial distribution SNAP2 for NMVOC, NOx, PM10 and SOx.

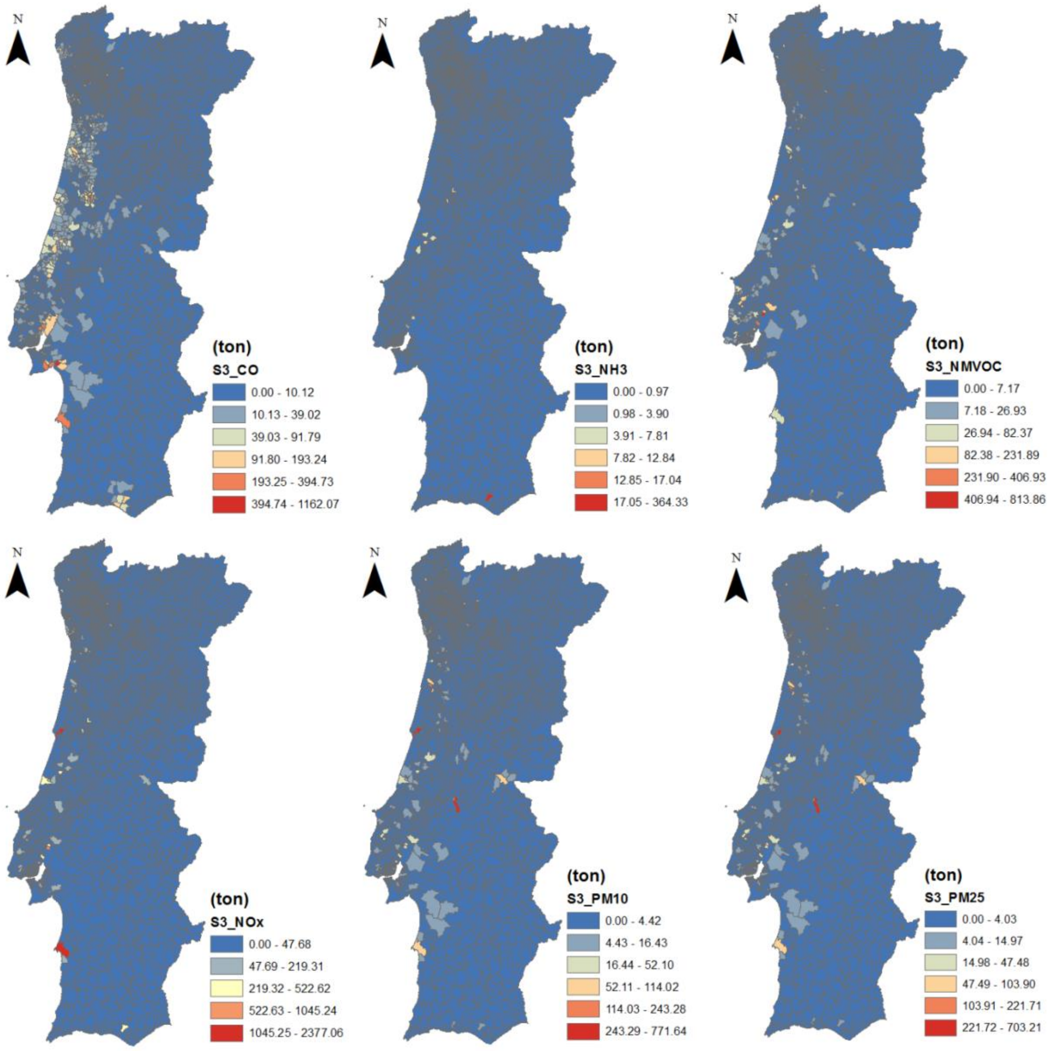


Figure A.2 – Spatial distribution SNAP3 for CO, NH_3, NMVOC, NOx, PM10 and PM2.5.

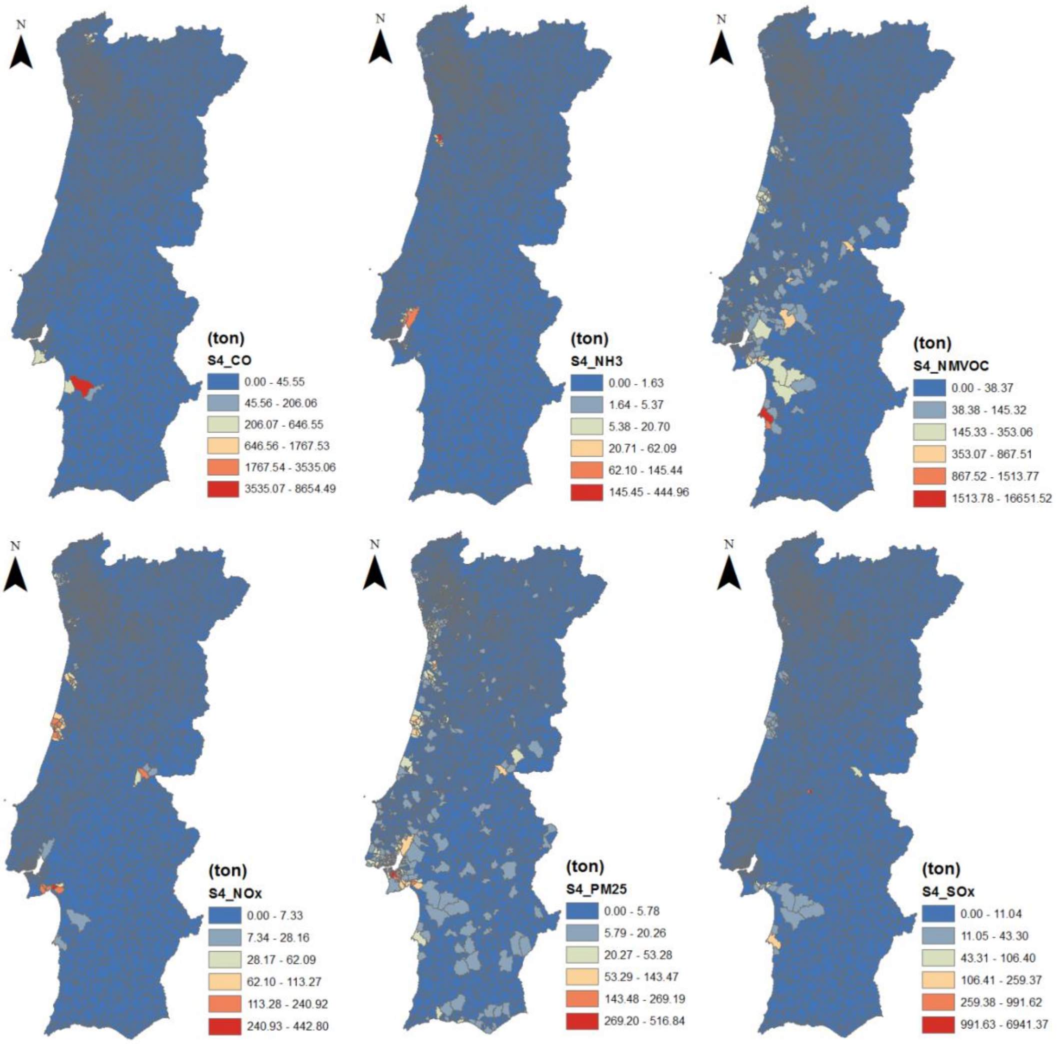


Figure A.3 – Spatial distribution SNAP4 for CO, NH_3, NMVOC, NOx, PM2.5 and SOx.

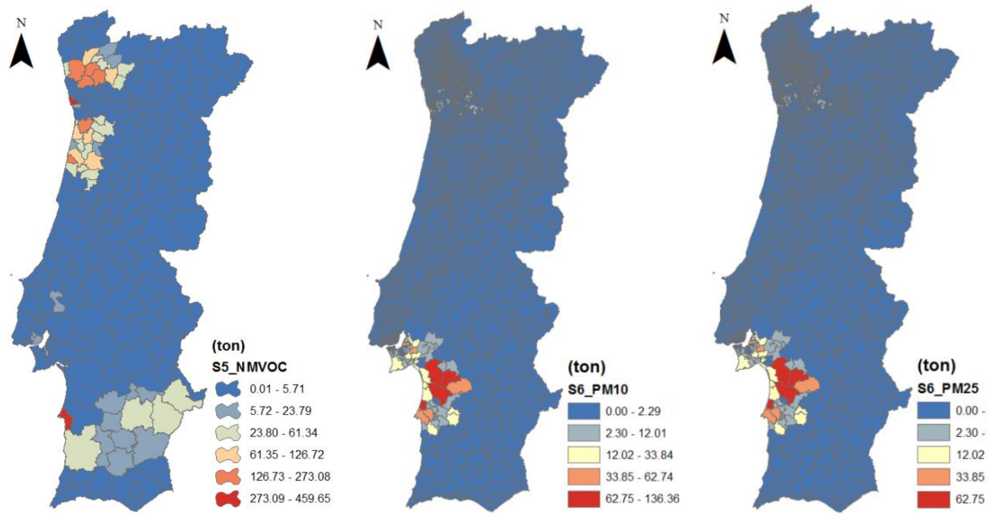


Figure A.4 – Spatial distribution SNAP5 and SNAP6 for NMVOC, PM10 and PM2.5.

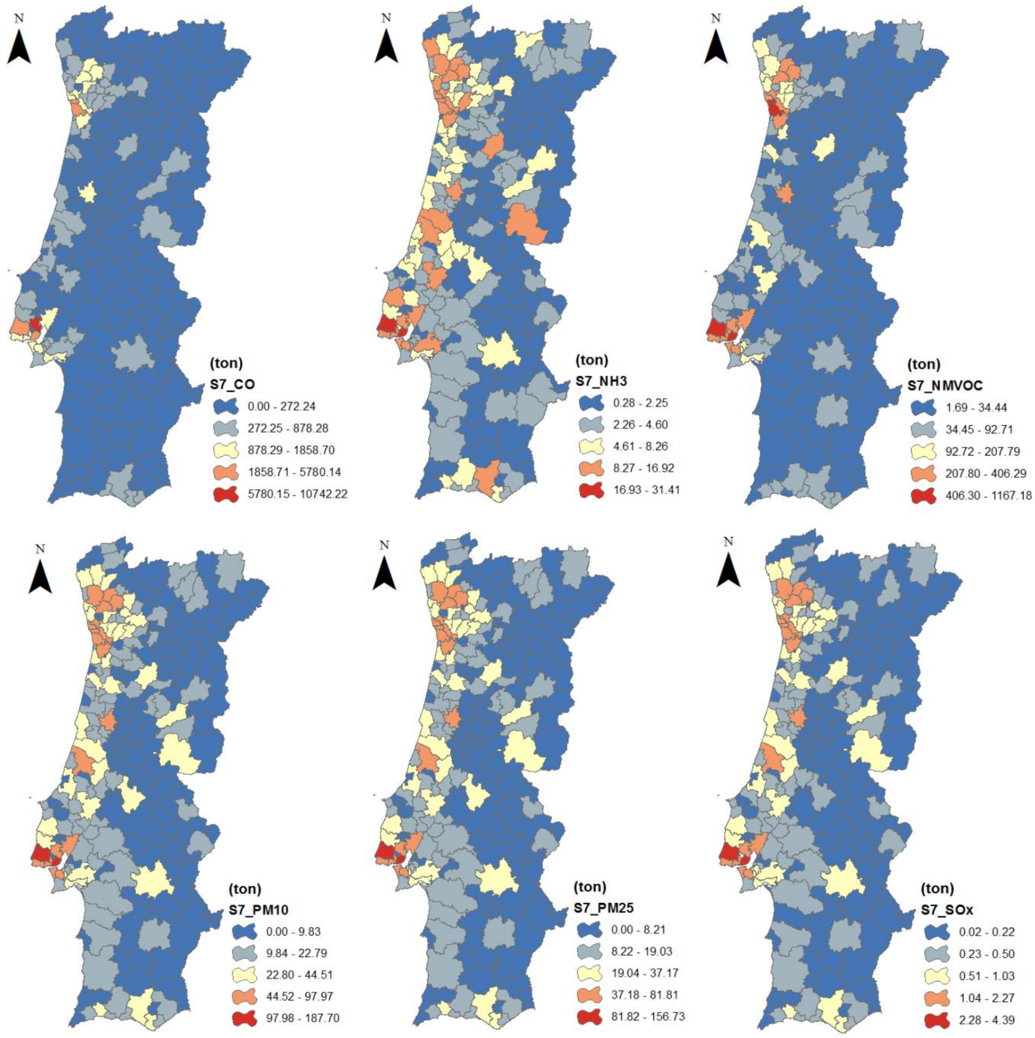


Figure A.5 – Spatial distribution SNAP7 for CO, NH_3, NMVOC, PM10. PM2.5 and SOx.

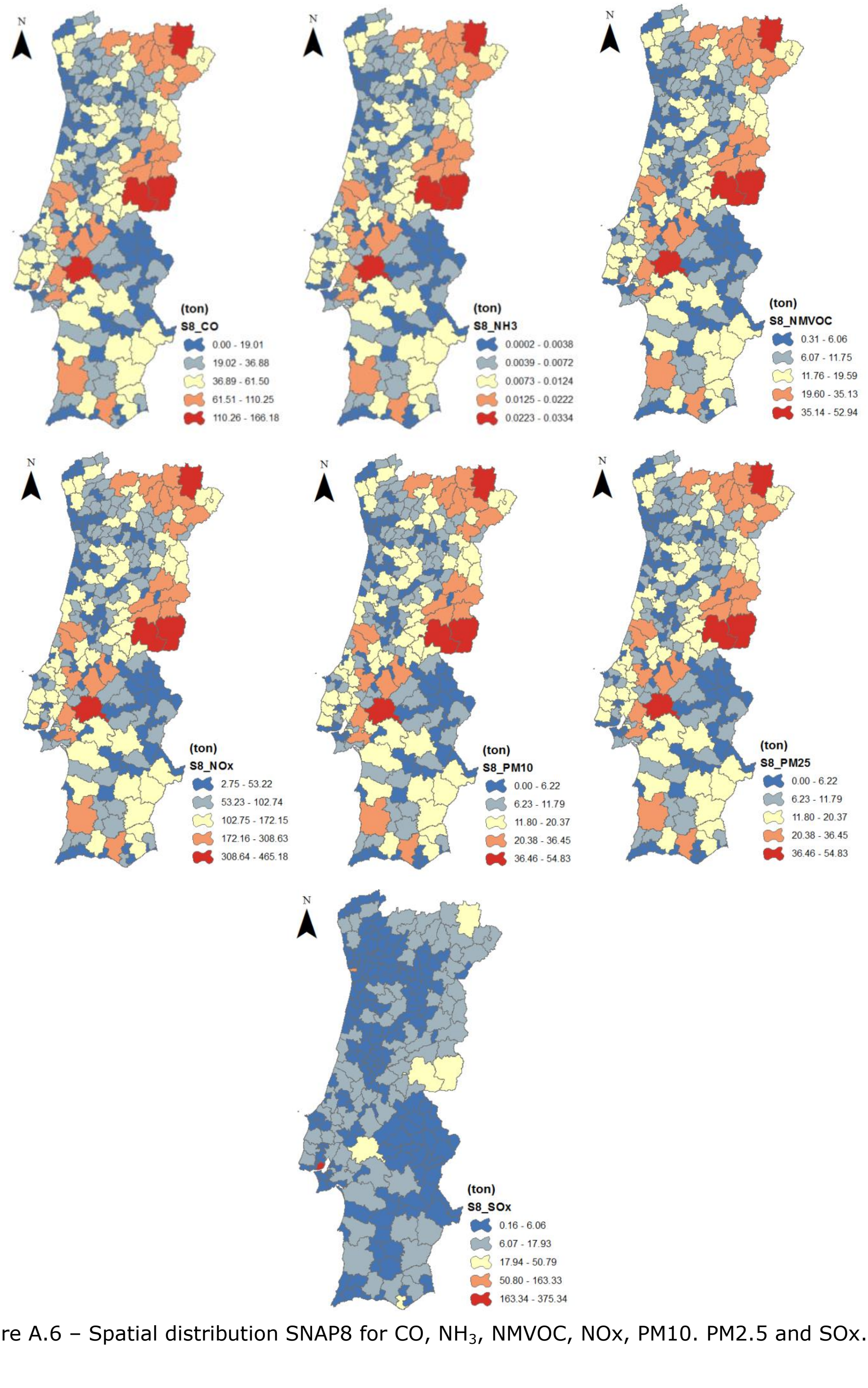


Figure A.6 – Spatial distribution SNAP8 for CO, NH_3, NMVOC, NOx, PM10. PM2.5 and SOx.

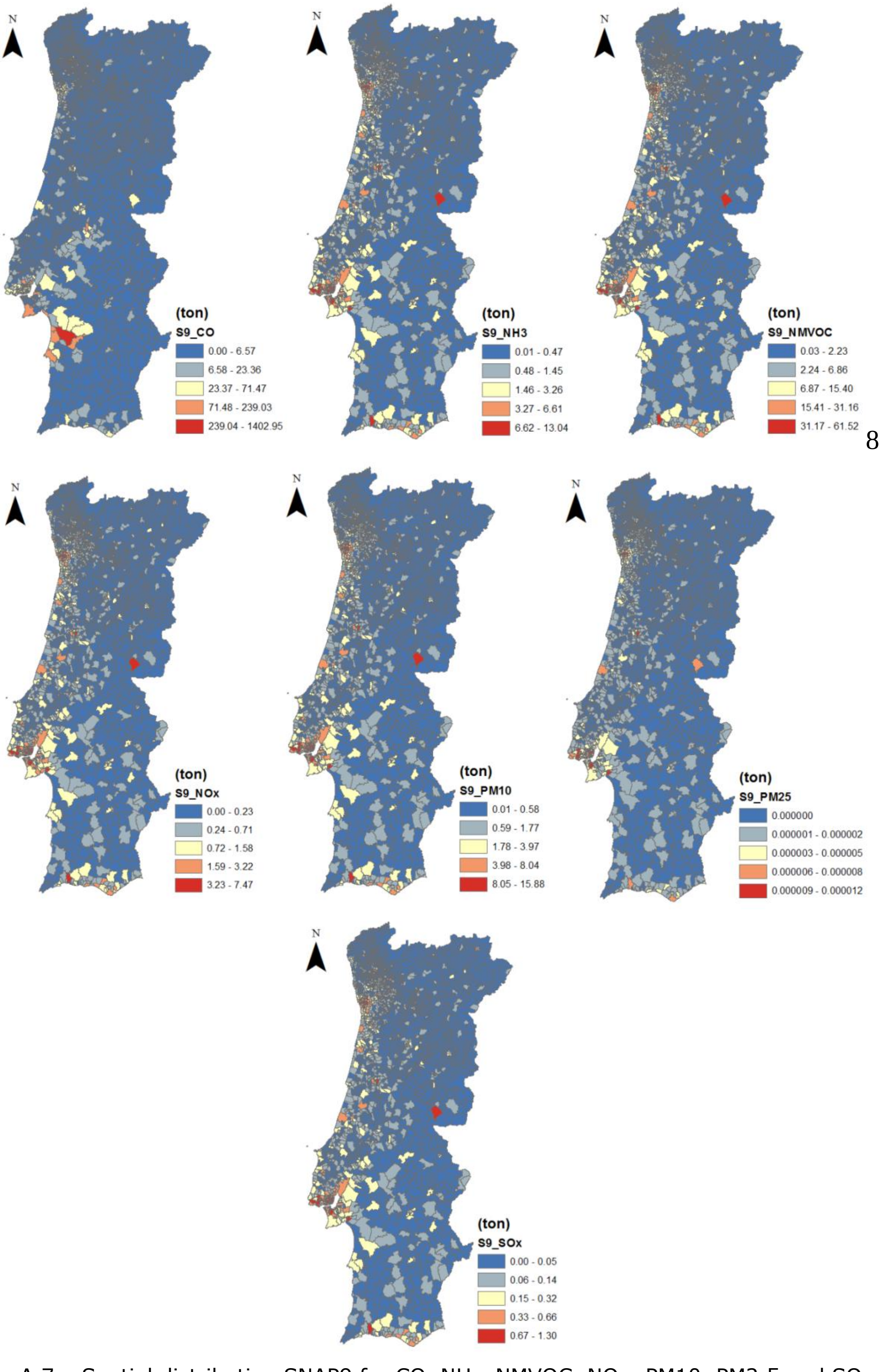


Figure A.7 – Spatial distribution SNAP9 for CO, NH_3, NMVOC, NOx, PM10. PM2.5 and SOx.

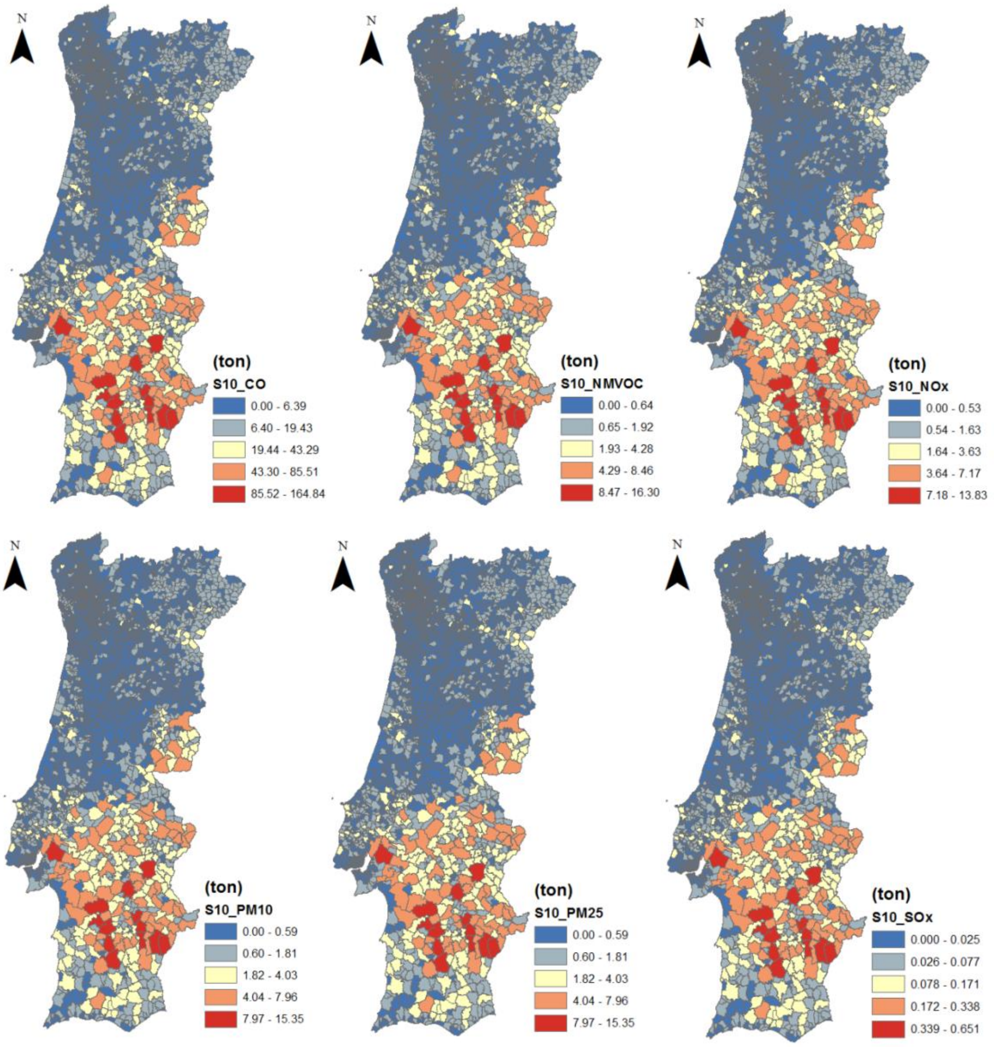


Figure A.8 – Spatial distribution SNAP10 for CO, NMVOC, NOx, PM10. PM2.5 and SOx.

Appendix C

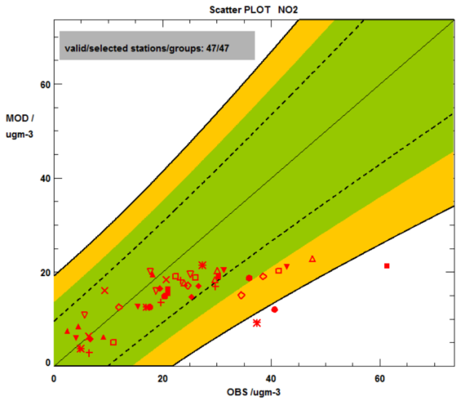
Appendix C - Delta tool plots for CAMx (D1, D2 and D3) for NO_2, O_3 and PM10.

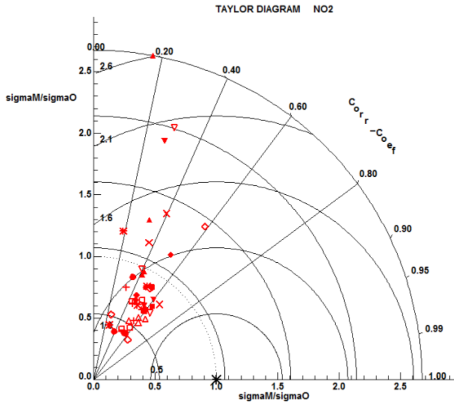
Table A.2 to Table A.4 present the other plots from the DELTA tool on the evaluation of BASE simulation.

Table A.2 – DELTA tool plot for CAMx D1 for NO_2, O_3 and PM10.

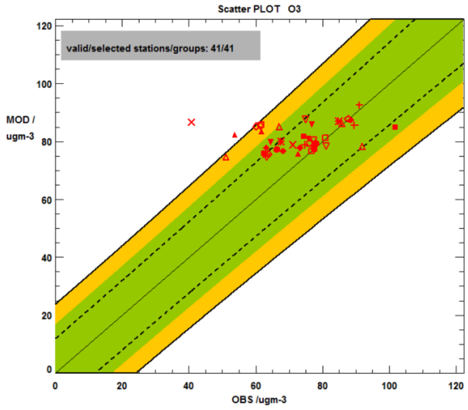
D1

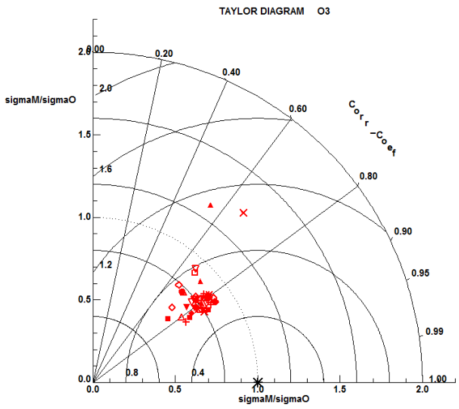
NO_2



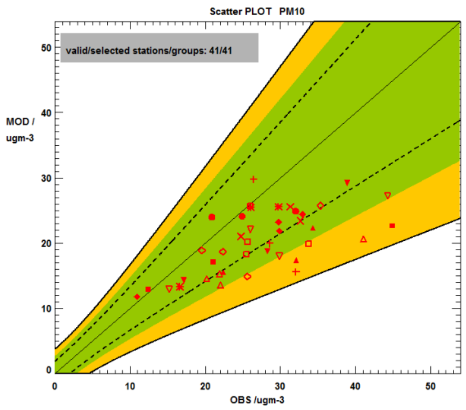


O_3





PM10



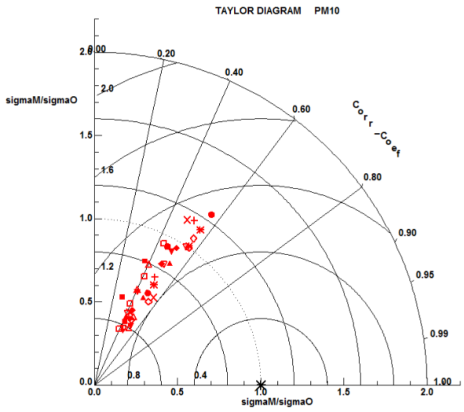
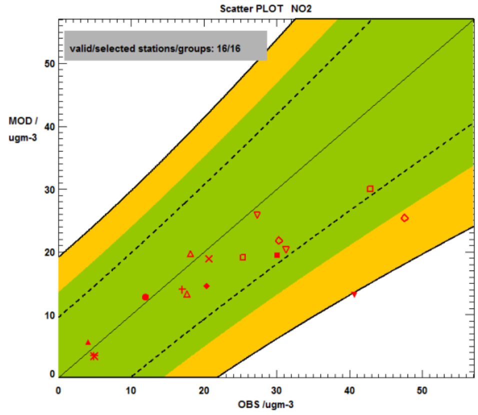
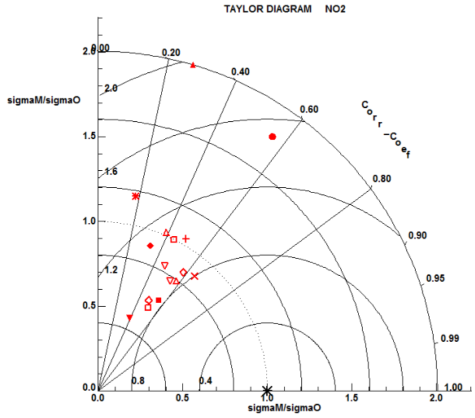


Table A.3 – DELTA tool plot for CAMx D2 for NO_2, O_3 and PM10.

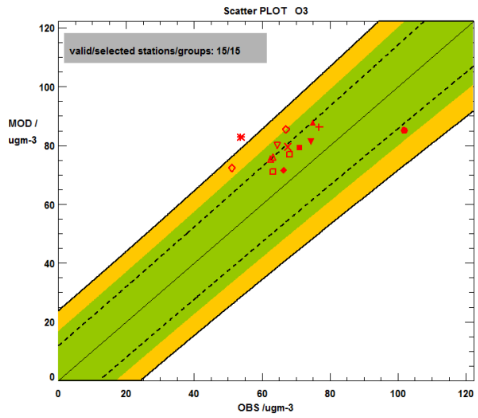
D2

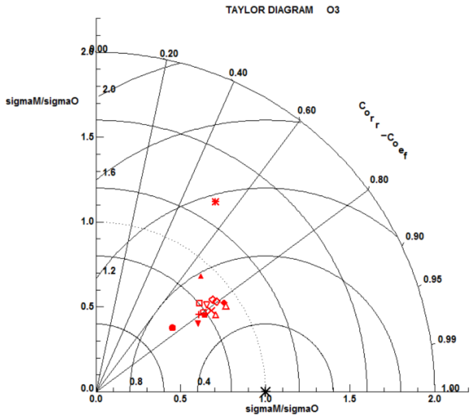
NO_2



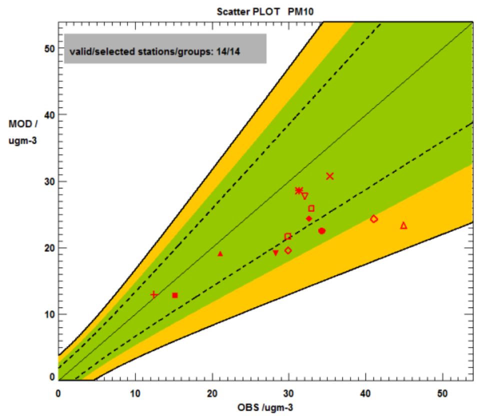


O_3





PM10



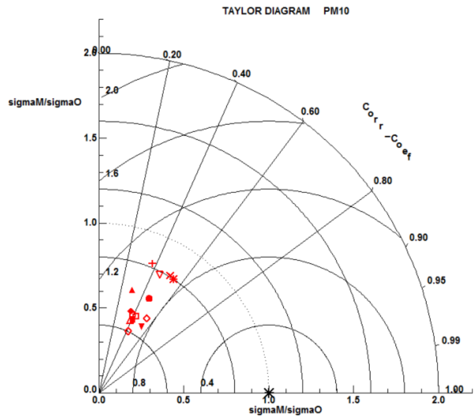
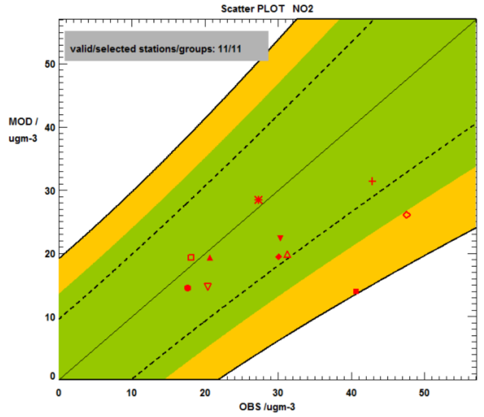
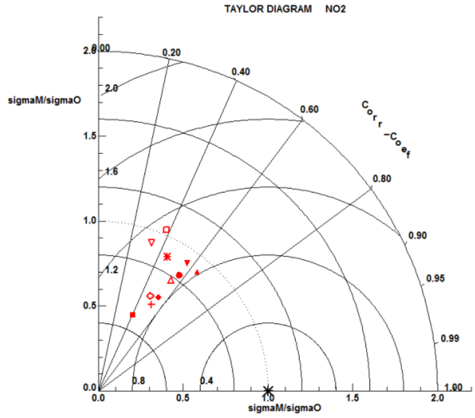


Table A.4 – DELTA tool plot for CAMx D3 for NO_2, O_3 and PM10.

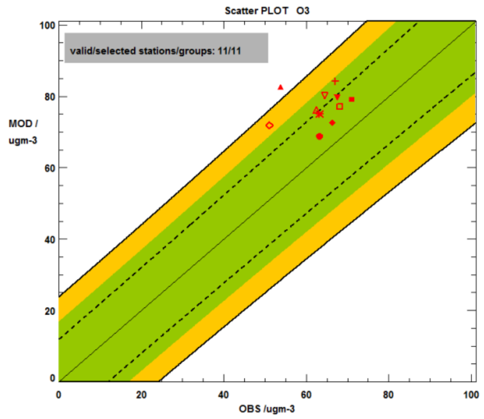
D3

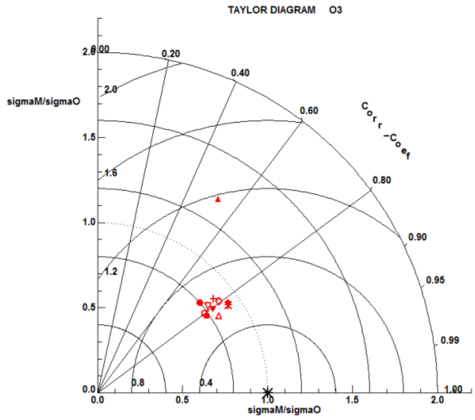
NO_2



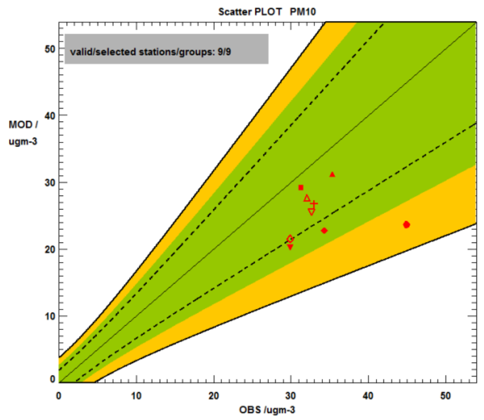


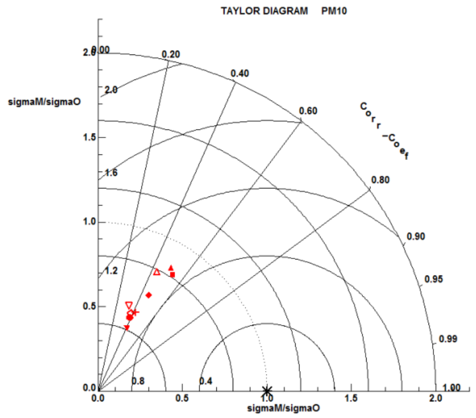
O_3





PM10





Appendix D

Appendix D – Statistical analysis of air quality data in each selected area.

Figure A.9 shows the areas to be statistical analysed in order to select which area has a synthetic data representative of the area.

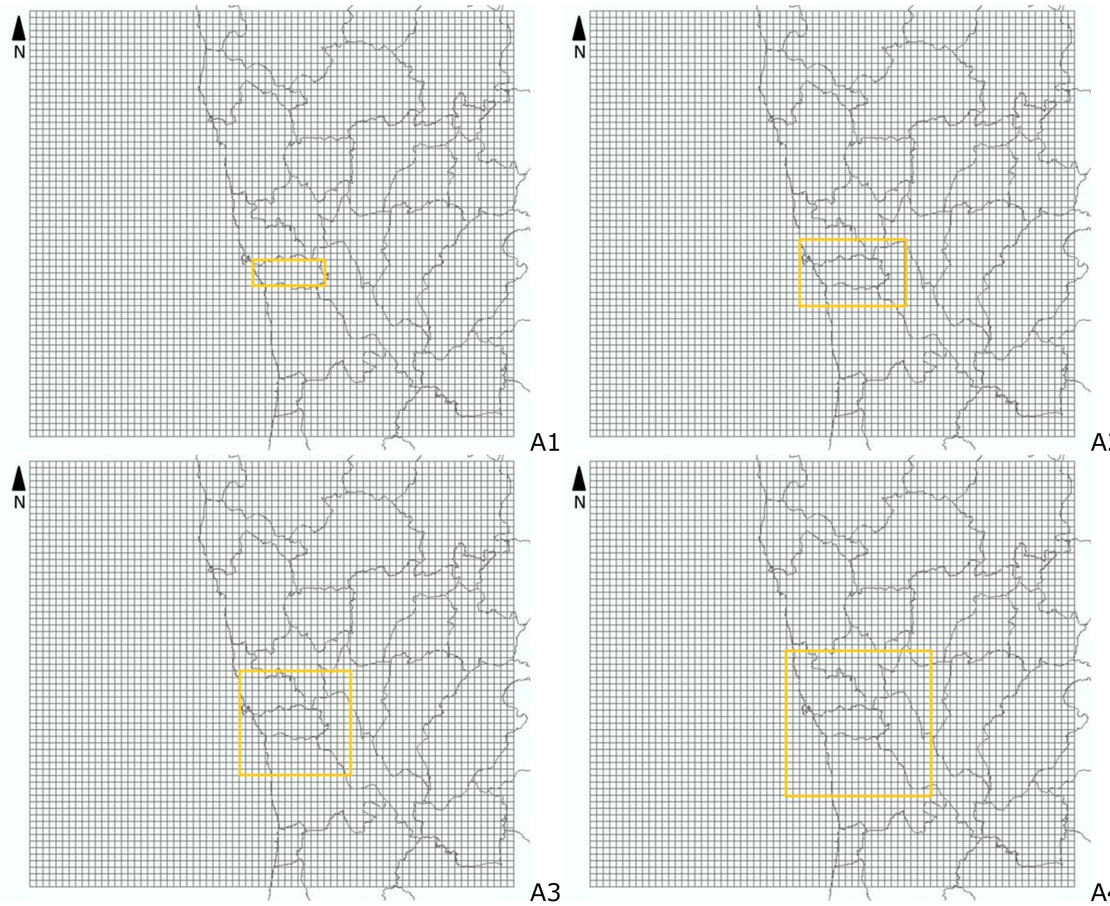


Figure A.9 – Selected areas of D3.

Figure A.10 have the statistical analysis of the synthetic data of each area, for each scenario for the three air pollutants.

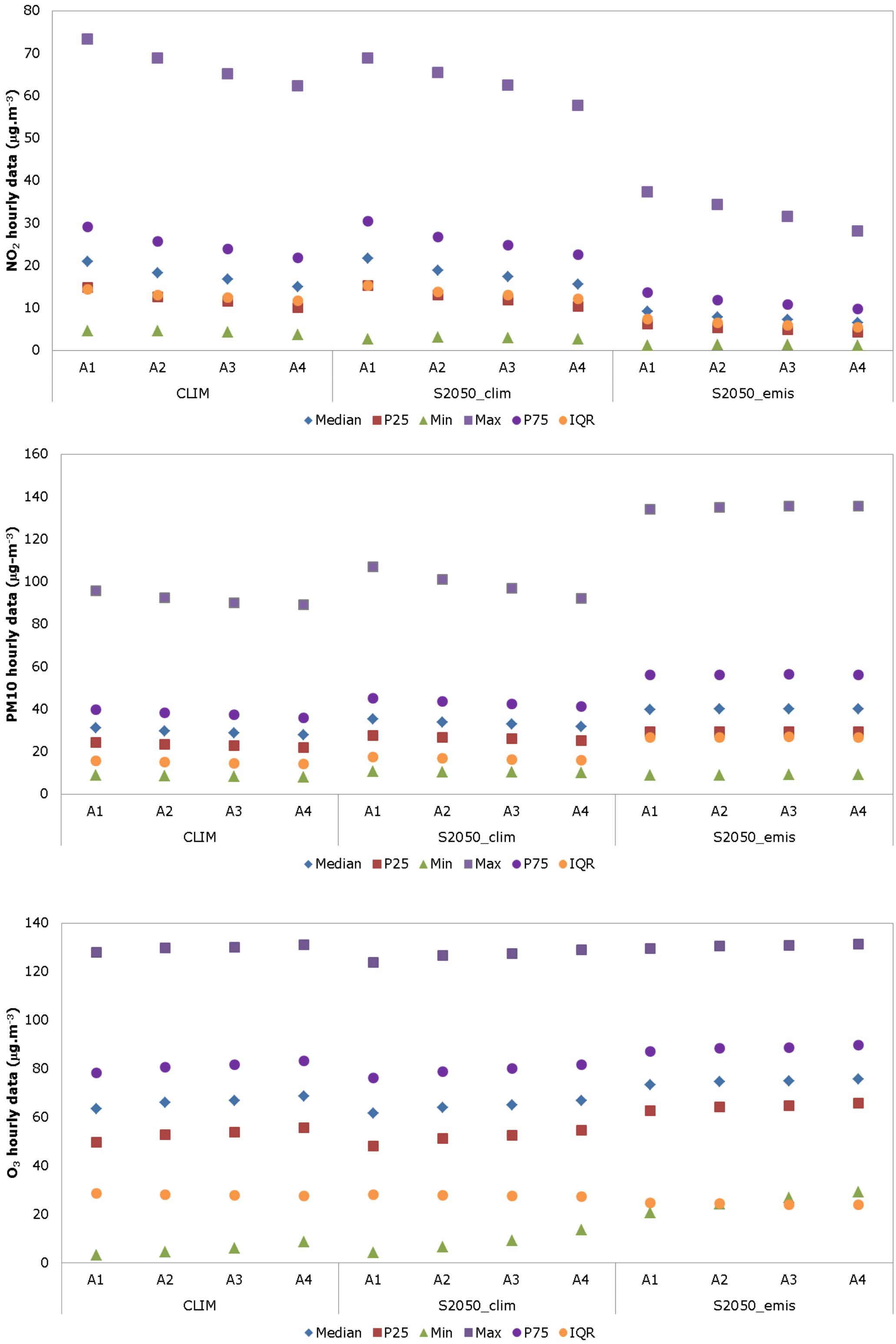


Figure A.10 – NO_2, PM10 and O_3 statistical analysis for each selected area for all the scenarios.