Universidade de Aveiro
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AVALIAÇÃO DA QUALIDADE DO AR NAS CIDADES EM CENÁRIOS DE ALTERAÇÃO CLIMÁTICA: ANÁLISE DA CONTRIBUIÇÃO DE FONTES

ASSESSING AIR QUALITY IN CITIES UNDER CLIMATE CHANGE SCENARIOS: A SOURCE APPORTIONMENT APPROACH

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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 da Doutora Myriam Lopes, Professora Associada do Departamento de Ambiente e Ordenamento da Universidade de Aveiro e coorientação científica da Doutora Joana Ferreira, Investigadora Auxiliar no Departamento de Ambiente e Ordenamento da Universidade de Aveiro.

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palavras-chave

Qualidade do ar; Modelação numérica; Impactos na saúde; Região de Aveiro; Apoio à decisão

resumo

As alterações climáticas e a poluição atmosférica são grandes desafios atuais e futuros das áreas urbanas. Abordagens integradas de combate à poluição atmosférica e às alterações climáticas podem desempenhar um papel importante no apoio à gestão do espaço urbano, orientado para o bem-estar dos cidadãos.

Neste contexto, o principal objetivo deste trabalho foi desenvolver uma abordagem de modelação capaz de avaliar os impactos do clima futuro e das projeções de emissões, incluindo a contribuição relativa de diferentes regiões e setores fonte, na qualidade do ar à escala urbana.

A modelação da qualidade do ar, associada a cenários de alterações climáticas, é uma ferramenta poderosa para compreender e avaliar os processos físicos e químicos que venham a ocorrer no futuro na atmosfera. A região de Aveiro, no centro de Portugal, foi o caso de estudo selecionado por ser reconhecida como uma das regiões afetadas por alguns eventos de má qualidade do ar em Portugal e vulnerável aos efeitos das alterações climáticas. O sistema de modelação WRF-CAMx foi aplicado a um domínio urbano sobre a Região de Aveiro, para o clima futuro de médio prazo (considerando o cenário SSP2-4.5) e para o passado recente (considerado como referência). Na modelação da qualidade do ar foram aplicadas ferramentas de quantificação de contribuição de fontes, com base na revisão da literatura, permitindo quantificar a contribuição relativa de vários setores e áreas fonte para os níveis de poluição atmosférica. Com base nos resultados da contribuição de fontes, para o futuro de médio prazo, foram definidos e testados dois cenários de redução de emissões para o setor industrial, identificado como o mais relevante na região. Adicionalmente, para os cenários climáticos e de projeção de emissões considerados (com e sem reduções adicionais de emissões), foram estimados os impactos na saúde humana, expressos em número de mortes prematuras, devido à exposição prolongada à poluição atmosférica.

Os resultados demostram que, devido à implementação de medidas de neutralidade carbónica, definidas para combater as alterações climáticas, no futuro a médio prazo a qualidade do ar na Região de Aveiro poderá melhorar, com reduções de até 4 $\mu g.m^{-3}$ nas concentrações de PM2.5 e PM10, e de até 22 $\mu g.m^{-3}$ para NO_2. Também as mortes prematuras associadas à poluição atmosférica irão diminuir (cerca de 76% para PM2.5 e 100% para NO_2). No entanto, deverão ser consideradas medidas adicionais de redução de emissões para o setor industrial, uma vez que este será o setor que mais contribuirá para os níveis de poluição atmosférica no futuro.

Este trabalho é inovador porque inclui alterações climáticas juntamente com uma avaliação de impacto na saúde no processo de gestão da qualidade do ar, focando-se na escala urbana onde a densidade populacional é maior e a poluição do ar representa um enorme desafio. Além disso, a abordagem aplicada fornece informações relevantes para o apoio ao planeamento urbano para proteção do ambiente e da saúde dos cidadãos.

keywords

Air quality; Numerical Modelling; Health impacts; Aveiro Region; Decision support

abstract

Climate change and air pollution are the current and future challenges of urban areas. Integrated approaches tackling air pollution and climate change can play an important role to support urban planning oriented for citizen welfare.

In this context, the main goal of this work was to develop a modelling approach able to assess the impacts of future climate and projected emissions, including the relative contribution of different source regions and sectors, on air quality at urban scale. The Aveiro Region, in central Portugal, was selected as a case study since it is recognized as one of the regions affected by some poor air quality events in Portugal and vulnerable to climate change effects.

Air quality modelling, associated with climate change scenarios, is a powerful tool to understand and assess the physical and chemical processes occurring in the atmosphere. In this scope, the WRF-CAMx modelling framework was applied to an urban domain over the Aveiro Region, for the medium-term future climate (considering the SSP2 4.5 scenario) and for the recent-past (considered as reference). For the air quality modelling, source apportionment tools were applied, based on literature review findings, allowing to quantify the relative contribution of several source activities and areas to ambient pollution. Based on the source apportionment results, for the medium-term future, two emission abatement scenarios were defined and tested for the industrial sector identified as the most relevant in the region. Additionally, for the climate scenarios and projected emissions (with and without additional emission reductions), mortality health indicators, expressed as the number of premature deaths, were used to estimate health impacts of long-term exposures.

Results show that in the medium-term future, air quality in the Aveiro Region may improve, with reduction up to 4 $\mu g.m^{-3}$ for PM2.5 and PM10 concentrations and 22 $\mu g.m^{-3}$ for NO_2, due to the implementation of carbon neutrality measures, defined to fight climate change. Also, the premature deaths linked to air pollution long-term exposure will decrease (76% for PM2.5 and 100% for NO_2). However, additional emission abatement measures should be considered for the industrial sector, as this will be the greater contributor to the concentration levels of the main pollutants in the future.

This work is innovative once it includes climate change together with a health impact assessment in the air quality management process, focusing on the urban scale where population density is higher and air pollution represents a huge challenge. Moreover, the applied approach provides valuable information to support urban planning for the protection of environment and citizens' health.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ORIGINAL PUBLICATIONS

This thesis is the result of the work contained in the following original publications:

(i) **Coelho S.**, Ferreira J., Lopes M. (2023) Source apportionment of air pollution in urban areas: a review of the most suitable source-oriented models. *Air Quality, Atmosphere & Health:* 1-10. https://doi.org/10.1007/s11869-023-01334-z

(ii) **Coelho S.**, Ferreira J., Rodrigues V., Lopes M. (2022). Source apportionment of air pollution in European urban areas: lessons from the ClairCity project. *Journal of Environmental Management* 320: 115899. https://doi.org/10.1016/j.jenvman.2022.115899

(iii) **Coelho S.**, Ferreira J., Carvalho D., Lopes M. (2022). Air quality impacts on human health, under a climate change scenario: methodology and case study application. *Sustainability* 14: 14309. https://doi.org/10.3390/su142114309

(iv) **Coelho S.**, Ferreira J., Lopes D., Carvalho D., Lopes M. (2023) Facing the challenges of air quality and health in a future climate: the Aveiro Region case study. *Science of The Total Environment*: 162767. https://doi.org/10.1016/j.scitotenv.2023.162767

Other authored and co-authored works (papers, conference proceedings and book chapters) were published during the course of the PhD work and enriched the research and findings presented here, namely:

(i) **Coelho S.**, Rafael. S., Fernandes A.P., Lopes M., Carvalho D. (2023) How the new climate scenarios will affect air quality trends: an exploratory research. *Urban Climate* 49: 101479. https://doi.org/10.1016/j.uclim.2023.101479

(ii) **Coelho S.**, Ferreira J., Carvalho D., Miranda A.I., Lopes M. (2022) Climate change impact on source contributions to the air quality in Aveiro Region. *In: Air Pollution Modeling and its Application XXVIII. ITM 2021.* [Mensink C., Jorba O. (eds.)] Springer Proceedings in Complexity. Springer International Publishing, p 308. http://doi.org/10.1007/978-3-031-12786-1_29

(iii) Rodrigues V., Gama C., Ascenso A., Oliveira K., **Coelho S.**, Monteiro A., Hayes E., Lopes M. (2021) Assessing air pollution in European cities to support a citizen centered approach to air quality management. *Science of The Total Environment* 799: 149311. http://doi.org/10.1016/j.scitotenv.2021.149311

(iv) Fernandes A.P., Rafael S., Lopes D., **Coelho S.**, Borrego C., Lopes M. (2021) The air pollution modelling system URBAIR: how to use a Gaussian model to accomplish high spatial and temporal resolutions. *Air Quality, Atmosphere & Health* 14: 1969-1988. http://doi.org/10.1007/s11869-021-01069-9

(v) **Coelho S.**, Rafael S., Lopes D., Miranda A.I., Ferreira, J. (2021) How changing climate may influence air pollution control strategies for 2030? *Science of The Total Environment* 758: http://doi.org/10.1016/j.scitotenv.2020.143911

(vi) Ascenso A., Augusto B., Silveira C., Rafael S., **Coelho S.**, Monteiro A., Ferreira J., Menezes I., Roebeling P., Miranda A.I. (2021) Impacts of nature-based solutions on the urban atmospheric environment: a case study for Eindhoven, The Netherlands. *Urban Forestry & Urban Greening* 57: 126870. http://doi.org/10.1016/j.ufug.2020.126870

(vii) Cherif S., Doblas-Miranda E., Lionello P., Borrego C., Giorgi F., Iglesias A., Jebari S., Mahmoudi E., Moriondo M., Pringault O., Rilov G., Somot S., Tsikliras A., Vila M., Zittis G. (2020) Drivers of change. *In: Climate and Environmental Change in the Mediterranean*

Basin – Current Situation and Risks for the Future. First Mediterranean Assessment Report [Cramer W., Guiot J., Marini K. (eds.)] Union for the Mediterranean, Plan Bleu, UNEP/MAP, Marseille, France, pp. 59-180. http://doi.org/10.5281/zenodo.4768833 (**Coelho S.** as contributing author)

(viii) **Coelho S.**, Rafael S., Coutinho M., Monteiro A., Medina J., Figueiredo S., Cunha S., Lopes M., Miranda A.I., Borrego C. (2020) Climate-Change Adaptation Framework for Multiple Urban Areas in Northern Portugal. *Environmental Management* 66: 395-406. http://doi.org/10.1007/s00267-020-01313-5

(ix) **Coelho S.**, Rodrigues V., Barnes J., Boushel C., De Vito L., Lopes M. (2018) Air pollution in the Aveiro region, Portugal: A citizens' engagement approach. *WIT Transactions on Ecology and the Environment. Air Pollution XXVI*. 230: 253–262. http://doi.org/10.2495/AIR180241

(x) Borrego C., Rafael S., Rodrigues V., Monteiro A., Sorte S., **Coelho S.**, Lopes M. (2018) Air quality, urban fluxes and cities resilience under climate change - A brief overview. *International Journal of Environmental Impacts: Management, Mitigation and Recovery* 1 (1): 14-27. http://doi.org/10.2495/ei-v1-n1-14-27

(xi) **Coelho S.**, Ferreira J., Rodrigues V., Rafael S., Borrego C., Lopes M. (2017) Identification and analysis of source contributions to the air quality in the Amsterdam region. *WIT Transactions on Ecology and the Environment. Air Pollution XXV*. 211: 31-40. http://doi.org/10.2495/AIR170031

(xii) Rafael S., Martins H., Marta-Almeida M., Sá E., **Coelho S.**, Rocha A., Borrego C., Lopes M. (2017) Quantification and mapping of urban fluxes under climate change: Application of WRF-SUEWS model to Greater Porto area (Portugal). Environ Res 155: 321-334. http://doi.org/10.1016/j.envres.2017.02.033

ABBREVIATIONS

C_0	Baseline Concentration
CAM-Chem	Community Atmosphere Model with Chemistry
CAMx	Comprehensive Air Quality Model with Extensions
CB6	Carbon Bond 6
CDR	Crude Death Rate
CI	Confidence interval
CLRTAP	Convention on Long-Range Transboundary Air Pollution
CMIP5	Coupled Model Intercomparison Project phase 5
CMIP6	Coupled Model Intercomparison Project phase 6
CO	Carbon Oxide
CO_2	Carbon Dioxide
CRF	Concentration-Response Function
Csb	Warm-Summer Mediterranean Climate
CTM	Chemical Transport Model
EC	European Commission
EEA	European Environment Agency
EMEP	European Monitoring and Evaluation Programme
ESM	Earth System Model
EU	European Union
FAIRMODE	Forum for Air Quality Modelling
GHG	Greenhouse Gas
ICD-10	International Classification of Diseases - 10th Revision
INE	Portuguese national institute for statistics
IPCC	International Panel for Climate Change
LE	Life Expectancy
MEGAN	Model of Emissions of Gases and Aerosols from Nature
MOZART	Model for Ozone and Related chemical Tracers
MPI	Max Planck Institute for Meteorology
MQI	Modelling Quality Indicator
NAPCP	National Air Pollution Control Programmes
NEC	National Emission Ceilings
NH_3	Ammonia
NMVOC	Non-Methane Volatile Organic Compound
NO_2	Nitrogen Dioxide
NOx	Nitrogen Oxides
O_3	Ozone

PAF	Population Attributable Fraction
PD	Premature Deaths
PM	Particulate Matter
PM10	Particulate Matter with an aerodynamic diameter of 10 microns or less
PM2.5	Particulate Matter with an aerodynamic diameter of 2.5 microns or less
PSAT	Particulate Source Apportionment Technology
RCP	Representative Concentration Pathway
RM	Receptor Model
RMSE	Root Mean Square Error
RNC2050	Roadmap for Carbon Neutrality 2050
RR	Relative Risk
SA	Source Apportionment
SNAP	Selected Nomenclature for Air Pollution
SO_2	Sulphur Dioxide
SOMO35	Sum of Ozone Means Over 35 ppb
SOx	Sulphur Oxides
SSP	Shared Socio-economic Pathway
UN	United Nations
UNECE	United Nations Economic Commission for Europe
UNFCCC	United Nations Framework Convention on Climate Change
WCRP	World Climate Research Programme
WHO	World Health Organization
WRF	Weather Research and Forecasting model
YLL	Years of Life Lost

CHAPTER 1

1. GENERAL INTRODUCTION

In this chapter an overview of the scientific context of the thesis is provided, where general knowledge on climate change, air quality, and their impacts on urban areas is introduced. This chapter also details the main objective and the research questions of this work, as well as the outline of the thesis.

1.1. OVERVIEW

Air pollution and climate change are serious environmental problems nowadays and for the next decades (MedECC 2020). Many cities across the world, particularly densely populated urban areas, are facing air pollution problems, with exceedances to legislated air quality levels. Nowadays air pollution is the biggest environmental risk factor, since exposure to air pollutants can affect human health, leading to increased mortality and morbidity (WHO 2022).

Air quality is linked to the earth's climate and ecosystems globally (WHO 2020). Many of the drivers of air pollution are also sources of greenhouse gases (GHGs) emissions. Policies to reduce air pollution, consequently, offer a win-win strategy for both climate and health, lowering the burden of disease attributable to air pollution, as well as contributing to the near- and long-term mitigation of climate change (WHO 2020, 2022). In this sense, a better understanding of the effects of climate change on urban air quality, and how they affect human health, are currently research challenges.

1.1.1. Climate Change

According to the International Panel for Climate Change (IPCC) Special Report on the impacts of global warming (IPCC 2018), climate change is "a change in the state of the climate that can be identified (e.g., by using statistical tests) by changes in the mean and/or the variability of its properties and that persists for an extended period, typically decades or longer". The same IPCC special report (IPCC 2018) also states that: "climate change may be due to natural internal processes or external forcings such as modulations of the solar cycles, volcanic eruptions and persistent anthropogenic changes in the composition of the atmosphere or in land use". However, for other organizations such as the United Nations Framework Convention on Climate Change (UNFCCC) (UN 1992) climate change means "a change of climate which is attributed directly or indirectly to human activity that alters the composition of the global atmosphere and which is in addition to natural climate variability observed over comparable time periods", making a distinction between climate change attributable to anthropogenic activities and climate variability resultant from natural causes.

Evidences of a changing climate have been supported by the changes documented in the main elements of the climate system, namely, atmosphere, land, cryosphere, biosphere and ocean (Arias et al. 2021). The Fourth Assessment Report of IPCC, published in 2007 (AR4; IPCC 2007), was the first to conclude that warming of the climate system is unequivocal. The increasing availability of satellite and ground-based observations datasets of diverse aspects of the climate system, as well as climate reconstructions and reanalysis, corroborate the transition to warmer conditions during the 20th century and that warming has accelerated during the last decades (MedECC 2020; Gulev et al. 2021). According to Gulev et al. (2021), since the late 19th century, global surface temperature over land and global average sea surface temperature have increased. The troposphere has warmed, and the near-surface specific humidity over land has increased. Warming oceans have caused ocean waters to expand, leading to rising sea levels. Also, the atmospheric circulation has been changing, including a poleward shift of mid-latitude storm tracks (Gulev et al. 2021).

As previously mentioned, climate change can be induced by natural or anthropogenic drivers (IPCC 2018; Arias et al. 2021). In the latest IPCC reports (Hegerl et al. 2007; Bindoff et al. 2013; Eyring et al. 2021), the climate change detection and attribution to natural-only or anthropogenic and natural forcings was made through the comparison of observed datasets of several climate change indicators with climate models results, based on the defined in the IPCC Good Practice Guidance Paper (Hegerl et al. 2010). Figure 1.1 shows observed and simulated changes of air temperature (over land and globally) and ocean heat content, compared to the 1995–2014 averages. For the simulated changes, results from the climate models assemble participating in the Coupled Model Intercomparison Project Phase 6 (CMIP6; Eyring et al. 2016; O'Neill et al. 2016) of the World Climate Research Programme (WCRP) were used, with and without anthropogenic forcings.

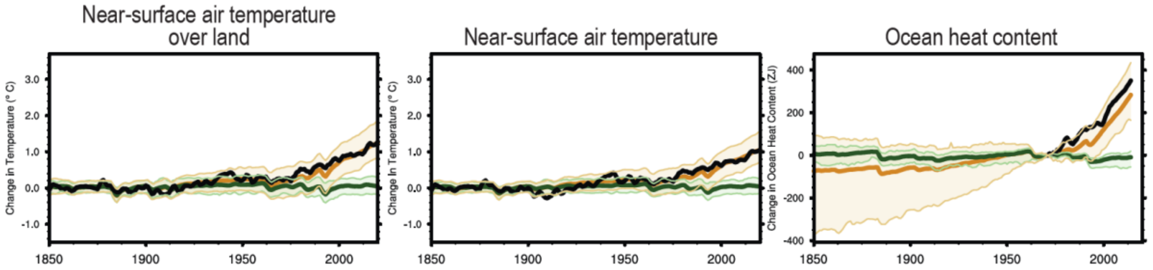

Figure 1.1. *Global near-surface air temperature (only over land and globally) and ocean heat content, simulated and observed changes for the period 1850-2014, compared to the 1995–2014 averages. Black lines show observations; orange lines and shading show averages over the CMIP6 historical simulations and 5-95th percentile ranges, considering both natural and anthropogenic forcings; green lines and shading show CMIP6 historical simulations ensemble and 5-95th percentile ranges, considering only natural forcing. Adapted from Arias et al. (2021).*

Figure 1.1 shows that, for the three climate change indicators considered, the simulations including only natural forcings differs considerably from the observations. A Better agreement between observations and simulations occurs when the models include both anthropogenic and natural forcings. This behaviour demonstrates how the anthropogenic drivers, such as carbon dioxide (CO_2) emitted by combustion of fossil fuels, atmospheric aerosols, non-CO_2 GHGs and land use changes, have impacted the climate system (Arias et al. 2021), and the importance of the human activities will have in the future climate.

As for the detection and attribution of climate change, climate modelling has become a powerful tool to represent and understand the complex climate system (Borrego et al. 2018; Lin et al. 2020) and has been applied as a research tool to study and simulate future climate changes (Flato et al. 2014; IPCC 2018). Depending on the aspects considered (e.g., the number of spatial dimensions; the extent to which physical, chemical or biological processes are explicitly characterized; or the level at which empirical parametrizations are involved), climate models can be of varying complexity (IPCC

2018). The climate simulations carried out under the CMIP6 framework (Eyring et al. 2016) and the Sixth Assessment Report of IPCC (AR6; IPCC 2021a), are made using a hierarchy of climate models, ranging from simple climate models, to models of intermediate complexity, to comprehensive climate models, and Earth System Models (ESMs).

CMIP6 projections of future climate were performed based on a set of five new generation scenarios, with different trajectories for human development and global environmental change, the Shared Socio-economic Pathways (SSPs; O'Neill et al. 2017; Riahi et al. 2017). The five SSPs comprise a textual description of reasonable human development strategies that lead to very different future challenges, within the context of climate impacts and climate mitigation and adaptation strategies (van Vuuren et al. 2017a). Each SSP scenario has quantitative descriptions of key elements, including population (KC and Lutz 2017), urbanization (Jiang and O'Neill 2017), economic growth (Crespo Cuaresma 2017; Dellink et al. 2017; Leimbach et al. 2017), energy system (Bauer et al. 2017), land use changes (Popp et al. 2017), and air pollutants emissions (Rao et al. 2017). The five SSPs narratives, described in detail in O'Neill et al. (2017) and shown in Figure 1.2, are:

(i) SSP1, "*Sustainability - Taking the Green Road"*. Based on global cooperation, this narrative highlights the use of eco-friendly technologies, increasing use of renewable energy, the transition to lifestyles with less intensive use of resources and low energy demand, still assuming a relatively high rate of economic growth. The combination of these factors will lead to relatively low challenges to mitigation and adaptation.

(ii) SSP2, *"Middle of the Road"*. This narrative follows a tradition of dynamics-as-usual, in which economic and social trends do not change considerably from historical patterns. Technological developments advance quickly, environmental systems will have some improvements and the intensity of energy and resources use will decrease, as well as the dependency of fossil fuels. These moderated development trends will lead to intermediate challenges to mitigation and adaptation, with significant differences across countries.

(iii) SSP3, *"Regional Rivalry - A Rocky Road"*. In this narrative the main focus is on the high fossil fuel dependency and the increasing intensive use of resources, along with difficulties in achieving international cooperation to address environmental and other global concerns, due to a resurgent nationalism and regional conflicts. Technological changes will be slow, there will be limited progress on human development, combined with slow economic growth. This pathway will imply high challenges for both mitigation and adaptation.

(iv) SSP4, *"Inequality - A Road Divided"*. This narrative considers increasing inequalities and stratification, both across and within the countries, mainly due to highly unequal investments in human capital, along with increasing disparities in economic growth and political power. For some regions, the development of low carbon technologies, together with the international political and business classes well integrated and able to decide and act quickly, will lead to low challenges for mitigation. On the other hand, a substantial part of the population, mainly at less developed regions with limited access to institutions capable of dealing with economic and environmental issues, will face high challenges for adaptation.

(v) SSP5, *"Fossil-fuelled Development – Taking the Highway"*. This narrative is driven by economic and social developments based on the exploitation of abundant fossil fuel resources and the adoption of resources and energy intensive lifestyles. Also, a strong investment in health, education and institutions will be considered to develop human and social capital. Highly engineered infrastructures, a robust economic growth and the attained human developments will result in low challenges to adaptation. However, the fossil fuels dependence coupled with the lack of environmental concerns will imply high challenges to adaptation.

The main dynamics and implementation of each SSP are described in five dedicated papers, namely: (i) van Vuuren et al. (2017b) for SSP1; (ii) Fricko et al. (2017) for SSP2; (iii) Fujimori et al. (2017) for SSP3; (iv) Calvin et al. (2017) for SSP4; and (v) Kriegler et al. (2017) for SSP5.

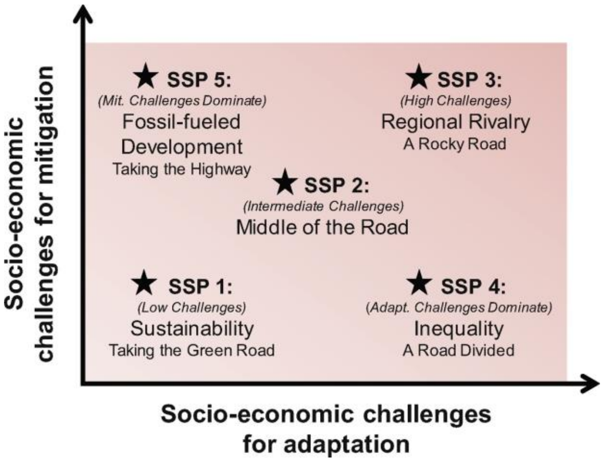

Figure 1.2. *The five SSPs, with different combinations of challenges to mitigation and to adaptation. From O'Neill et al. (2017).*

According to Chen et al. (2021), the five SSPs narratives can lead to several global radiative forcing levels, depending on the pathways of CO_2, non-CO_2 GHGs, aerosols and land use. This approach is aligned and follows-up the radiative forcings considered in the four Representative Concentration Pathways (RCPs) scenarios of CMIP5 (Coupled Model Intercomparison Project Phase 5), namely, RCP2.6, RCP4.5, RCP6.0 and RCP8.5 (with a radiative forcing of 2.6, 4.5, 6.0 and 8.5 $W.m^{-2}$ at the end of the century, respectively), and adds new forcing pathways that, according to O'Neill et al. (2016), would be of interest to the climate science communities. The full set of the five SSPs and multiple climate forcing levels forms a matrix of possible integrated scenarios (Kriegler et al. 2012; van Vuuren et al. 2014). From that matrix, O'Neill et al. (2016), identify four priority scenarios on which CMIP6 climate modelling groups should focus: SSP1 2.6 for sustainable pathways, SSP2 4.5 for middle-of-the-road, SSP3 7.0 for regional rivalry, and SSP5 8.5 for fossil fuel-rich development. Lee et al. (2021) also considered SSP1-1.9 as one of the scenarios that should be analysed, since it is directly relevant to the assessment of the 1.5°C Paris Agreement goal. In these new scenario labels, the first number refers to the assumed SSP, and the second refers to the approximate global radiative forcing in 2100, in $W.m^{-2}$. For these five high-priority scenarios, Figure 1.3 shows the example of the global surface air temperature changes, from the CMIP6 simulations ensemble, relative to the 1995–2014 average (considered as recent past) and relative to the 1850–1900 average (pre-industrial period).

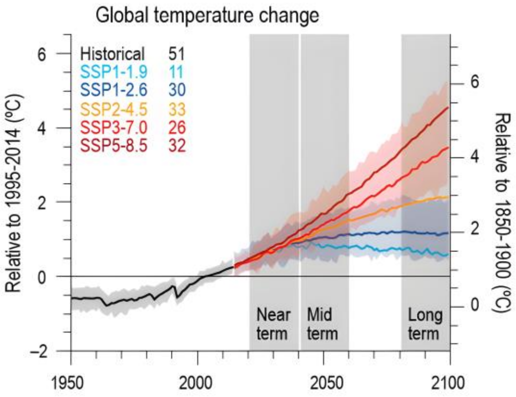

Figure 1.3. *Global surface air temperature changes, relative to the 1995–2014 average (left axis) and relative to the 1850–1900 average (right axis). The curves show averages over the CMIP6 simulations, the shadings around show 5-95th percentile ranges (only for SSP1-2.6 and SSP3-7.0), and the numbers near the top show the number of model simulations used. Adapted from Lee et al. (2021).*

From Figure 1.3, and according to Lee et al. (2021), for the SSP1-2.6 scenario, the 5-95th percentile range of global temperature change for 2081–2100 (long-term future), relative to 1995–2014, is between 0.6 °C and 2.0 °C. The corresponding ranges for SSP2-4.5 and SSP3-7.0, the intermediate and high emissions scenarios where CO_2 concentrations increase to 2100, but less rapidly than SSP5-8.5, are from 1.4 °C to 3.0 °C and from 2.2°C to 4.7 °C, respectively. The range for SSP5-8.5, the highest overall emissions scenario, is between 2.7 °C and 5.7 °C. The range for SSP1-1.9, the lowest emissions scenario, is 0.2 °C to 1.3 °C. The ensemble difference between 1850–1900 and 1995–2014 average values is around 0.82 °C, providing an estimate of the changes in the global surface air temperature since the pre-industrial period.

In addition to the future changes on global air temperature, the five high-priority scenarios will affect other key components of the climate system, changing the magnitude and frequency of meteorological parameters as well as natural cycles. IPCC (2021b) enumerated the main impacts on the climate system projected for the future, transversal to the five high-priority scenarios, namely:

(i) Global surface air temperature will continue to increase until at least mid-century under all emissions scenarios considered. Global warming of 1.5°C and 2°C will be exceeded during the 21st century unless deep reductions in CO_2 and non-CO_2 GHGs emissions occur in the coming decades.

(ii) An increase in the frequency and intensity of hot extremes, heavy precipitation, agricultural/ecological droughts, and intense tropical cyclones, will be also expected, as well as a decrease in Arctic Sea ice, snow cover and permafrost.

(iii) The global water cycle will be intensified, including its variability, global monsoon precipitation and the severity of wet and dry events.

(iv) For the scenarios with higher CO_2 emissions, it is expected that the ocean and land carbon sinks will be less effective at slowing the accumulation of CO_2 in the atmosphere.

(v) Past and future changes in GHGs emissions will be irreversible during centuries, especially changes in the ocean, ice sheets and global sea level.

A recent study conducted over Europe (Carvalho et al. 2021) investigated future changes in the surface temperature, using a CMIP6 multi-model ensemble. According to this study, temperature will increase across Europe particularly at northern (during winter) and southernmost (during summer) latitudes, where, according to SSP5-8.5, the warming can reach 3 °C in the middle of the century (as shown in Figure 1.4). According to Coelho et al. (2021), by 2030 and considering the RCP8.5 climate scenario, it is expected an increase of near-surface temperature up to 2.0 °C, and a general decrease in the annual total precipitation. Several studies (e.g., Kjellström et al. 2011; Jacob et al. 2014; Coelho et al. 2021) have pointed out that that strongest warming will occur in Southern Europe during the warmest months, with a consequent increase of the frequency, magnitude, and duration of heat waves. Increasing temperature, combined with decreasing precipitation, generates strong trends towards drier conditions, as it has been observed in southern Europe (Vicente-Serrano et al. 2014).

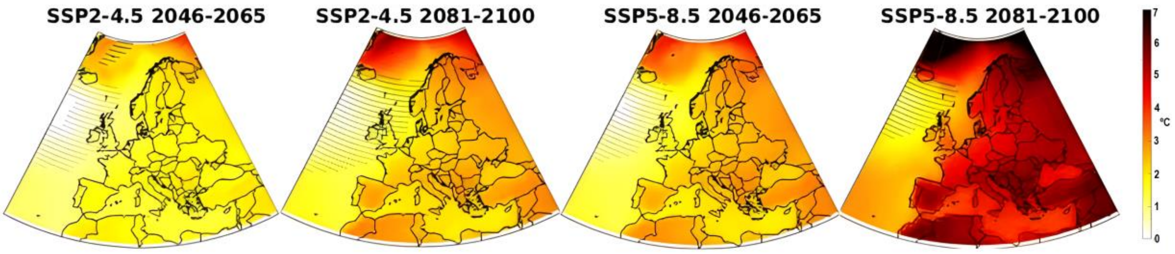

Figure 1.4. *Spatial maps of the mean surface temperature between the baseline (1995–2014) and the 2046–2065 and 2081–2100 future periods, for SSP2-4.5 and SSP5-8.5. Adapted from Carvalho et al. (2021).*

Over the last years, several studies have been published on the future climate projections in Portugal, since it has been identified as a climate hotspot, along with the whole Mediterranean region (Giorgi and Lionello 2008; MedECC 2020). Despite the studies using distinct climate scenarios and/or time scales (i.e., near-, mid-, or long-term future), which could lead to different results, four general conclusions can be drawn about the future climate in Portugal, namely:

(i) Projected temperature changes are rather severe and significant (Cardoso et al. 2019), with increases of up to 2 °C by the mid-century (Coelho et al. 2020, 2023a) and up to 4 °C by 2100 (Pereira et al. 2021);

(ii) Strongest warming is projected in summer, when high temperature events and heat waves are likely to become more frequent and/or more extreme (Rafael et al. 2017; Cardoso et al. 2019; Coelho et al. 2020, 2023a; Pereira et al. 2021);

(iii) Projected precipitation changes show a considerable decline in precipitation for all seasons except winter (Soares et al. 2017; Pereira et al. 2021; Coelho et al. 2023a);

(iv) The number of intense precipitation days will increase (Soares et al. 2017; Coelho et al. 2020; Pereira et al. 2021), as well as dryness periods (Soares et al. 2017; Pereira et al. 2021).

In order to combat climate change, in the end of 2021, Portugal has approved the climate framework law (AR 2021), establishing targets and requirements for the design of public policies across economic sectors and levels of government. Also, in line with the Paris Agreement (UNFCCC 2015), Portugal has defined the Roadmap for Carbon Neutrality 2050 (RNC2050), in which it committed to reduce its GHGs emissions so that the balance between emissions and removals from the atmosphere will be zero by 2050 (APA 2019). RNC2050 proposes three carbon neutrality trajectories, based on distinct narratives and macroeconomic scenarios, namely:

(i) Off-Track, the least ambitious scenario, that retains the essentials of the economic structure and current trends as well as the decarbonisation policies already adopted or in force;

(ii) Peloton, the medium ambition scenario, with socioeconomic developments compatible with the development and application of new technologies that, however, do not significantly change either the production structures or the population’s lifestyles, and foresees a modest incorporation of circular economy models and the maintenance of population concentration in the Metropolitan Areas;

(iii) Yellow Jersey, the most ambitious scenario, with a socio-economy characterised by a structural and transverse change in production chains, made possible by the combination of a series of technologies of the 4th Industrial Revolution, foreseeing a more effective incorporation of circular economy models and greater growth of the importance of medium-sized cities.

Independently of the scenario or the study region considered, climate change will result in an impact on the future meteorological patterns, with more relevance on temperature and precipitation (IPCC 2021b; Carvalho et al. 2022). This changes in the meteorological conditions will impact the atmospheric processes responsible for the chemistry, transport and deposition of air pollutants, leading also to air quality changes (Jacob and Winner 2009; Von Schneidemesser et al. 2015).

1.1.2. Air quality

According to the World Health Organization (WHO), air pollution is the “contamination of the indoor or outdoor environment by any chemical, physical or biological agent that modifies the natural characteristics of the atmosphere”. Seinfeld and Pandis (2006) defines it as “a situation in which substances that result from anthropogenic activities are present at concentrations sufficiently high above their normal ambient levels to produce a measurable effect on humans, animals, vegetation, or materials”, identifying already sources and key problems associated with air pollution. The state of air pollution is often expressed as air quality. Therefore, air quality is a measure of how clean or polluted the air is.

Air pollution has been regulated for many years, leading to relevant progresses in the past decades. Reducing air pollution is a complex challenge that requires coordinated efforts, from global to regional and local levels, to control pollutant emissions and to establish air quality standards, through the implementation of effective legislative and non-legislative measures. At European level, the Convention on Long-range Transboundary Air Pollution (CLRTAP) from the United Nations Economic Commission for Europe (UNECE) is an important regulatory framework. Under CLRTAP, established in 1979, parties are obligated to report several air pollutants emission data, including sulphur dioxide (SO_2), nitrogen oxides (NOx), non-methane volatile organic compound (NMVOC), carbon oxide (CO), ammonia (NH_3), particulate matter (PM, namely PM10 - particulate matter with a diameter of 10 microns or less, and PM2.5 - with a diameter of 2.5 microns or less).

At the European Union (EU) level, three main policy instruments can be identified, namely: (i) Directive 2008/50/EC (EC 2008) on ambient air quality and cleaner air for Europe; (ii) Directive 2004/107/EC (EC 2005) relating to arsenic, cadmium, mercury, nickel, and polycyclic aromatic hydrocarbons in ambient air; and (iii) Directive 2001/81/EC (EC 2001) on national emission ceilings (NEC). The 2008 air quality directive (EC 2008) merges most of the existing legislation, establishing the standards and requirements to ensure that EU Member States effectively monitor and/or assess air quality on their territory, in a harmonized and comparable manner. This includes the definition of limit values on the key air pollutants concentrations.

Based on the human health impacts, the WHO has published guidelines for several air pollutants and supports extensive research into the effects of air pollution. WHO air quality guidelines, whilst not legally binding, play a key role in policy discussions, serving as a global target for national, regional and city governments to work towards improving their citizen’s health by reducing air pollution. These guidelines are subject to periodic scientific review, typically every 10 years. The new WHO air quality guidelines (WHO 2021), revised in September 2021, are more ambitious than the previous ones, which were updated in 2005 (WHO 2006), reflecting the huge impact that air pollution has on global health (Hoffmann et al. 2021).

In the European Green Deal (EC 2019a), published in December 2019, the European Commission (EC) has committed to further improve air quality and to align EU air quality standards more closely with the WHO air quality guidelines. On October 26th, 2022, the EC proposed a revision of the EU Ambient Air Quality Directive (EC 2022), which should be considered by the European Parliament and the Council. Upon adoption, the new EU Ambient Air Quality Directive will be carried out gradually, with specific targets for 2030, 2040 and 2050 allowing authorities to adjust and invest accordingly.

Table 1.1 compares the air quality limit and target values defined in the Directive 2008/50/EC (EC 2008), the new EU air quality directive proposal (EC 2022), and the WHO air quality guidelines (WHO 2006, 2021) for the pollutants considered as most relevant for the human health protection.

Table 1.1. *Comparison between EU air quality directive (EC 2008), the new EU air quality directive proposal (EC 2022), and WHO air quality guidelines updated in 2005 and 2021 (WHO 2006, 2021), to human health protection, for main air pollutants.*

Pollutant	**Averaging period**	**Limit or target value**			
		EU		**WHO**	
		2008	**2021 (a)**	**2005**	**2021**
PM10 $\mu g.m^{-3}$	24-hour	50 (b)	45 (c)	50 (d)	45 (d)
	Year	40	20	20	15
PM2.5 $\mu g.m^{-3}$	24-hour	-	25 (c)	25 (d)	15 (d)
	Year	25	10	10	5
NO_2 $\mu g.m^{-3}$	1-hour	200 (e)	200 (f)	-	-
	24-hour	-	50 (g)	-	25 (d)
	Year	40	20	40	10
O_3 $\mu g.m^{-3}$	8-hour max (h)	120 (i)	120 (i)	100 (d)	100 (d)
	Peak season (j)	-	-	-	60
SO_2 $\mu g.m^{-3}$	1-hour	350 (k)	350 (f)	-	-
	24-hour	125 (l)	50 (g)	20 (d)	40 (d)
	Year	-	20	-	-
CO $mg.m^{-3}$	8-hour max (h)	10	10	-	-
	24-hour	-	4 (g)	-	4 (d)

Notes: (a) To be attained by 1 January 2030.
(b) Not to be exceeded more than 35 times per year.
(c) Not to be exceeded more than 18 times per year.
(d) 99th percentile (i.e., not to be exceeded more than 3-4 times per year).
(e) Not to be exceeded more than 18 times per year.
(f) Not to be exceeded more than once per year.
(g) Not to be exceeded more than 18 times per calendar year
(h) Maximum daily 8-hour mean concentration, selected from 8-hour running averages. Each 8-hour average so calculated will be assigned to the day on which it ends.
(i) Not to be exceeded more than 25 times per year, averaged over 3 years.
(j) Average of daily maximum 8-hour mean concentration, in the 6 consecutive months with the highest 6-month running-average.
(k) Not to be exceeded more than 24 times per year.
(l) Not to be exceeded more than 3 times per year.

In 2019, the EC published a fitness check of the EU Ambient Air Quality Directives (EC 2019b), assessing the performance of the two complementary EU Ambient Air Quality Directives (2004/107/EC and 2008/50/EC (EC 2005, 2008)), and concluded that they have been partly effective in improving air quality and achieving air quality standards. For the EU, the reduction trend in the annual concentrations of the main pollutants, PM10 and PM2.5, nitrogen dioxide (NO_2), and ozone (O_3), can be seen in Figure 1.5.

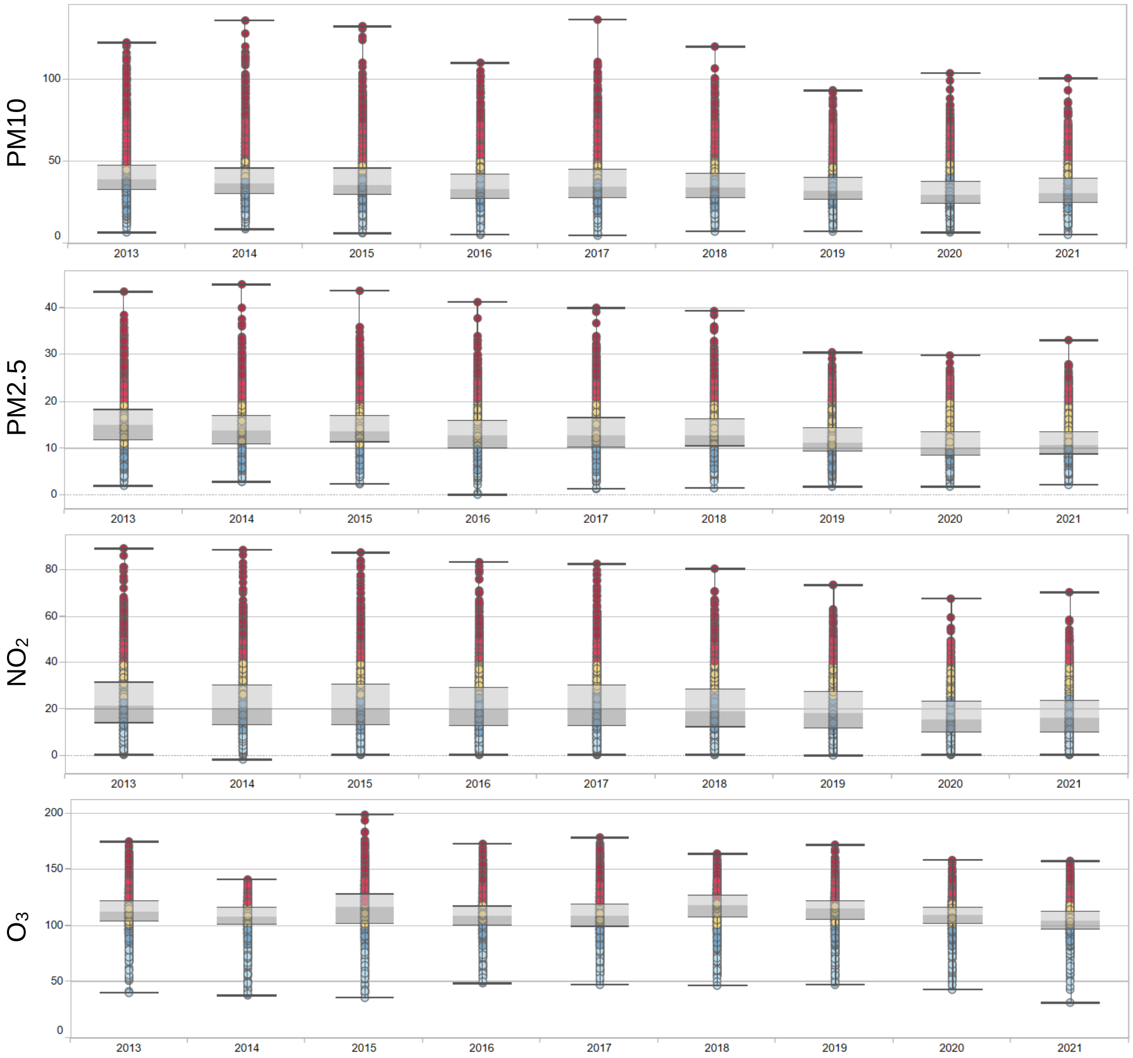

Figure 1.5. *Annual concentration ranges, in µg.m^{-3}, of PM10 (90.4 percentile of daily mean), PM2.5 (annual average), NO_2 (annual average) and O_3 (93.2 percentile of maximum daily 8-hour average), from 2013 to 2021, for each air quality monitoring station in EU. Red points represent non-compliance of the legislated limit or target value. From EEA (2022a).*

Despite the improvements already achieved (as shown in Figure 1.5), in some European countries/regions, ambient air concentration levels continue to surpass the legislated values for PM10, PM2.5, NO_2 and O_3, damaging human health and the environment (EEA 2022b). Figure 1.6 shows annual mean concentrations, for 2020, for PM10, PM2.5, NO_2 and O_3, across Europe.

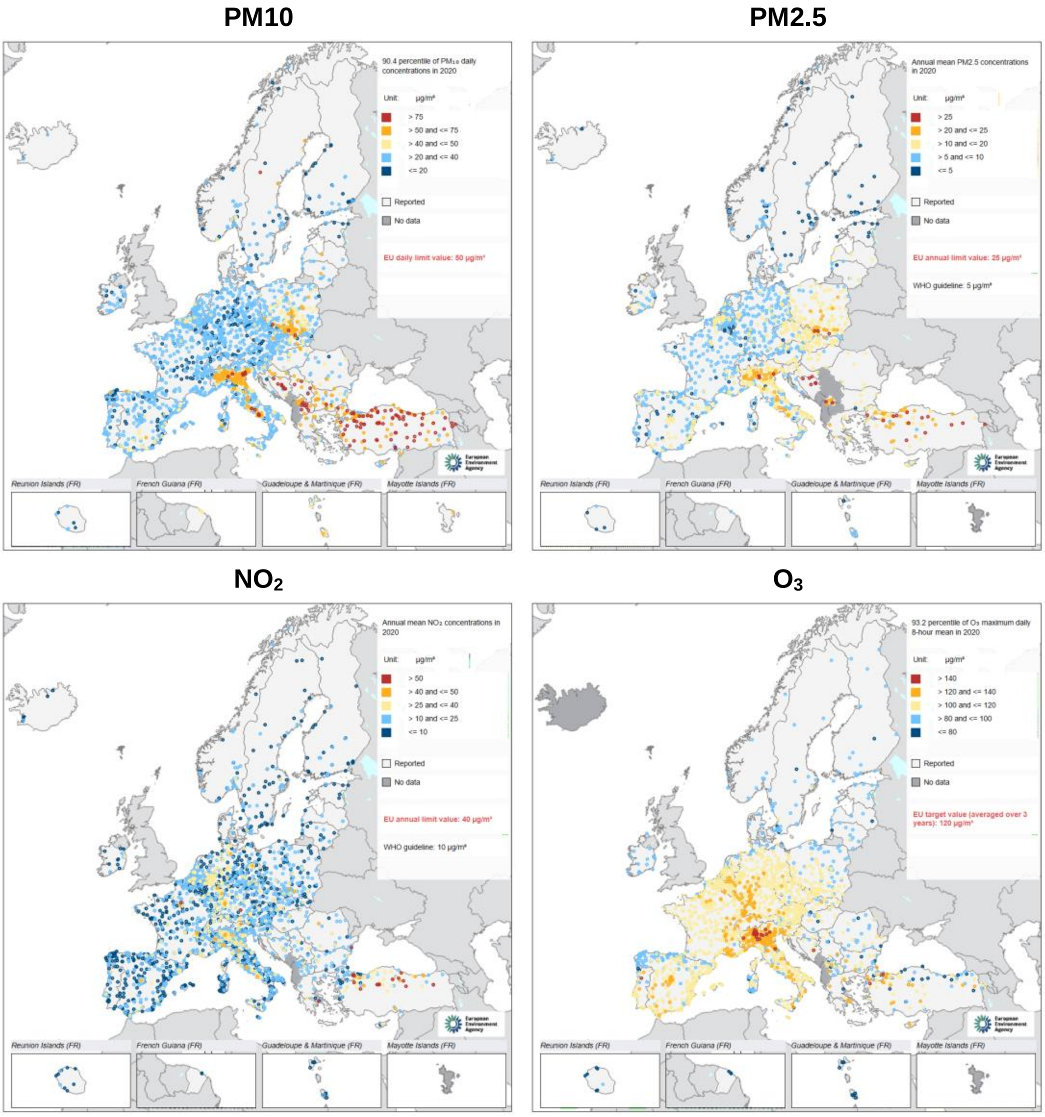

Figure 1.6. *Observed concentrations, in µg.m^{-3}, of PM10 (90.4 percentile of daily mean), PM2.5 (annual mean), NO_2 (annual mean) and O_3 (93.2 percentile of maximum daily 8-hour mean), for 2020 in Europe. From EEA (2022b).*

According to Figure 1.6, in Europe, the annual limit and target values of PM10, PM2.5, NO_2 and O_3 were surpassed in 2020 in several air quality monitoring stations. Nevertheless, it should be noted that, in 2020, lockdown measures introduced to minimise the spread of Covid-19 had an impact on air pollutant emissions from road transport, leading to a temporary reduction in NO_2 concentrations. These concentrations have led to most of the EU's urban population being exposed to levels of key air pollutants that are harmful to health, as shown in Figure 1.7.

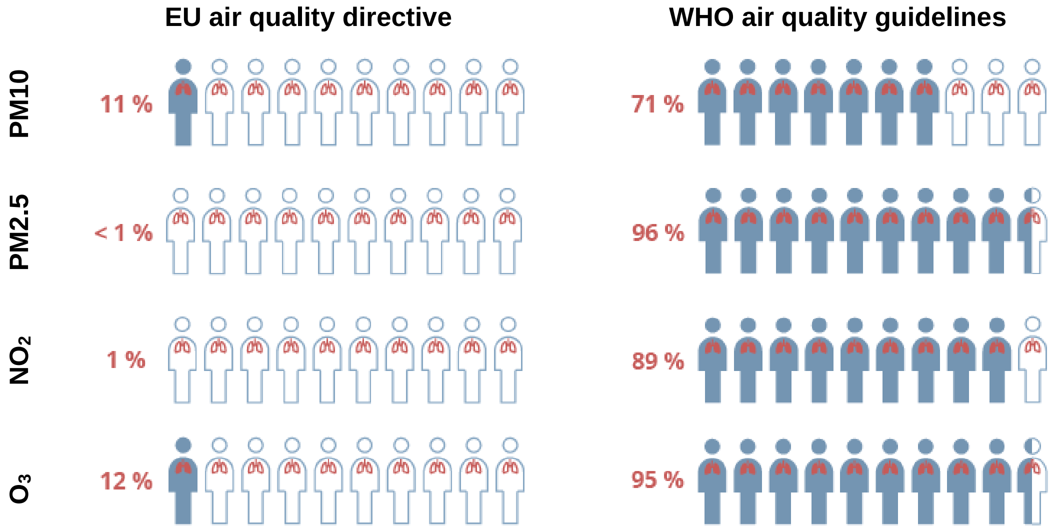

Figure 1.7. *Share of the EU urban population exposed to air pollutant concentrations above EU standards and WHO guidelines in 2020. Adapted from EEA (2022b).*

Figure 1.7 shows that, in 2020, 12% of the EU urban population lived in zones with O_3 concentrations above the EU standards, while 11% were exposed to PM10 and less than 1% to PM2.5 and NO_2 concentrations above the EU limit values. However, when considering the 2021 WHO air quality guidelines, the EU urban population exposure raised to 93% for O_3, 71% for PM10, 96% for PM2.5 and 89% for NO_2. If the new WHO air quality guidelines are met in 1000 European cities (with a total population of more than 168 million people), it is estimated that more than 100 thousand premature deaths could be prevented annually due to the exposure to PM2.5, and nearly 60 thousand dues to NO_2 (UN-Habitat 2022).

Both short- (over a few hours or days) and long-term (over months or years) exposure to air pollution may lead to adverse health effects, such as mortality and morbidity, mainly due to cardiovascular and respiratory diseases (WHO 2013a, b). Mortality, measured in terms of premature deaths, reflects a reduction in life expectancy as a result of exposure to air pollution, while morbidity refers to the occurrence of illness and years lived with a disease or disability, which may require hospitalisation (EEA 2020).

In order to reduce the air pollution and the impacts on the human health and the environment, EU Member states should develop an air quality plan for the zones and/or agglomerations where the air quality directive is not fulfilled, putting in risk the population's health and the environment (EC 2008). To achieve the established air quality objectives, the air quality plan must define air pollution abatement measures, based on a detailed analysis of the origin of the pollution, including the main emission sources and information on pollution imported from other regions, among others (EC 2008).

According to the EU emission inventory reported under the CLRTAP (EEA 2021), in 2020, in the EU, the main emission sectors were: manufacturing and extractive industry for NMVOC; transport for NOx; and residential, commercial and institutional for PM10 and PM2.5. For Portugal, the shares of emission by sector group are similar to those of the EU, with the exception of PM10 and PM2.5 emissions, mostly from manufacturing and extractive industry, followed by the residential, commercial and institutional sector.

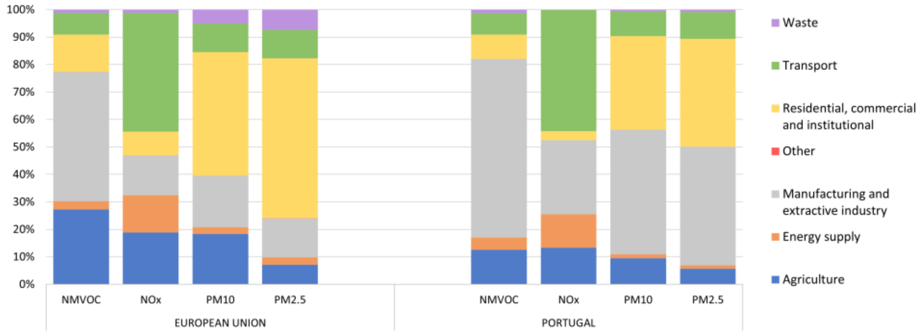

Figure 1.8. *Share of the EU and Portuguese emissions of the main pollutants, by sector group in 2020. Adapted from EEA (2021).*

As in Europe, in recent years air quality has been improving in Portugal, with pollutants concentrations above EU legislated values not being recorded. However, improvements can still be made in order to attain WHO air quality guidelines. Figure 1.9 shows exceedances to the daily limit value of PM10, annual limit value of NO_2, and information and alert thresholds of O_3, in Portugal in 2021, published in the latest edition of the Portuguese State of the Environment Report (APA 2022).

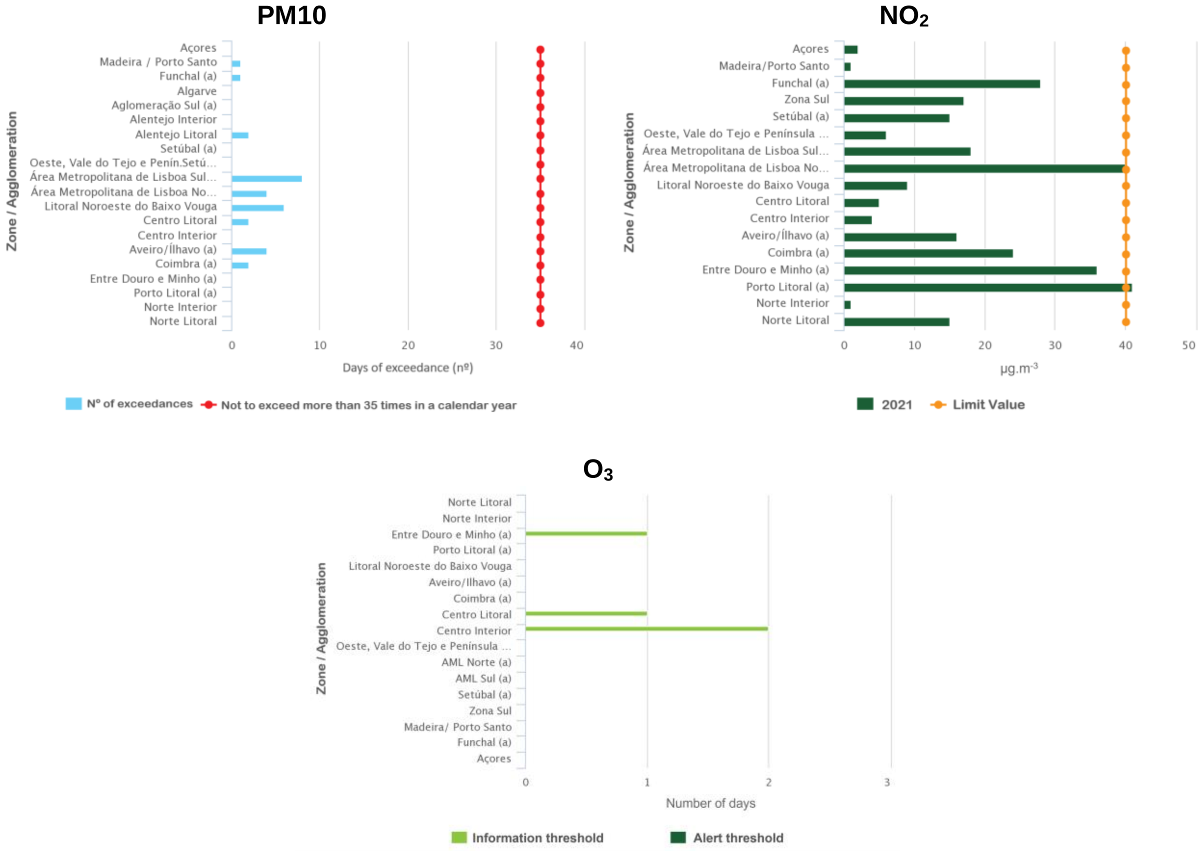

Figure 1.9. *Exceedances to the EU daily limit value of PM10, annual limit value of NO_2, and information and alert thresholds of O_3, in each zone/agglomeration (a) in Portugal in 2021. Adapted from APA (2022).*

According to Figure 1.9, of the 20 zones/agglomerations delimited to assess PM10 in Portugal, 10 did not register any exceedance of the daily limit value of 50 µg.m^{-3}. The central region ("Coimbra", "Aveiro/Ílhavo", "Litoral Noroeste do Baixo Vouga", and "Centro Litoral") and Lisbon metropolitan area ("Área Metropolitana de Lisboa Norte" and "Área Metropolitana de Lisboa Sul") registered 87% of the number of exceedances recorded in Portugal in 2021. For NO_2, the highest annual concentrations were recorded in the metropolitan area of Lisbon ("Área Metropolitana de Lisboa Norte") with 40 µg.m^{-3}, and in "Porto Litoral" with 41 µg.m^{-3}, the only agglomeration with an exceedance to the EU annual limit value.

Despite the air quality improvements achieved in recent years, with most zones/agglomerations with pollutants concentrations below the EU legislated values, it is necessary to understand whether this will continue in the future due to expected climate changes impacts. As meteorology plays an important role in chemistry, transport, and deposition of air pollutants (Jacob and Winner 2009; Von Schneidemesser et al. 2015), it is expected that climate change will affect temporal and spatial variations of air pollutants (Szopa et al. 2021). Thus, the three possible trajectories for carbon neutrality, proposed for Portugal in RNC2050 to combat climate change, must be analysed in terms of their impact on future atmospheric pollutants emissions and consequent impacts on air quality, making possible to be ahead and improve the air quality management over a region/country.

1.1.3. Urban Areas

On November 15th, 2022, according to the United Nations (UN), the world's population has reached 8 billion people, a landmark resulting from people living longer and healthier lives than ever before (UN 2022). In 2021, 56% of the world's population lived in urban areas (see Figure 1.10), and it is expected to grow to 68% by 2050 (UN-Habitat 2022). When analysing only Europe, the number is ever bigger, with 75% of European citizens living in urban areas nowadays, and projected to be 84% in 2050 (UN-Habitat 2022). Globally, urban areas are major contributors to climate change, consuming over 65% of the world's energy, responsible for more than 70% of CO_2 emissions. As a result, urban areas are facing important challenges related to pressures induced by changes on urban metabolism, as well as by climate change (Borrego et al. 2018).

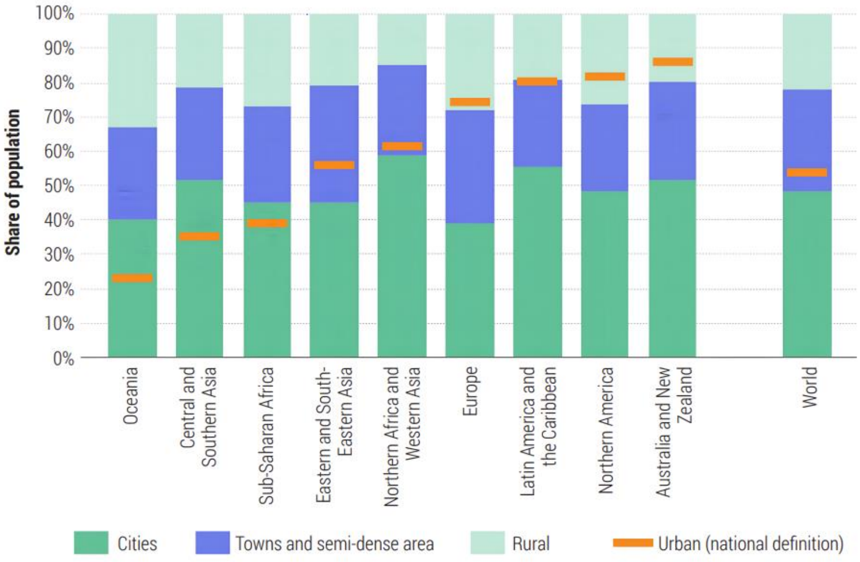

Figure 1.10. *Population by degree of urbanization and in nationally defined urban areas in 2015. Adapted from UN-Habitat (2022).*

The future of urban areas has been of concern to policy makers, researchers, and urban residents, particularly in monitoring the divergent demographic, economic, social, environmental and political pathways that will lead towards more sustainable outcomes (UN-Habitat 2022). Due to the complex interactions between social, economic, and environmental stressors, urban areas are the ones that must respond to climate change risks (Revi et al. 2014; Radhakrishnan et al. 2017, 2018; Dodman et al. 2022). Therefore, it is important that urban areas act as experimentation and innovation ecosystems to help all other areas in their transition to become climate-neutral (UN-Habitat 2022). Indeed, climate change impacts in urban areas have recently become an important and urgent research topic (Jiang et al. 2017).

Alongside climate change, air pollution has also become a growing concern, and it is expected that this issue will continue for the next decades (Von Schneidemesser et al. 2015). Unless action is taken, air pollution will be the largest environmental cause of premature death worldwide by 2050 (OECD 2012; Von Schneidemesser et al. 2015). According to the best practices for local and regional air quality management (Pisoni et al. 2022), defined by the Forum for Air quality Modelling (FAIRMODE), efficient modelling tools can be used to support air quality management in developing and implementing plans and measures to improve air quality.

Numerical models, associated with both air quality and climate change modelling, have emerged as a useful and reliable tool to understand the atmospheric physical and chemical processes from global to local scales (Borrego et al. 2018). The capability to study multi-pollutant in long-term simulations for different scenarios is crucial to anticipate the effects of climate change on air quality, to assess impacts on human health, and to evaluate adaptation measures for urban systems, highly advantageous characteristics for policy makers and stakeholders' decision making (Lacressonnière et al. 2014; Borrego et al. 2018).

1.2. RESEARCH QUESTIONS AND OBJECTIVES

The previously presented framework highlights climate change and air pollution as the current and future challenges of urban areas for environment and human health protection. Air quality management and climate change mitigation and adaptation measures should rely on scientific research support. This work contributes to enrich the knowledge in this scientific field and serves as a policy-oriented support, by answering to four main research questions:

1. Which are the best available tools to account for air pollution and climate change impacts on air quality and human health in an integrated way?
2. What is the relative contribution of emission source regions/activities to air quality, now and in the future, considering a climate change scenario?
3. What is the impact of existing climate policies on air quality and health?
4. What additional policies are needed to support win-win strategies for air quality and climate mitigation and/or adaptation, at urban scale?

The main goal of this work was to develop a modelling approach able to assess the impacts of future climate and projected emissions, including the relative contribution of different source regions and sectors, on air quality at urban scale. Within this goal, the specific objectives were:

- To better Understand the relationship between air quality and emissions by source region and sector, at urban scale;

- To identify and select the adequate urban scale modelling tools to accurately reproduce the relevant atmospheric physical and chemical processes, and at same time, to quantify the impacts of climate mitigation and/or adaptation measures on future air quality;
- To quantify the impacts of current and future air pollution exposure on human health;
- Provide recommendations for the best win-win strategies on climate and air quality management, for the future, to support urban planning oriented for citizen welfare.

The work was developed having as case study the Aveiro Region in Portugal. Aveiro Region, integrating eleven municipalities (Águeda, Albergaria-a-Velha, Anadia, Aveiro, Estarreja, Ílhavo, Murtosa, Oliveira do Bairro, Ovar, Sever do Vouga and Vagos), is an interesting and challenging case study since it is recognized as one of the regions affected by some poor air quality events in Portugal and vulnerable to climate change effects.

This work is innovative once it incorporates state-of-the-art techniques and includes climate change together with a health impact assessment in the air quality management process. Moreover, it focuses on the urban scale where population density is higher and air pollution represents a huge challenge.

Furthermore, this work is pertinent, complex and multidisciplinary and it is in accordance with the Horizon Europe Cluster 5 - Climate, Energy and Mobility. The objectives defined are also aligned with two goals of the 2030 Agenda for Sustainable Development, namely: Goal 11 - make cities and human settlements inclusive, safe, resilient, and sustainable; and Goal 13 - take urgent action to combat climate change and its impacts. More specifically, this work will help to increase awareness of climate change (goal 13.3) and air pollution in urban areas (goal 11.6). Also, the comprehensive approach of this work will improve the knowledge of integrated measures and policies for mitigation and adaptation to climate change (goal 11.B), to be included in national and regional policies, strategies and planning (goal 13.2).

1.3. OUTLINE OF THE THESIS

This document presents the main results and the discussions of the PhD thesis entitled “Assessing air quality in cities under climate change scenarios: a source apportionment approach”, which arose from a set/sequence of scientific papers. To achieve the thesis main objectives, the research structure shown in Figure 1.11 was adopted.

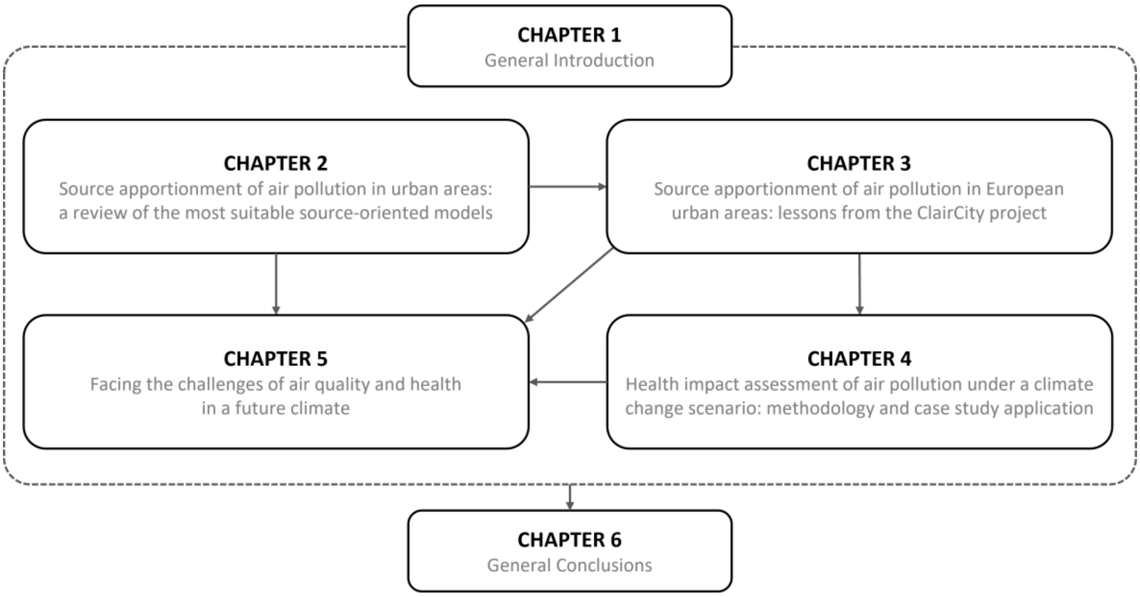

Figure 1.11. *Overview of the thesis structure.*

Chapter 1 provides an overview of the scientific context of this thesis, which will be the basis of the scientific developments presented in the following chapters. General knowledge on climate change, air quality, and their impacts on urban areas have been introduced. The main focus was placed on urban areas, where greater effects of climate change and air pollution are expected in the future, as well as more serious impacts on human health. The contents of this chapter were enriched by research and findings from other authored and co-authored publications, that are not fully included in this thesis (i.e., Borrego et al. 2018; MedECC 2020; Coelho et al. 2020, 2021, 2023a). Taking into consideration all this information, the main goal and research questions of this work have been detailed.

Chapter 2 presents a literature review of air quality studies based in source apportionment techniques. This review summarizes the differences among the different source apportionment techniques, highlights their purpose and their advantages and disadvantages. Given the growing concern about air pollution in urban areas, this study also tries to understand which source apportionment approaches are the most appropriate to assess air quality and to support the design of air quality plans in these areas. The main findings of this chapter play a key role in the development of the source apportionment methodologies applied in the work presented in Chapters 3 and 5. Furthermore, the work developed in this chapter contributed to the knowledge gain necessary to answer the first research question. The full text of this chapter has been published in the *Air quality, Atmosphere & Health* journal (Coelho et al. 2023b).

Chapter 3 focuses on the diagnosis of the current air quality, and the quantification of the main source activities' contributions to PM10, PM2.5 and NO_2, based on the application of a source apportionment tool, over six European urban areas, including the Aveiro Region. The performance evaluation of the modelling system applied in this chapter, with both meteorological and chemical transport models, allowed to improve the numerical model's accuracy to realistically reproduce the physical and chemical processes that occur in urban areas. This information was extremely relevant for the application of the numeral system presented in Chapters 4 and 5. The main conclusions presented in this chapter also allowed to improve the application of source apportionment tools applied in Chapter 5. Furthermore, this work contributed to answer research questions number 1 and 2. The full text of this chapter has been published in the *Journal of Environmental Management* (Coelho et al. 2022a).

Chapter 4 assesses air pollution effects on human health at urban scale for the middle of the century, considering the SSP2-4.5 climate change scenario and the emission projections from the Portuguese RNC2050 intermediate scenario, through the application of a numerical modelling system, having the Aveiro Region as a case study. The main conclusions of this chapter, regarding the impact of existing climate policies on air quality and human health, play an important role to answer to the first and third research questions, as well as the development of the work presented in Chapter 5. The full text of this chapter has been published in the *Sustainability* journal (Coelho et al. 2022b).

The information obtained from the previous Chapters culminated in the work presented in **Chapter 5**. This chapter presents an integrated modelling approach able to assess the impacts of future climate and projected emissions, including the relative contribution of different source regions and sectors, on air quality at urban scale, as well as their impact on human health. Moreover, additional abatement strategies to improve future air quality in Aveiro Region are provided. This work was a key contribution to answer to all research questions. This chapter has been published in the *Science of The Total Environment* journal (Coelho et al. 2023c).

Finally, the main outcomes achieved in this thesis are presented in **Chapter 6.** In addition, Chapter 6 provides suggestions of future work to be developed following the work presented throughout this thesis.

This thesis comprises adapted versions of published or submitted papers to peer-reviewed Science Citation Index (SCI) journals. The papers' adaptations only concern references and document formatting, in order to make the text easier to read. In most papers, the author was responsible for the study design, as well as for the results analysis and for the manuscript writing. The co-authors were responsible for the critical revision of the manuscript, and, when applicable, to provide initial climate data, and software support.

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CHAPTER 2

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2. SOURCE APPORTIONMENT OF AIR POLLUTION IN URBAN AREAS: A REVIEW OF THE MOST SUITABLE SOURCE-ORIENTED MODELS

Abstract

Notwithstanding the improvements already achieved in recent decades through regional and urban scale actions implemented across Europe, air pollution is still a major environment and health concern for Europeans. The quantitative assessment of the different sources of air pollution in regional/urban areas is crucial to support the design of accurate air quality plans. Source apportionment techniques are capable to relate air pollutant concentrations to existing emission sources activities and regions. The selection of the appropriate source apportionment technique to apply to a given area should take into account the ultimate goal of the study. Despite the growing number of studies that include source apportionment techniques, there is still a lack of works that summarise information on this topic in a systematic way. In this work, a literature review of studies applying SA techniques, published between 2010 and 2021, was performed. Additionally, this review summarises the differences among the different source apportionment techniques, with focus on source-oriented models, highlighting their purpose and their advantages and disadvantages. Results shows that the number of studies using source apportionment source-oriented models has been increasing across the years, with 59% using tagged species methods, 28% brute force methods, and 13% other methods. This source-oriented models have been mostly applied for PM2.5, to assess the causes of air pollution levels.

Keywords: air pollution; source apportionment; source-oriented models; urban areas; numerical modelling; review

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2.1. INTRODUCTION

Despite the already achieved improvements over recent decades, air pollution is still a major environment and health concern for Europeans. Across Europe, the levels of air pollutants are still exceeding the European Union (EU) standards prescribed by the Air Quality Directive (EEA 2020). Due to the geographical concentration of people and economic activities, which result in higher emissions from different sources, air pollution in urban areas is often higher than in other areas of a country (OECD 2020). The most serious pollutants in European urban areas, in terms of harm to human health, are particulate matter (PM) and nitrogen dioxide (NO_2). In the EU, 97% of the urban population is exposed to levels of fine PM above the latest guideline levels set by the WHO, published in September 2021 (WHO 2021). Population living in bigger cities tend also to be exposed to higher concentrations of NO_2 due to the road traffic emissions (EEA 2021). Negative impacts on respiratory and cardiovascular health and premature deaths are two of the major effects of human exposure to air pollution (WHO 2013; Kelly and Fussell 2015). In 2018, estimates of the health impacts attributable to long-term exposure to air pollution indicate that PM and NO_2 concentrations were responsible for about 417 and 55 thousand premature deaths, respectively, in the EU-28 (EEA 2020).

Although regional and urban scale actions to reduce air pollution have been implemented across Europe (e.g., Giannouli et al. 2011; Miranda et al. 2015; Borrego et al. 2016), there are still problems that need to be addressed (Thunis et al. 2019). Air pollution hotspots remain in the Po-valley region and Eastern Europe for PM and in most European big cities for NO_2 (EEA 2019). One of the key issues is to understand the origin of the pollution (Thunis et al. 2019). For that, the quantitative assessment of the different origins of air pollution in urban areas is crucial to support the design of accurate air quality plans. As indicated in the European Air Quality Directive (EC 2008), this assessment can be made through the application of source apportionment (SA) techniques. In that sense, The Forum for Air quality Modelling (FAIRMODE), a joint response initiative of the European Environment Agency (EEA) and the European Commission Joint Research Centre (JRC), developed a European guide (Mircea et al. 2020) to provide an overview and recommendations for the application of air quality models in estimating source contributions to PM and guides the choice of the most effective mitigation strategies and measures to include in air quality plans.

SA techniques are capable to relate air pollutant concentrations to existing emission source activities (e.g.: domestic heating, road transport, industries) and regions (e.g.: local, urban, metropolitan areas) and may be based on the measured concentrations of pollutants, known as receptor models, or on chemistry, transport, and dispersion models, known as source-oriented models (Mircea et al. 2020). The selection of the SA technique to be used to inform about the influence that one or more sources have in a specific area and period of time, depends on the purpose of the study. According to Belis et al. (2020), the most reported purposes for the SA applications are: (i) to assess the causes of air pollution levels, (ii) to support the design of air quality plans, (iii) to evaluate the impact of abatement measures, (iv) to quantify the contribution of different areas within a country/region, and (v) to quantify transboundary transport.

Despite the growing number of studies that include SA techniques and their relevant contributions given to this research field (e.g., Belis et al. 2014, 2020; Hopke 2016; Thunis et al. 2019; Mircea et al. 2020), there is still a lack of works that summarise information on this topic in a systematic way. To overcome this gap, in this work a literature review of studies, based on SA techniques, was performed. Furthermore, this review summarises the differences among the different SA techniques, highlights their purpose and their advantages and disadvantages. Given the growing concern about air pollution in urban areas, this study also tries to understand which SA approaches are the most appropriate to assess air quality and to support the design of air quality plans in these areas. Then, the final goal was to highlight the research needs for this field of study. The multi-analysis feature of this work, combining a literature review with a content analysis, and focusing on the purpose of air quality management in urban areas, can enrich the existing knowledge, making it innovative and relevant in the context of SA-based studies.

This review is organized as follows. Section 2.2, Materials and Methods, presents the methodology used. In Section 2.3, Source Apportionment Models and Techniques, models and techniques used in SA studies, as well as the main advantages and disadvantages of each one, are reviewed. Section 2.4, Source Apportionment Applications, compiles the main applications of the source-oriented models/techniques. Finally, Section 2.5, Conclusions and Recommendations, summarises the major findings and discusses the challenges for future SA applications.

2.2. MATERIALS AND METHODS

A literature review was performed to gather all the relevant literature to fulfil the objective of this study. This review was based on peer-reviewed papers published in international scientific journals, and was limited to articles in English, published between 2010 and 2021. Following the objective of this study, the search included the following keywords: (i) “air pollution" or "air quality" or "atmospheric pollution”; and (ii) “source contribution*" or "source apportionment" or "source oriented"; and (iii) "modelling" or "model*"; and (iv) “urban area*" or "city" or "cities”. The search was performed in order include singular/plural and related words, in the categories “title, abstract and keywords” in the Scopus database (www.scopus.pt, accessed in October 2021).

Figure 2.1 shows that a total of 557 papers were found and their abstracts were carefully read. For the 557 studies found, a simple analysis was carried out in section 2.3.1, which resulted in the exclusion of 438 papers for being considered out of the scope of this study. The remaining 119 papers were fully read, and a more detailed content analysis was performed (Sections 2.3.2 and 2.4). A full list of the 119 papers, including the main characteristics of each study, is presented in Supplementary Material (section 2.7).

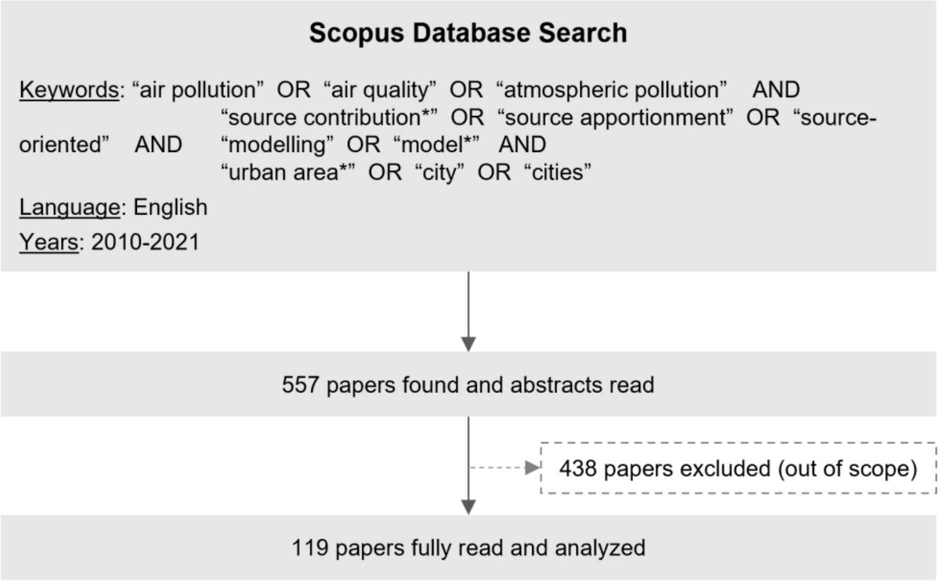

Figure 2.1. *Review process.*

To keep a coherent analysis, a data sheet was developed with the author, publishing year and content information, such as the numerical model, SA technique, case study location, scale of analysis, and pollutants analysed. This information was used to perform a detailed analysis, where descriptive statistics were derived for the year of publication, the methodology used, and the pollutants analysed.

2.3. SOURCE APPORTIONMENT MODELS AND TECHNIQUES

2.3.1. Receptor and source-oriented models: advantages and limitations

According to the SA models and techniques used, the 557 studies analysed can be divided in two major groups: (i) SA studies based on receptor models and (ii) SA studies based on source-oriented models. Figure 2.2 summarises the number of SA studies, according to the model used, during different periods between 2010 and 2021.

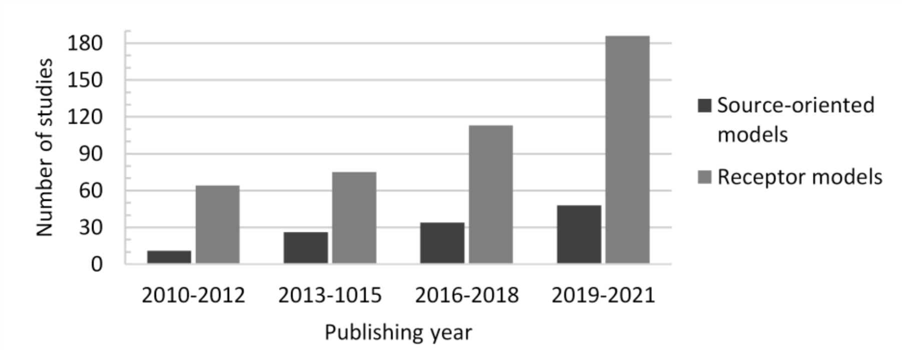

Figure 2.2. *Summary of SA studies published between 2010 and 2021.*

According to Figure 2.2, during the last 12 years, there was an increase in the number of studies using SA techniques. Overall, receptor models have been the most popular method for SA studies since 2010. On the other hand, a few source-oriented model studies have been carried out, accounting for only 21% of all studies. Each method has its own features. Receptor models, based on the mass conservation principles, are used to perform SA by analysing the chemical and physical parameters measured at one or more specific sites (receptors) (Belis et al. 2020). They include many tools ranging from simple techniques, with elementary mathematical and basic physical assumptions, to complex models requiring data processing (Mircea et al. 2020). Principal component analysis, chemical mass balance and positive matrix factorization are the most applied receptor model techniques (Belis et al. 2014; Hopke 2016). Alternatively, source-oriented models are usually based on the application of air quality models, being Eulerian, Gaussian and Lagrangian models the most commonly used (Fragkou et al. 2012). In a simple or more complex way, they try to mimic the physical and chemical processes taking place in the atmosphere in the presence of emissions of pollutants. Common source-oriented model approaches are: (i) brute force method, in which separate model runs are performed, each one considering a different set of sources of interest; and (ii) tagged species method that earmarks the mass of chemical species to track the atmospheric fate of every source throughout a unique model run (Mircea et al. 2020; Belis et al. 2020).

Although SA techniques have advanced considerably in the last years due to the growing interest in the scientific community, both receptor and source-oriented models still have some limitations inherent to their formulation and data availability (Mircea et al. 2020). The advantages and limitations of each SA technique most identified in the analysed studies are compiled in Table 2.1.

Table 2.1. *Advantages and limitations of receptor and source-oriented models.*

	Receptor models	***Source-oriented models***
Advantages	It derives information about sources from measured data It estimates the contribution of sources for most of the PM chemical components It does not require an extensive input data set (e.g., 3D meteorological data, 3D emission data, air concentrations at boundaries) It does not require significant computing resources and data storage is negligible The uncertainty of the output is estimated	It evaluates the contributions of sources in the absence of measured data It is possible to predict air quality changes in relation to emissions changes The definition of the sources depends on the emission inventories, so it can be detailed in terms of activity sectors It quantifies the contribution of transported pollutants It is possible to explore the variability in time (with high temporal resolution) and space of source contributions
Limitations	It is limited to sites where monitoring data are available It provides information for specific time windows Some methods require prior knowledge of the composition of the emission sources	It is limited by the quality of the input data (e.g., emissions, meteorology) It is limited by the formulation of the chemical transport model used It requires significant computing resources and data storage

According to Table 2.1, one of the main advantages of receptor models is related to the reduced input data set that this SA technique requires, as well as the computing resources and data storage being almost negligible. As limitation, since receptor models derive information on sources from measured data, it is limited to sites, and time periods, for which these data are available. On the other side, the application of source-oriented models does not require the use of measured data, making it possible to apply for any location and time period. Another advantage of source-oriented models is the possibility to predict air quality impacts from emission changes, as well as the quantification of the contribution from different activity sectors or the transport of pollutants. However, source-oriented models are limited by the quality of the input data and the formulation of the chemical transport models used. Also, the significant amount of computing resources and data storage can be another limitation of this SA technique.

Based on the findings of Table 2.1, and considering the objective of this study to understand which SA approaches are the most appropriate to assess air quality in urban areas and to support the design of air quality plans, the following sections will focus on the analysis of papers with source-oriented models, 119 out of a total of 557 papers.

2.3.2. Source-oriented models: brute force and tagged species methods

The 119 studies with SA source-oriented models, identified in the previous section, can be divided in 3 major groups, according to the type of method used, namely: (i) brute force method; (ii) tagged species method; and (iii) other methods such as path integral method, response surface modelling, among others.

The underlying question related to SA brute force method is: "*What would be the reduction in the pollutant concentrations corresponding to a given reduction in the emissions of its precursors?*", as stated by Belis et al. (2020). Brute force method consists of running a model simulation with all emissions (baseline) and then performing several additional model simulations, each one varying emissions from a defined activity source and/or the geographical location. The difference between the results from the baseline and each additional simulation is considered as the contribution of that source (Mircea et al. 2020). This method, also known as sensitivity analysis, can be used with any numerical model, without the need of a specific model module to execute the brute force method approach (Belis et al. 2020).

For the SA tagged species method, Belis et al. (2020) defined the underlying question as: "*What is the actual mass transferred from a pollutant source to its concentration in a given location and period?*". This methodology is designed for SA purposes, using labels in each precursor in every time step according to its activity source and/or the geographical origin, making it possible to quantify the mass contributed by every source/area to the pollutant concentration (Mircea et al. 2020). Tagged species method is based on the mass balance equation, ensuring the sum of the concentrations corresponding to each source is always equal to the total concentration due to all sources (Yarwood et al. 2007). Several Eulerian Chemical Transport Models (CTMs), such as CAMx PSAT (Yarwood et al. 2007; ENVIRON 2016), CMAQ-ISAM (Kwok et al. 2013; Appel et al. 2018) and LOTOS-EUROS (Kranenburg et al. 2013; Manders-Groot et al. 2016), have already a SA tagged species algorithm implemented.

Figure 2.3 summarizes the number of SA studies, according to the source-oriented model used, during different periods between 2010 and 2021.

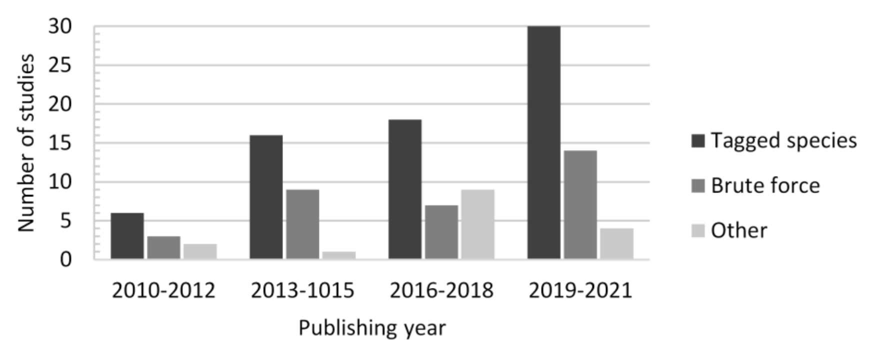

Figure 2.3. *Summary of SA source-oriented models studies published between 2010 and 2021.*

As in the previous section analysis, the number of studies using SA source-oriented models has been increasing across the years. Tagged species method has been the most used in the 119 SA studies analysed, accounting for 59% of all studies, while studies using brute force methods correspond to only 28%. The remaining 13% of the studies use other methods. The use of tagging and brute force methods has been constantly increasing over time, while the application of other approaches is more recent and varies over the years, according to Figure 2.3.

The advantages and limitations of brute force and tagged species methods most identified in the analysed studies are compiled in Table 2.2.

Table 2.2. *Advantages and limitations of brute force and tagged species methods.*

	Brute force method	***Tagged species method***
Advantages	Can be used to evaluate the impact of abatement measures Can be used with any numerical model	Apportion all the sources in one single run Assess the contributions referred to the specific emission inventory and meteorological fields used as input Can be used to attribute the actual impacts of sources on health and vegetation
Limitations	Requires as many runs as the sources to apportion plus the run with the base case (all sources) Due to the non-linear behaviour, the sum of source contributions may not match the total pollutant mass obtained in the base case (mass is not always conserved) Dependent on the sectorial detail available in the emissions inventory	Dependent on the sectorial detail available in the emissions inventory Could require additional coding efforts For non-linear pollutants, the source contribution cannot be extrapolated to situations different than the modelled case

One of the advantages of using brute force methods is the evaluation of the impact of abatement measures, and the possibility to use this SA technique with any numerical model. As limitation, brute force method requires the base case run (with all sources) plus as many runs as the sources to apportion. Also, due to the non-linear behaviours, the sum of the source concentrations allocated in each single run can differ from the concentrations of the pollutants in the base case run. On the other hand, tagged species method can apportion all sources in one single run. Another advantage of tagged species method is that it can be used to attribute the actual impacts of sources on health and vegetation. However, this SA technique could require additional coding efforts and is dependent on the sectorial detail available in the emissions inventory.

In summary, tagged species method quantifies the mass that is transferred from the source to the receptor. For that reason, according to Belis et al. (2020), tagged species methods can be grouped under the category of mass-transfer SA. In opposition, the brute force method is a sensitive analysis that estimates the changes in concentrations that would result from a change in emissions (Belis et al. 2020).

2.4. SOURCE APPORTIONMENT APPLICATIONS

2.4.1. Main purposes

The 119 papers analysed in this section reported several purposes for the applications of SA techniques. Table 2.3 summarizes the main purpose of the SA studies, divided by the source-oriented models used, i.e., tagged species method, brute force method or other, published between 2010 and 2021.

Table 2.3. *Summary of the main purpose of the SA studies, using source-oriented models, published between 2010 and 2021.*

Purpose of the study	***Total number of studies***	***Source-oriented models***		
		Tagged species method	***Brute force method***	***Others***
Assess the causes of air pollution levels	42	24	13	5
Quantify the contribution of different areas within a country/region	18	11	5	2
Quantify transboundary transport	10	8	0	2
Evaluate the impact of abatement measures	11	6	4	1
Support the design of air quality plans	3	1	1	1
Others	35	20	10	5

Table 2.3 shows that most of the papers applied SA source-oriented models to assess the causes of air pollution levels (42; ~35%), of which 24 used tagged species methods (e.g., Huang et al. 2012a; Wang et al. 2014a, 2019a; Yang et al. 2020), 13 used brute force methods (Wang et al. 2014b, 2015; Zhang et al. 2014b; Lu et al. 2019b), and 5 used other methods (e.g., Lee et al. 2014; Qiao et al. 2018). Following, 18 (~15%) papers aimed to quantify the contribution of different areas within a country/region, with 11 of these papers using tagged species methods (e.g., Valverde et al. 2016; Zhang et al. 2017), 5 using brute force methods (e.g., Huang et al. 2018; Wang et al. 2020b), and 2 using other methods (e.g., Zhu et al. 2018). The quantification of transboundary transport was the purpose of 10 papers, but only using tagged species methods (8 papers) (e.g., Wagstrom and Pandis 2011; Chen et al. 2017a), and other methods (2 papers) (e.g., Dunker et al. 2017). 6 papers used tagged species methods to evaluate the impact of abatement measures (e.g., Li et al. 2013; Zhang et al. 2014a). For the same purpose, 4 papers used brute force methods (e.g., East et al. 2021), and only 1 used other method (Xing et al. 2011). SA source-oriented models were also applied to support the design of air quality plans in 3 papers, of which Lu et al. (2019a) used tagged species method, Borge et al. (2014) used brute force method, and You et al. (2017) used other method. Finally, 35 papers applied SA source-oriented models for other purposes, like Bove et al. (2014) that used tagged species method to compare and to integrate receptor models and CTMs, or Minoura et al. (2016) that investigated the vertical circulation of atmospheric pollutants using a brute force method, among others.

2.4.2. Pollutants

The pollutants analysed in the SA source-oriented models' studies reviewed in this work are summarized in Table 2.4. Some studies analysed multiple pollutants, so the total number of the pollutants may be greater than the total number of studies.

Table 2.4. *Summary of pollutants analysed in the SA source-oriented models studies published between 2010 and 2021.*

Source-oriented models	Number of studies	Pollutants							
		PM10	PM2.5	O_3	NO_2 / NOx	SO_2	VOC	CO	Other
Tagged species method	70	8	43	24	10	4	4	3	11
Brute force methods	33	6	22	9	5	3	1	1	7
Others	16	2	11	5	1	2	1	0	2

SA source-oriented models have been applied for many air pollutants such as PM10 (e.g., Cheng et al. 2013a - with tagged species method; Guttikunda et al. 2019 - with brute force method), O_3 (e.g., Coelho et al. 2017 - with tagged species method; Wang et al. 2020b - with brute force method), NO_2 and/or NOx (e.g., Rafee et al. 2017 - with brute force method; Parvez and Wagstrom 2019 - with tagged species method), among others. However, most of the studies focus on PM2.5, such as Jat and Gurjar (2021) that, using the brute force method, quantified the contribution of emissions from major source sectors and source regions of Indo-Gangetic Plain to local and regional PM2.5 pollution during winter; and Qiao et al. (2021) that quantified the contributions from different sectors and regions to PM2.5 in the Sichuan Basin, using the tagged species method.

2.4.3. Case studies' locations

The location of case studies included in the papers was also evaluated (Figure 2.4). In order to simplify this analysis, the case study locations, which range from one to several cities in more than one country, have been grouped by country. Some papers used multiple case studies, located in different countries, so the total number of case studies may be greater than the total number of studies. Figure 2.4 shows that most of the case studies (65; ~42%) are located in China. Following, 22 (~14%) case studies are located in the United States, 8 (~5%) in Italy and 6 (~4%) in Portugal. When analysing the location of each case study by SA source-oriented model, no relationship was found between the use of a specific method and the location of the case study.

Figure 2.4. *Summary of case studies location, grouped by country, used in the SA source-oriented models studies published between 2010 and 2021.*

According to WHO (2016), China, here identified as the most used case study location, is considered the region of the world with more air pollution-related premature deaths. China is also located in the world region where the highest levels of air pollution are recorded. Despite these relationships, there is no clear relationship between the case studies locations of the analysed papers and the places with poorer air quality and, consequently, more associated premature deaths.

2.4.4. Case studies' periods

Figure 2.5 presents the summary of case studies' periods, used in the SA source-oriented model studies analysed. To simplify this analysis, the periods were clustered in three different types, namely: (i) all year, that includes only studies that used the entire year; (ii) months(s), that includes studies with one or more analysed months, or an entire season, but never an entire year; and (iii) day(s), including studies that analysed short periods and/or pollution episodes (always less than a month) or used representative days to characterize a specific period. Some studies analysed multiple periods (but always from the same type of period clustering), so the total number of the periods may be greater than the total number of studies. The case studies' periods belong to years from 1970 to 2020. One of the studies (Guttikunda et al. 2019) also analysed future years, until 2030, where emission projections were considered, but without taking into account the variations in meteorology caused by climate change.

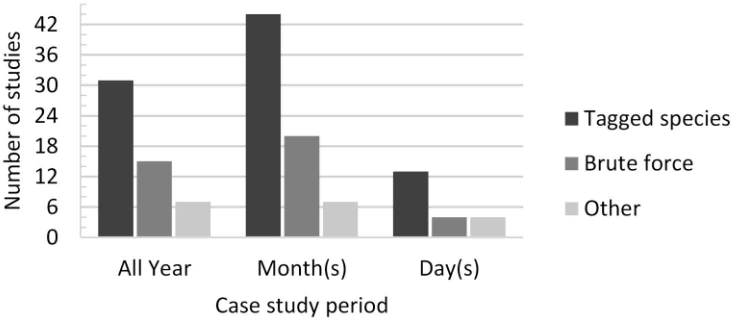

Figure 2.5. *Summary of case studies periods, used in the SA source-oriented models studies published between 2010 and 2021.*

Figure 2.5 shows that most of case studies' periods (71, ~49%) are less than a year, ranging from one to several months or an entire season, of which 44 used tagged species methods (e.g., Du et al. 2020; Shen et al. 2020; Bai et al. 2021), 20 used brute force methods (e.g., Wang et al. 2014b; Dolwick et al. 2015; East et al. 2021), and 7 used other methods (e.g., Dunker et al. 2017; Langner et al. 2020). Following, 53 (~37%) case studies' periods have focused on one or more entire years, with 31 of these being analysed with tagged species methods (e.g., Lang et al. 2017; Lonati et al. 2020; Jiang et al. 2021), 15 with brute force methods (e.g., Cho et al. 2012; Wang et al. 2018a; Zhou et al. 2021), and 7 with other methods (e.g., Liu et al. 2018; Wang et al. 2020a). Finally, only 21 (~14%) case studies have periods with several days (Martins et al. 2015 - with tagged species method; Baker et al. 2016 - with brute force method; Liu et al. 2017 - with other method).

2.4.5. Discussion

From the 119 papers fully analysed in this review, tagged species methods are the most used SA technique. Although tagged species method has been applied for several purposes, the most common is the assessment of the causes of air pollution levels. According to its formulation, tagged species methods can be an asset in quantifying transboundary transport and the contribution of different areas within a country/region. Brute force methods, less applied in the 119 analysed papers, are also mostly used in the assessment of the causes of air pollution levels. Contrary to the tagged species method, the added value of brute force method is the capability to evaluate the impact of abatement measures, which is one of the keys to support the design of air quality plans.

Both tagged species and brute force methods have been used for many pollutants such NO_2, O_3, and SO_2. However, most of the studies focus on PM due to the high number of exceedances and pollution episodes and the higher experience of SA techniques on this pollutant, mostly due to the FAIRMODE work (Mircea et al. 2020), that provides an overview on air pollution SA for PM. The location and time period analysed for each case study varies according to the air quality problems identified by the authors, and no relationship was found between these characteristics of the study and the SA method chosen. In most cases, the location of the case study seems to be related to the affiliation location of the authors of the works.

In this sense, the selection of the best SA technique to be used in each study must be defined based on its purpose. Due to its ability to quantify the transboundary transport, in addition to contributions from several source regions and sectors, in a single run, the tagged species method may be a better choice when the objective of a study is a more complete diagnosis of the air quality. On the other hand, if the objective is to evaluate the impact of abatement measures and/or to support the design of air quality management plans, brute force method must be chosen.

2.5. CONCLUSIONS AND RECOMMENDATIONS

Due to the growing number applications of SA techniques and the lack of works that summarize information on this topic in a systematic way, a literature review of studies, from 2010 to 2021, that include SA techniques was performed in this work. The papers analysis shows that the number of studies using SA source-oriented models has been increasing across the years, with tagged species (59%) and brute force (28%) methods being the most used models in the majority of the 119 fully analysed studies. Also, other methods (13%) such as path integral method or response surface modelling, was applied in some studies. From the content analysis, it is possible to conclude that both SA source-oriented models have been mostly applied for PM2.5, to assess the causes of air pollution levels. Approximately 42% of the case studies are located in China and the most used time period of analysis covers month(s) and/or seasons.

Given the growing concern about air pollution in urban areas, another objective of this study was to assess the main advantages and limitations of each SA source-oriented model, to understand which one is the most appropriate to assess air quality and to support the design of air quality plans in urban areas. The tagged species method appears to be the one that provides a more complete assessment of air quality, identifying contributions from both source regions and sectors, as well as the transboundary transport. However, this method is not capable of evaluate the impact of abatement measures to support the design of air quality plans, requiring the use of brute force methods. Therefore, to better assess air quality and support the design of air quality management plans in urban areas, the two methods must be used in a complementary way.

The final goal of this review was to highlight the needs for future source apportionment applications. One of the recommendations is that longer time periods should be considered in the SA analysis since results based on day(s), month(s) and season(s) are indicative of that period patterns and contributions, but they should not be linearly extrapolated to other specific periods, as they do not fully describe the role of transboundary transport and the defined source sectors and/or regions. In addition, since air pollutants dispersion is highly driven by climate-related events and it is expected that climate change will affect future air quality patterns, SA works considering future years, with both emission and meteorological projections, should be done.

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2.7. SUPPLEMENTARY MATERIAL

▪ MATERIALS AND METHODS

Table 2.5. *Full list of the 119 papers analysed, including the main characteristics of each study.*

Reference	*Title*	*Year of publication*	*Models*	*SA type*	*Case study*	*Year of study*	*Spatial resolution*	*Pollutants*	*Purpose*
East et al. (2021)	Air quality modelling to inform pollution mitigation strategies in a Latin American megacity	2021	CMAQ	BFM	Bogota, Colombia	2014 (6 months)	1 km	PM2.5	Evaluate the impact of abatement measures
Dimitrova and Velizarova (2021)	Assessment of the contribution of different particulate matter sources on pollution in Sofia city	2021	ADMS-Urban	BFM	Sofia, Bulgaria	2014 (all year)	50 m	PM10; PM2.5	Assess the causes of air pollution levels
Yang et al. (2021)	Characteristics of regional transport during two-year wintertime haze episodes in North China megacities	2021	NAQPMS	BFM	Beijing–Tianjin–Hebei, China	2017 (2 months); 2018 (1 month)	-	PM2.5	Assess the causes of air pollution levels
Jat and Gurjar (2021)	Contribution of different source sectors and source regions of Indo-Gangetic Plain in India to PM2.5 pollution and its short-term health impacts during peak polluted winter	2021	WRF-Chem	BFM	India	2016 (1 month)	20 km	PM2.5	Assess the causes of air pollution levels
Kumar et al. (2021)	Contributions of international sources to PM2.5 in South Korea	2021	GEOS-Chem	BFM	Republic of Korea	2015-2016 (all years)	0.25° x 0.3125°	PM2.5	Assess the causes of air pollution levels
Kim et al.(2021)	Effects of vertical turbulent diffusivity on regional PM2.5 and O_3 source contributions	2021	CAMx	TAG	Seoul, South Korea	2016 (1 month)	9 km	PM2.5; O_3	Others
Zhao et al. (2021)	Impacts of COVID-19 on air quality in mid-eastern China: An insight into meteorology and emissions	2021	WRF-Chem	BFM	Mid-eastern China	2019 (1 month); 2020 (1 month)	9 km	PM2.5	Others

Yan et al. (2021)	Impacts of synoptic circulations on summertime ozone pollution in Guanzhong Basin, northwestern China	2021	WRF-CAMx	TAG	Guangzhou, China	2018 (3 months)	12 km	O_3	Quantify the contribution of different areas within a country/region
Zheng et al. (2021)	Long-range transport of ozone across the eastern China seas: A case study in coastal cities in southeastern China	2021	CMAQ	TAG	China	2017 (14 days)	9 km	O_3	Quantify transboundary transport
Li et al. (2021)	Modelling air quality during the EXPLORE-YRD campaign – Part II. Regional source apportionment of ozone and PM2.5	2021	CMAQ	TAG	Yangtze River Delta, China	2018 (2 months)	12 km	PM2.5; O_3	Assess the causes of air pollution levels
Lu et al. (2021)	Nonlinear response of SIA to emission changes and chemical processes over eastern and central China during a heavy haze month	2021	NAQPMS	BFM	Eastern and Central China	2014 (1 month)	27 km	SIA (Secondary Inorganic Aerosols)	Quantify the contribution of different areas within a country/region
Gong et al. (2021)	Quantifying the impacts of inter-city transport on air quality in the Yangtze River Delta urban agglomeration, China: Implications for regional cooperative controls of PM2.5 and O_3	2021	CMAQ	TAG	Yangtze River Delta, China	2018 (1 months)	12 km	PM2.5; O_3	Quantify the contribution of different areas within a country/region
Zhou et al. (2021)	Real-time numerical source apportionment of PM2.5 concentrations over the Yangtze River Delta region, China	2021	RAEMS	BFM	Yangtze River Delta, China	2019 (all year)	6 km	PM2.5	Quantify the contribution of different areas within a country/region
Qiao et al. (2021)	Revealing the origin of fine particulate matter in the Sichuan Basin from a source-oriented modeling perspective	2021	CMAQ	TAG	Sichuan Basin, China	2015 (all year)	12 km	PM2.5	Assess the causes of air pollution levels
Bai et al. (2021)	Spatial-temporal variation characteristics of air pollution and apportionment of contributions by different sources in Shanxi province of China	2021	CAMx	TAG	Shanxi, China	2012 (2 months); 2015 (2 months)	9 km	PM2.5; PM10; SO_2; NO_2	Evaluate the impact of abatement measures

Kitagawa et al. (2021)	Source apportionment modelling of PM2.5 using CMAQ-ISAM over a tropical coastal-urban area	2021	CMAQ	TAG	Southeast Brazil	2019 (2 months); 2020 (2 months)	1 km	PM2.5	Assess the causes of air pollution levels
Chen et al. (2021)	Source apportionment of fine secondary inorganic aerosol over the Pearl River Delta region using a hybrid method	2021	CAMx	TAG	Pearl River Delta, China	2015 (4 months)	3 km	SIA	Others
Ning et al. (2021)	Study on the influence of regional transportation on PM2.5 based on the RAMS-CMAQ model in Weihai, a typical coastal city of northern China	2021	CMAQ	TAG	Weihai, China	2018 (2 months)	1.5 km	PM2.5	Quantify the contribution of different areas within a country/region
Fernandes et al. (2021)	The air pollution modelling system URBAIR: how to use a Gaussian model to accomplish high spatial and temporal resolutions	2021	URBAIR	BFM	Estarreja, Portugal	2017 (all year)	200 m	PM10; NO_2	Others
Jiang et al. (2021)	Understand the local and regional contributions on air pollution from the view of human health impacts	2021	CMAQ	TAG	Beijing-Tianjin-Hebei, China	2017 (all year)	9 km	PM2.5	Quantify transboundary transport
Lang et al. (2021)	Understanding the impact of vehicular emissions on air pollution from the perspective of regional transport: A case study of the Beijing-Tianjin-Hebei region in China	2021	CAMx	TAG	Beijing-Tianjin-Hebei, China	2014 (4months)	9 km	CO; NH_3; SO_2; NOx; Elemental Carbon; PM2.5; Ammonium; Sulfate; Nitrate	Quantify the contribution of different areas within a country/region
Zhong et al. (2020)	Characteristics and source apportionment of PM2.5 and O_3 during winter of 2013 and 2018 in Beijing	2020	CAMx	TAG	Beijing, China	2013 (1 month); 2018 (1month)	12 km	PM2.5; O_3	Evaluate the impact of abatement measures
Lonati et al. (2020)	Combined eulerian-lagrangian hybrid modelling system for pm2.5 and elemental carbon source apportionment at the urban scale in Milan	2020	CAMx; AUSTRAL	TAG	Milan, Italy	2010 (all year)	1.7 km	PM2.5; Elemental Carbon	Assess the causes of air pollution levels

Du et al. (2020)	Effects of Regional Transport on Haze in the North China Plain: Transport of Precursors or Secondary Inorganic Aerosols	2020	NAQPMS	TAG	North China	2017 (2 months)	9 km	SIA	Quantify transboundary transport
Shen et al. (2020)	Insights into source origins and formation mechanisms of nitrate during winter haze episodes in the Yangtze River Delta	2020	CMAQ	TAG	Yangtze River Delta, China	2015 (3 months); 2016 (1 month)	36 km	Nitrate	Evaluate the impact of abatement measures
Wang et al. (2020a)	Mapping ozone source-receptor relationship and apportioning the health impact in the Pearl River Delta region using adjoint sensitivity analysis	2020	CMAQ	OTH	Pearl River Delta, China	2010 (all year)	3 km	O_3	Others
Langner et al. (2020)	Model-simulated source contributions to PM2.5 in Santiago and the central region of Chile	2020	WRF-Chem	OTH	Santiago, Chile	2012 (6 months)	2 km	PM2.5; NOx; SO_2; O_3	Assess the causes of air pollution levels
Yang et al. (2020)	Numerical study of air pollution over a typical basin topography: Source appointment of fine particulate matter during one severe haze in the megacity Xi'an	2020	CAMx	TAG	Xi'an, China	2016 (1 month); 2017 (2 months)	3 km	PM2.5	Assess the causes of air pollution levels
Daneshpajooh et al. (2020)	PM dispersion during stable winter episodes in Tehran and effect of governmental emission regulations	2020	CMAQ	BFM	Tehran	2010 (1 month)	2 km	PM	Evaluate the impact of abatement measures
Wang et al. (2020b)	Regional source apportionment of summertime ozone and its precursors in the megacities of Beijing and Shanghai using a source-oriented chemical transport model	2020	CMAQ	BFM	Beijing, China	2013 (1 month)	36 km	O_3	Quantify the contribution of different areas within a country/region
Guo et al. (2020)	Spatial distribution and source contributions of PM2.5 concentrations in Jincheng, China	2020	CALPUFF	OTH	Jincheng, China	2017 (2 months)	3 km	PM2.5	Quantify the contribution of different areas within a country/region

Xia (2020)	Temporal and Spatial Distribution Characteristics of PM2.5 Concentration in Tianjin and Simulation Analysis of Sources of Heavy Pollution Process	2020	WRF-Chem	BFM	Tianjin, China	2016 (3 months)	9 km	PM2.5	Assess the causes of air pollution levels
Parvez and Wagsrom (2019)	A hybrid modeling framework to estimate pollutant concentrations and exposures in near road environments	2019	CAMx; R-LINE	TAG	Connecticut	2011 (all year)	12 km (CAMx); 40 m (R-LINE)	NOx; PM2.5; Elemental Carbon	Others
Guttikunda et al. (2019)	Air quality, emissions, and source contributions analysis for the Greater Bengaluru region of India	2019	CAMx	BFM	Greater Bengaluru Region, India	2015 - 2030 (all years)	1 km	PM10; PM2.5	Others
Chang et al. (2019)	Contributions of inter-city and regional transport to PM2.5 concentrations in the Beijing-Tianjin-Hebei region and its implications on regional joint air pollution control	2019	CMAQ	TAG	Beijing-Tianjin-Hebei, China	2014 (all year)	12 km	PM2.5	Assess the causes of air pollution levels
Guo et al. (2019)	Contributions of local and regional sources to PM2.5 and its health effects in north India	2019	CMAQ	TAG	North India	2015	12 km	PM2.5	Quantify the contribution of different areas within a country/region
Lu et al. (2019a)	Differences in concentration and source apportionment of PM 2.5 between 2006 and 2015 over the PRD region in southern China	2019	CAMx	TAG	Pearl River Delta, China	2006 (all year); 2015 (all year)	3 km	PM2.5	Support the design of air quality plans
Pepe et al. (2019a)	Enhanced air quality modelling through AUSTAL2000 model in Milan urban area	2019	CAMx; AUSTRAL	TAG	Milan, Italy	2010 (all year)	1.7 km	PM2.5	Quantify the contribution of different areas within a country/region
Pepe et al. (2019b)	Enhanced CAMx source apportionment analysis at an urban receptor in Milan based on source categories and emission regions	2019	CAMx	TAG	Milan, Italy	2010 (all year)	1.7 km	NO_2; PM2.5	Assess the causes of air pollution levels
Liu et al. (2019)	Episode analysis of regional contributions to tropospheric ozone in Beijing using a regional air quality model	2019	CMAQ	TAG	Beijing, China	2015 (1-10/Jul)	16 km	O_3; NOx; VOC	Quantify the contribution of different areas within a country/region

Shu et al. (2019)	Episode study of fine particle and ozone during the CAPUM-YRD over Yangtze River Delta of China: Characteristics and source attribution	2019	CAMx	TAG	Yangtze River Delta, China	2016 (3 months)	9 km	PM2.5; O_3	Assess the causes of air pollution levels
Lu et al. (2019b)	Exploring 2016-2017 surface ozone pollution over China: Source contributions and meteorological influences	2019	GEOS-Chem	BFM	China	2016 (all year); 2017 (all year)	0.25° x 0.3125°	O_3	Assess the causes of air pollution levels
Ivey et al. (2019)	Investigating fine particulate matter sources in Salt Lake City during persistent cold air pool events	2019	CMAQ	OTH	USA	2007 (all year)	36 km	PM2.5	Others
Qiao et al. (2019a)	Local and regional contributions to fine particulate matter in the 18 cities of Sichuan Basin, southwestern China	2019	CMAQ	TAG	Sichuan Basin, China	2014 (1 month); 2015 (5 months)	12 km	PM2.5	Quantify the contribution of different areas within a country/region
Li et al. (2019)	Ozone source apportionment over the Yangtze River Delta region, China: Investigation of regional transport, sectoral contributions and seasonal differences	2019	CAMx	TAG	Yangtze River Delta, China	2015 (all year)	12 km	O_3; NOx; VOC	Assess the causes of air pollution levels
Wang et al. (2019a)	Source apportionment of summertime ozone in China using a source-oriented chemical transport model	2019	CMAQ	TAG	China	2013 (1 month)	36 km	O_3	Assess the causes of air pollution levels
Wang et al. (2019b)	Source estimation of SO42− and NO_3− based on monitoring-modeling approach during winter and summer seasons in Beijing and Tangshan, China	2019	CAMx	TAG	Beijing and Tangshan, China	2017 (6 months)	12 km	PM2.5	Others
Qiao et al. (2019b)	Spatial-temporal variations and source contributions to forest ozone exposure in China	2019	CMAQ	TAG	China	2013 (all year)	36 km	O_3	Others
Collet et al. (2018)	Future year ozone source attribution modeling study using CMAQ-ISAM	2018	CMAQ; CAMx	TAG	USA Cities	2011 (all year)	12 km	O_3	Assess the causes of air pollution levels

Wang et al. (2018b)	How aerosol direct effects influence the source contributions to PM2.5 concentrations over Southern Hebei, China in severe winter haze episodes	2018	WRF-Chem	BFM	Southern Hebei, China	2013 (1 month)	12 km	PM2.5	Others
Ying et al. (2018)	Improve regional distribution and source apportionment of PM2.5 trace elements in China using inventory-observation constrained emission factors	2018	CMAQ	TAG	China	2012 (2 months); 2013 (all year)	36 km	PM2.5	Others
Han et al. (2018)	Modeling study of impacts on surface ozone of regional transport and emissions reductions over North China Plain in summer 2015	2018	CMAQ	TAG	North China Plain	2015 (1 month)	16 km	O_3	Assess the causes of air pollution levels
Huang et al. (2018)	Numerical simulations for the source apportionment and control strategies of PM2.5 over Pearl River Delta, China, part I: Inventory and PM2.5 sources apportionment	2018	CMAQ	BFM	Pearl River Delta, China	2010 (1 month)	3 km	PM2.5	Quantify the contribution of different areas within a country/region
Thunis et al. (2018)	PM2.5 source allocation in European cities: A SHERPA modelling study	2018	CHIMERE	OTH	EU cities	2009	7 km	PM2.5	Assess the causes of air pollution levels
Qiao et al. (2018)	Source apportionment of PM2.5 for 25 Chinese provincial capitals and municipalities using a source-oriented Community Multiscale Air Quality model	2018	CMAQ	OTH	25 municipalities in China	2013 (all year)	36 km	PM2.5	Assess the causes of air pollution levels
Wang et al. (2018a)	Source Contributions to PM2.5 under Unfavorable Weather Conditions in Guangzhou City, China	2018	CAMQ	BFM	Guangzhou, China	2013-2016 (all years)	9 km	PM2.5	Others
Zhu et al. (2018)	Sources of particulate matter in China: Insights from source apportionment studies published in 1987–2017	2018	CMAQ; CAMx; ADMS; etc	-	China	-	-	PM	Quantify the contribution of different areas within a country/region

Liu et al. (2018)	Spatial-temporal variation characteristics of air pollution in Henan of China: Localized emission inventory, WRF/Chem simulations and potential source contribution analysis	2018	WRF-Chem	OTH	Henan, China	2012 (all year)	3 km	PM2.5	Assess the causes of air pollution levels
Dunker et al. (2017)	Contributions of foreign, domestic and natural emissions to US ozone estimated using the path-integral method in CAMx nested within GEOS-Chem	2017	CAMx	OTH	North America	2010 (7 months)	12 km	O_3	Quantify transboundary transport
Rafee et al. (2017)	Contributions of mobile, stationary and biogenic sources to air pollution in the Amazon rainforest: A numerical study with the WRF-Chem model	2017	WRF-Chem	BFM	Manaus, Brazil	2014 (3 days)	3 km	CO; NOx; SO_2; O_3; PM2.5; PM10; VOCs	Assess the causes of air pollution levels
Lang et al. (2017)	Development and application of a new PM2.5 source apportionment approach	2017	CAMx	TAG	North China	2011-2013 (all years)	3 km	PM2.5	Others
Chen et al. (2017a)	Estimating the contribution of regional transport to PM2.5 air pollution in a rural area on the North China Plain	2017	CMAQ	TAG	North China Plain	2013 (35 days)	9 km	PM2.5	Quantify transboundary transport
Coelho et al. (2017)	Identification and analysis of source contributions to the air quality in the Amsterdam region	2017	CAMx	TAG	Amsterdam, Netherlands	2010 (8-12/Jul)	5 km	O_3	Assess the causes of air pollution levels
Wang et al. (2017)	Local and regional contributions to fine particulate matter in Beijing during heavy haze episodes	2017	CAMx	TAG	Beijing, China	2013 (17 days)	12 km	PM2.5	Quantify transboundary transport
You et al. (2017)	Response surface modeling-based source contribution analysis and VOC emission control policy assessment in a typical ozone-polluted urban Shunde, China	2017	CMAQ	OTH	Pearl River Delta, China	2014 (1 month)	3 km	O_3	Support the design of air quality plans
Chen et al. (2017b)	Ship emission inventory and its impact on the PM2.5 air pollution in Qingdao Port, North China	2017	WRF-Chem	BFM	Qingdao, China	2014 (4 months)	3 km	PM2.5	Others

Nopmongcol et al. (2017)	Source contributions to United States ozone and particulate matter over five decades from 1970 to 2020	2017	CAMx	TAG	USA	1970; 1980; 1990; 2000; 2005; 2020 (all years)	36 km	O_3; PM2.5	Assess the causes of air pollution levels
Liu et al. (2017)	Source-receptor relationships for PM2.5 during typical pollution episodes in the Pearl River Delta city cluster, China	2017	WRF-Chem	OTH	Pearl River Delta, China	2012; 2013; 2014 (1 day each year)	3 km	PM2.5	Quantify transboundary transport
Zhang et al. (2017)	The source apportionment of primary PM2.5 in an aerosol pollution event over Beijing-Tianjin-Hebei region using WRF-Chem, China	2017	WRF-Chem	TAG	Beijing-Tianjin-Hebei, China	2015 (14 days)	12 km	PM2.5	Quantify the contribution of different areas within a country/region
Baker et al. (2016)	Contribution of regional-scale fire events to ozone and PM2.5 air quality estimated by photochemical modeling approaches	2016	CMAQ	BFM	USA	2011 (52 days)	12 km	O_3	Others
Pepe et al. (2016)	Development and application of a high resolution hybrid modelling system for the evaluation of urban air quality	2016	CAMx; AUSTAL2000	TAG	Milan, Italy	2010 (all year)	1.7 km (CAMx) 20m (AUSTRAL)	NOx	Others
Diamantopoulou et al. (2016)	Estimation of the local and long-range contributions to particulate matter levels using continuous measurements in a single urban background site	2016	CAMx	TAG	4 European cities	2010 (1 month); 2009 (1 month)	36 km	PM2.5; Elemental Carbon	Others
Lin et al. (2016)	Local and distant source contributions to secondary organic aerosol in the Beijing urban area in summer	2016	CAMx	TAG	Beijing, China	2007 (1 month)	9 km	O_3; NOx; VOCs; SOA	Others
Thunis et al. (2016)	On the design and assessment of regional air quality plans: The SHERPA approach	2016	CHIMERE	OTH	EU cities	2010 (all year)	7 km	PM2.5	Others
Valverde et al. (2016)	Ozone attributed to Madrid and Barcelona on-road transport emissions: Characterization of plume dynamics over the Iberian Peninsula	2016	CMAQ	TAG	Iberian Peninsula	2012 (6 days)	4 km	O_3	Quantify the contribution of different areas within a country/region

Vijayaraghavan et al. (2016)	Source apportionment of emissions from light-duty gasoline vehicles and other sources in the United States for ozone and particulate matter	2016	CAMx	TAG	Eastern USA	2008 (2 months)	12 km	PM2.5; VOC; NOx; CO; SO_2	Evaluate the impact of abatement measures
Wen et al. (2016)	Source apportionment of PM2.5 in Tangshan, China—Hybrid approaches for primary and secondary species apportionment	2016	CAMx	TAG	Tangshan, China	2012 (2 months); 2013 (2 months)	9 km	PM2.5	Others
Lu and Fung (2016)	Source apportionment of sulfate and nitrate over the pearl river delta region in China	2016	CAMx	TAG	Pearl River Delta, China	2011 (2 months)	3 km	PM	Quantify the contribution of different areas within a country/region
Li et al. (2016)	Source apportionment of surface ozone in the Yangtze River Delta, China in the summer of 2013	2016	CAMx	TAG	Yangtze River Delta, China	2013 (1 month)	4 km	O_3	Assess the causes of air pollution levels
Karamchandani et al. (2016)	Source-sector contributions to European ozone and fine PM in 2010 using AQMEII modeling data	2016	CAMx	TAG	16 cities in Europe	2010 (all year)	23 km	PM2.5; O_3	Assess the causes of air pollution levels
Stroud et al. (2016)	Toxic volatile organic air pollutants across Canada: multi-year concentration trends, regional air quality modelling and source apportionment	2016	AURAMS	OTH	North America	2006 (8 months)	22.5 km	VOCs	Others
Minoura et al. (2016)	Vertical circulation of atmospheric pollutants near mountains during a Southern California ozone episode	2016	CMAQ	BFM	California, USA	2005 (4 days)	4 km	O_3	Others
Dolwick et al. (2015)	Comparison of background ozone estimates over the western United States based on two separate model methodologies	2015	CMAQ; CAMx	BFM	North America	2007 (7 months)	12 km	O_3	Others
Morino et al. (2015)	Diurnal variations of fossil and nonfossil carbonaceous aerosols in Beijing	2015	CMAQ	TAG	Beijing, China	2010 (1 month)	4 km	Elemental and Organic Carbon	Others
Baker et al. (2015)	Photochemical grid model estimates of lateral boundary contributions to ozone and particulate matter across the continental United States	2015	CAMx	TAG	North America	2011 (all year)	12 km	O_3; PM2.5	Quantify transboundary transport

Progiou and Ziomas (2015)	Predicting annual average particulate concentration in urban areas	2015	CAMx	TAG	Athens, Greece	2002 (4 months)	5 km	PM	Others
Li et al. (2015a)	Source apportionment of fine particles and its chemical components over the Yangtze River Delta, China during a heavy haze pollution episode	2015	CAMx	TAG	Yangtze River Delta, China	2013 (1 month)	3 km	PM2.5	Assess the causes of air pollution levels
Wang et al. (2015)	Source apportionment of PM2.5 in top polluted cities in Hebei, China using the CMAQ model	2015	CMAQ	BFM	Hebei, China	2013 (2 months)	12 km	PM2.5	Assess the causes of air pollution levels
Li et al. (2015b)	Source contributions of urban PM2.5 in the Beijing-Tianjin-Hebei region: Changes between 2006 and 2013 and relative impacts of emissions and meteorology	2015	CAMx	TAG	Beijing–Tianjin–Hebei, China	2006; 2013 (all years)	36 km	PM2.5	Assess the causes of air pollution levels
Shahid et al. (2015)	Source Sector Contributions to Aerosol Levels in Pakistan	2015	WRF-Chem	BFM	Pakistan	2010 (4 months)	30 km	Sulphate; Nitrate; Ammonium; Black and Organic Carbon	Assess the causes of air pollution levels
Martins et al. (2015)	The role of ammonia on particulate matter pollution over Portugal	2015	CAMx	TAG	Portugal	2011 (18 days)	9 km	PM2.5	Others
Bove et al. (2014)	An integrated PM2.5 source apportionment study: Positive Matrix Factorisation vs. the chemical transport model CAMx	2014	CAMx	TAG	Genoa, Italy	2011 (4 months)	1.1 km	PM10; PM2.5	Others
Borge et al. (2014)	Emission inventories and modeling requirements for the development of air quality plans. Application to Madrid (Spain)	2014	CMAQ	BFM	Madrid, Spain	2007; 2014 (all years)	1 km	NO_2	Support the design of air quality plans
Lefohn et al. (2014)	Estimates of background surface ozone concentrations in the United States based on model-derived source apportionment	2014	CAMx	TAG	USA	2006 (all year)	12 km	O_3	Others

Lee et al. (2014)	Evaluation of concentrations and source contribution of PM10 and SO2 emitted from industrial complexes in Ulsan, Korea: Interfacing of the WRF-CALPUFF modeling tools	2014	CALPUFF	OTH	Ulsan, Korea	2012 (all year)	3 km	PM10; SO_2	Assess the causes of air pollution levels
Kota et al. (2014)	Evaluation of on-road vehicle CO and NOx national emission inventories using an urban-scale source-oriented air quality model	2014	CMAQ	TAG	Southeast Texas, USA	2006 (19 days)	4 km	CO; NOx	Others
Zhang et al. (2014b)	Impacts of updated emission inventories on source apportionment of fine particle and ozone over the southeastern U.S.	2014	CMAQ	BFM	Southeastern USA	2002 (2 months)	12 km	PM2.5; O_3	Assess the causes of air pollution levels
Ying et al. (2014)	Local and inter-regional contributions to PM2.5 nitrate and sulfate in China	2014	CMAQ	BFM	China	2009 (2 months)	36 km	PM2.5; Nitrate; Sulphate	Quantify the contribution of different areas within a country/region
Zhang et al. (2014a)	Source apportionment of sulfate and nitrate particulate matter in the Eastern United States and effectiveness of emission control programs	2014	CMAQ	TAG	Eastern USA	2000 (2 months); 2006 (2 months)	4 km	PM2.5	Evaluate the impact of abatement measures
Wang et al. (2014a)	Source contributions to primary and secondary inorganic particulate matter during a severe wintertime PM2.5 pollution episode in Xi'an, China	2014	CMAQ	TAG	Xi'an, China	2013 (1 month)	4 km	PM2.5	Assess the causes of air pollution levels
Wang et al. (2014b)	The 2013 severe haze over southern Hebei, China: Model evaluation, source apportionment, and policy implications	2014	CMAQ	BFM	Hebei, China	2013 (2 months)	12 km	PM2.5	Assess the causes of air pollution levels
Cheng et al. (2013b)	A new monitoring-simulation-source apportionment approach for investigating the vehicular emission contribution to the PM2.5 pollution in Beijing, China	2013	CMAQ	BFM	Beijing, China	2011 (4 months)	4 km	POA; Elemental Carbon; SO_2; NOX; NH_3; PM2.5	Others

Wu et al. (2013)	A study of control policy in the Pearl River Delta region by using the particulate matter source apportionment method	2013	CAMx	TAG	Pearl River Delta, China	2011 (1 month); 2012 (1 month)	3 km	PM2.5	Assess the causes of air pollution levels
Ma et al. (2013)	Application of AERMOD on near future air quality simulation under the latest national emission control policy of China: A case study on an industrial city	2013	AERMOD	BFM	Xuanwei, China	2008 (4 months); 2015 (1 month)	500 m	SO_2, NOx, PM10	Evaluate the impact of abatement measures
Cheng et al. (2013a)	Application of trajectory clustering and source apportionment methods for investigating trans-boundary atmospheric PM10 pollution	2013	CAMx	TAG	Pearl River Delta, China	2006 (3 months)	9 km	PM10	Quantify transboundary transport
Farooqui et al. (2013)	Modeling analysis of the impact of anthropogenic emission sources on ozone concentration over selected urban areas in Texas	2013	CAMx	TAG	Texas, USA	2002 (5 days)	4 km	O_3	Assess the causes of air pollution levels
Zhang et al. (2013)	Source apportionment of formaldehyde during TexAQS 2006 using a source-oriented chemical transport model	2013	CMAQ	TAG	Southeast Texas, USA	2006 (16 days)	4 km	HCHO	Assess the causes of air pollution levels
Li et al. (2013)	Systematic evaluation of ozone control policies using an Ozone Source Apportionment method	2013	CAMx	TAG	Pearl River Delta, China	2006 (4 months)	3 km	O_3	Evaluate the impact of abatement measures
Huang et al. (2012b)	Assessment of PM10 emission sources for priority regulation in urban air quality management using a new coupled MM5-CAMx-PSAT modeling approach	2012	CAMx	TAG	Beijing, China	2002 (all year)	-	PM10	Others
Cho et al. (2012)	Emission sources sensitivity study for ground-level ozone and PM 2.5 due to oil sands development using air quality modelling system: Part II - Source apportionment modelling	2012	CMAQ	BFM	Alberta, Canada	2006 (all year)	4 km	O_3; PM2.5	Assess the causes of air pollution levels

Fu et al. (2012)	Sensitivity and linearity analysis of ozone in East Asia: The effects of domestic emission and intercontinental transport	2012	CMAQ	BFM	East Asia	2001 (2 months)	36 km	O_3	Assess the causes of air pollution levels
Huang et al. (2012a)	Use of a MM5-CAMx-PSAT Modeling System to Study SO_2 Source Apportionment in the Beijing Metropolitan Region	2012	CAMx	TAG	Beijing, China	2002 (1 month)	3 km	SO_2	Assess the causes of air pollution levels
Samaali et al. (2011)	Application of a tagged-species method to source apportionment of primary PM2.5 components in a regional air quality model	2011	AURAMS	TAG	North America	2002 (3 months)	42 km	PM2.5	Others
Wagstrom and Pandis (2011)	Contribution of long-range transport to local fine particulate matter concerns	2011	CAMx	TAG	Eastern USA	2001 (35 days); 2002 (34 days)	36 km	PM	Quantify transboundary transport
Zhang and Ying (2011)	Contributions of local and regional sources of NOx to ozone concentrations in Southeast Texas	2011	CMAQ	TAG	Texas, USA	2000 (13 days)	4 km	O_3	Assess the causes of air pollution levels
Xing et al. (2011)	Modeling study on the air quality impacts from emission reductions and atypical meteorological conditions during the 2008 Beijing Olympics	2011	CMAQ	OTH	Beijing, China	2007 (1 month); 2008 (month)	4 km	O_3; PM2.5	Evaluate the impact of abatement measures
Bedogni and Pirovano (2011)	Source apportionment technique: Inorganic aerosol transformation processes in the Milan area	2011	CAMx	TAG	Milan, Italy	2004 (all year)	5 km	PM10	Others
Hixson et al. (2010)	Influence of regional development policies and clean technology adoption on future air pollution exposure	2010	UCD	BFM	San Joaquin Valley, USA	2030 (22 days)	4 km	PM2.5; Elemental and Organic Carbon; Nitrate	Evaluate the impact of abatement measures
Zhang and Ying (2010)	Source apportionment of airborne particulate matter in Southeast Texas using a source-oriented 3D air quality model	2010	UCD/CIT	OTH	Eastern Texas, USA	2000 (13 days)	4 km	PM2.5; Elemental and Organic Carbon	Others

CHAPTER 3

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3. SOURCE APPORTIONMENT OF AIR POLLUTION IN EUROPEAN URBAN AREAS: LESSONS FROM THE CLAIRCITY PROJECT

Abstract

Air pollution has become a major threat to human health in the last decades, with an increase of acute air pollution episodes in many cities worldwide. Source apportionment modelling provides valuable information on the contribution from different emission source sectors and source regions to distinct air pollutants concentrations. In this study, the CAMx model, with its PSAT tool, was applied to quantify the contribution of multiple source areas, categories and pollutant types to ambient air pollution, namely to PM and NO_2 concentrations, over six European urban areas: Bristol (United Kingdom), Amsterdam (The Netherlands), Ljubljana (Slovenia), Liguria Region (Italy), Sosnowiec (Poland) and Aveiro Region (Portugal). Results indicate overall higher annual NO_2, and PM concentrations located in the urban centres of the case studies. A comparison between the different areas showed that Liguria is the region with highest NO_2 annual mean concentrations, while Ljubljana, Liguria Region and Sosnowiec are the case studies with the highest PM annual mean concentrations. The annual average contributions denote a major influence from road transport to NO_2 concentrations, with up to 50%, except in Aveiro region, where road transport presents a lower contribution to NO_2 concentrations, and the greatest contributor is the industrial combustion and processes sector with 45%. These results indicate a negligible contribution of the transboundary transport to NO_2 concentrations, highlighting the relevance of local sources, while for PM concentrations the transboundary transport is the major contributor. The results highlight the relevance of long-range transport of PM across Europe. The transboundary transport reduces its importance during winter, when residential and commercial combustion increases its contribution. In the case of the Aveiro region, the industrial combustion and processes sector also plays an important contribution to PM concentrations.

Keywords: air pollution, air quality modelling, source apportionment, urban areas

3.1. INTRODUCTION

Air pollution has become a growing concern in the past few decades and it is expected that this issue will continue for the next few decades (Von Schneidemesser et al. 2015). Many urban areas, particularly densely populated cities, are facing air pollution problems, and it is estimated that 91% of the world's population lives in places where air quality exceeds World Health Organization guidelines (WHO 2016). Developing and low-income countries are those that face the greatest burden; however, air quality is still a problem in most developed countries as well (Pepe et al. 2019). Citizens' exposure to air pollutants like particulate matter (PM) and nitrogen dioxide (NO_2) has been threatening human health, increasing mortality and morbidity. In 2019, the estimated impacts attributable to long-term exposure to PM and NO_2 concentrations in the European Union population were around 307000 and 40400 premature deaths, respectively (EEA 2021).

To reduce the effects of air pollution, it is important to define strategies for air quality improvement. Although air quality improvement strategies, at both European (e.g. Amann et al. 2011; Miranda et al. 2015), regional (e.g. D'Elia et al. 2009; Borrego et al. 2016) and urban (e.g. Giannouli et al. 2011; Duque et al. 2016) scales, have been effective in reducing air pollution over the years, there are still problems localised in specific cities/regions (Thunis et al. 2019). Air pollution hotspots remain in the Po-valley region and Eastern Europe for PM and in most European big cities for NO_2 (EEA 2019a). One of the key issues is to understand the origin of the pollution (Thunis et al. 2019).

For that, a diagnosis of current air pollution and of the main source contributions must be made. Source apportionment (SA) tools can provide valuable information on quantifying the contribution of different source categories (e.g.: transport, domestic heating, industrial activities) and source regions (e.g.: local, urban, metropolitan areas, counties) to several pollutant concentrations. SA approaches have been widely used as part of air pollution reduction strategies for PM (e.g. Timmermans et al. 2017; Qiao et al. 2018), O_3 (e.g. Borrego et al. 2016) and NO_2 (Borge et al. 2014).

SA can be performed either through the application of Receptor Models (RM) or Chemical Transport Models (CTM). RM are based on the analysis of direct observations of air quality monitoring stations, relying on three main statistical techniques: principal component analysis (PCA), chemical mass balance (CMB), and positive matrix factorization (PMF) (Belis et al. 2014; Hopke 2016). CTM are numerical models able to simulate air pollutant concentrations, like CAMx (ENVIRON 2016), CMAQ (Wyat Appel et al. 2018) and LOTOS-EUROS (Manders-Groot et al. 2016) that include specific tools for SA analysis. Source apportionment with RM is generally limited to pollutants involved in linear processes. Because these models cannot distinguish the spatial origin of pollution, they are limited to sectorial apportionments. While CTMs can be used for both spatial and sectorial apportionments and they manage non-linear species, use of the results depends on the method used. Source apportionment based on brute force directly reflects the impact of emission reductions and are therefore useful to support air quality planning, but its application is limited in terms of emission reduction strength, hence the need to carefully assess the range of applicability. On the other hand, while tagging algorithms are suited to identify sources, they are not to quantify their impact on pollution, unless pollutants are involved in linear processes (Thunis et al. 2019). Although they are widely used to conduct SA analysis, both RM and CTM have some limitations. RM are more suitable for PM while other pollutants like NO_2 would be less straightforward (Belis et al. 2013). RM also are site-dependent and then SA results are only valid close to the air quality station considered. Thus, to represent larger areas, it is necessary to carry out additional measurement campaigns. According to Belis et al. (2013, 2015), RM also have limited capabilities to trace the origin of secondary PM whose precursors can be emitted from a varied range of sources. On the other hand, CTM can be used for any area of interest, but their reliability strongly depends on the accuracy, and spatial and temporal resolution of input data, such as emission inventories (Guevara et al. 2014) and meteorological datasets (Bessagnet et al. 2016). Additionally, secondary organic aerosols are generally hardly simulated and often underestimated (Bergström et al. 2012; Meroni et al. 2017).

This study focuses on the quantification of the contribution of distinct source categories to PM (PM10 and PM2.5) and NO_2 concentrations, based on the application of the SA tool available in CAMx model, over six urban areas, which are distinguished by their location in Europe, population density, environmental context, namely in terms of air pollution, and economic and social backgrounds, among others. Bristol (United Kingdom), Amsterdam (The Netherlands), Ljubljana (Slovenia), Liguria Region (Italy), Sosnowiec (Poland) and Aveiro Region (Portugal), were the six urban areas chosen, as they are the case studies of the ClairCity project, in which this study is integrated (www.claircity.eu). The outputs of this work provided technology-independent activity data on a sector level that acknowledges the heterogenic nature of each case study.

ClairCity intended to apportion air pollution emissions and concentrations, carbon footprints and health outcomes by city citizens' behaviour and day-to-day activities in order to make these challenges relevant to how people chose to live, behave and interact within their city environment. In this context, this study presents the first level assessment through a CTM-based SA study identifying the main contributions of multiple source areas, categories and pollutant types to ambient air pollution, namely to PM10, PM2.5 and NO_2 concentrations in the six ClairCity cities/ regions.

The main objective of this study is to quantify the contributions to PM10, PM2.5 and NO_2 from different sectors and regions, under distinct case studies. For that, this work presents a diagnosis of the current air quality, and investigates the main source activities contributions, for the six cities/regions under analysis. The multi-urban area feature of this work, together with the contribution from source categories and regions in the SA analysis of PM and NO_2, make its novelty and relevance in the context of CTM-based SA studies. Other SA studies have already been done, however some research gaps remain unfulfilled. For example, Thunis et al. (2021) published a PM2.5 urban atlas with a sectorial and spatial source apportionment for 150 European cities, Timmermans et al. (2017) applied LOTOS-EUROS model for the cities of Beijing and Shanghai, Qiao et al. (2018) performed CMAQ simulations for 25 Chinese provincial capitals and municipalities and Li et al. (2015) applied CAMx-PSAT for Beijing–Tianjin–Hebei region, however all studies were focused only on PM2.5. Other studies like Kim et al. (2017), for Seoul, and Wang et al. (2017), for Beijing, applied CAMx-PSAT, but only for high pollution episodes and focusing only on source regions without investigating source sectors contributions.

The present work is organized as follows: Section 3.2 provides a short characterization of the six case studies; Section 3.3 describes the air quality modelling system setup; the air quality modelling evaluation is presented in Section 3.4; Section 3.5 analyses the main modelling results, as well as the main limitations of this work; conclusions are drawn in Section 3.6.

3.2. CASE STUDIES CHARACTERIZATION

Using six cities/regions (Bristol (BRS), UK; Amsterdam (AMS), NL; Ljubljana (LJB), SI; Liguria (LIG), IT; Sosnowiec (SOS), PO; and Aveiro (CIRA), PT) this study captures the variability across EU cities considering distinct indicators, namely: (i) spatial scales and population demographics; (ii) geographic distribution; (iii) air quality and carbon sources, emissions and concentrations; (iv) social, economic and health-related challenges; and (v) management capacities and capabilities.

A summary description of these cities/regions characteristics is provided in Table 3.1. The full description of these cities/regions and how they fulfil the above criteria can be found in supplementary material (section 3.8).

Table 3.1. *Cases studies' characteristics.*

Case study name & abbreviation	***Population [habitants]***	***Area [Km²]***	***Pollutants of Concern***	***Case study specificities***	***References***
Bristol, BRS	450 000	140	NO_2	Locate in North-western Europe with a maritime climate Big city with a lot of commuting Port and airport facilities	(Slingerland et al. 2018, 2020a, b; Rodrigues et al. 2021)
Amsterdam, AMS	835 000	220	PM; NO_2	Locate in Western Europe with a maritime climate Important centre of enterprise and industry Airport facilities Largest petrol and second largest coal port of Europe	(Slingerland et al. 2017, 2020b, c; Rodrigues et al. 2021)
Ljubljana, LJB	289 000	275	PM	Locate in Central Europe with a continental climate Situated in the Sava River basin Big city with industry, services, and tourism	(Slingerland et al. 2018, 2020b, c; Rodrigues et al. 2021)
Liguria Region, LIG	1 565 000	5 420	NO_2	Locate in Southern Europe with a Mediterranean climate Located between sea and steep hills Large industrial centre in the coastal area Important port facilities	
Sosnowiec, SOS	200 000	90	PM; NO_2	Locate in Central Europe with a continental climate Medium sized city Strong industrial sector and coal mines	
Aveiro Region, CIRA	370 000	1 690	PM	Locate in South-western Europe with a Mediterranean climate Coastal region with six thousand hectares of estuary Geographic and economic diversity (from coastal towns to rural inland areas) Several chemical and manufacturing companies	

3.3. MODELLING APPLICATION

The CAMx - Comprehensive Air Quality Model with Extensions, version 6.3, (ENVIRON 2016), forced by the WRF - Weather Research and Forecasting, version 3.7, (Skamarock et al. 2008) meteorological fields, was applied over the 6 cities/regions, for the year of 2010, that represents typical weather conditions for the various case studies (NOAA 2011). The CAMx Particulate Source

Apportionment Technology (PSAT) (Yarwood et al. 2007) was applied to quantify the contributions of multiple source areas, categories, and pollutant types to ambient pollution, over each case study.

Both models were applied using a two-nests approach based on a European domain with 0.25 degrees' horizontal resolution and 6 domains of interest centred in each case study, with 0.05 degrees' horizontal resolution (see Figure 3.1 and Table 3.2 of supplementary material (section 3.8)). For each case study, to better consider all areas that may influence the air quality conditions of the city/region under analysis, the size of the domains was selected together with local partners.

Meteorological inputs to the chemical simulations were driven by the meteorological model WRF, forced by ERA-Interim reanalysis data (Berrisford et al. 2011) from European Centre for Medium Range Weather Forecast at 6 hours and 0.75 degrees temporal and spatial resolution respectively. The following set of physical parameterizations, applied to all cities/regions, and selected based on previous studies (e.g. Kong et al. 2015; Coelho et al. 2021; Ascenso et al. 2021), were used in the WRF model: WRF Single-moment 5-class Microphysical Scheme (Hong et al. 2004); Dudhia Shortwave radiation scheme (Dudhia 1989); RRTM (Rapid RadiativeTransfer Model) longwave radiation model (Mlawer et al. 1997); MM5 similarity surface layer scheme (Zhang and Anthes 1982); Unified Noah Land Surface Model (Tewari et al. 2004); Yonsei University Planetary Boundary Layer scheme (Hong et al. 2006) and Grell 3D Ensemble Scheme for cumulus parametrization (Grell and Dévényi 2002).

CAMx initial and boundary conditions for the first domain were provided by the global chemical model MOZART (Emmons et al. 2010) with a temporal and spatial resolution of 6 hours and 1.9 by 2.5 degrees, respectively. The ozone column data was obtained from the Total Ozone Mapping Spectrometer dataset (Mcpeters et al. 1998). Anthropogenic emissions for both domains were taken from the TNO-MACC_II European emission inventory (Kuenen et al. 2014) available at a resolution of 0.125 by 0.0625 degrees, and were speciated into the CB6 chemical mechanism species considered in the CAMx simulation (Yarwood et al. 2010). As this inventory comprises annual total emissions by area for the main activity sources, monthly, daily and hourly coefficients provided by the TNO database (Van Der Gon et al. 2011) were used.

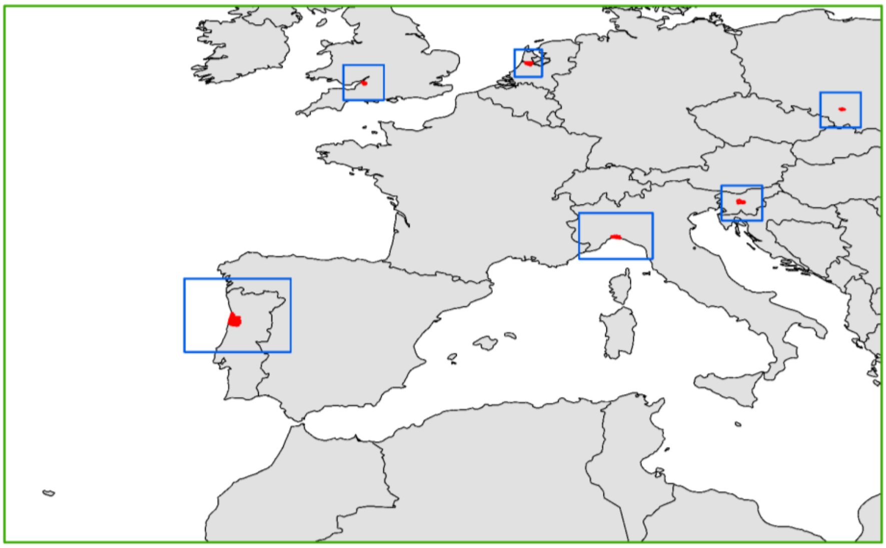

Figure 3.1. *Map of the WRF-CAMx European domain (within the green rectangle) and the 6 domains of interest (within the blue rectangles) for each case study location (in red).*

The PSAT application requires the definition of source groups to be tracked and thus the input of extra emission files for each of the groups to be considered. Based on the national emission inventories of each case study (Dębski et al. 2018; Logar et al. 2018; Pereira et al. 2018; Taurino et al. 2018; Wakeling et al. 2018; Wever et al. 2018) and on the emission sources, the main sectors contributing to PM and NO_2 emissions in the year under study, that are common to all case studies, are: (i) residential and commercial combustion; (ii) road transport; and (iii) industrial combustion and processes. All the remaining sectors, which were grouped and treated as one, consider energy industries, extraction and distribution of fossil fuels, product use, non-road transport and other mobile sources, waste treatment and agriculture. In this sense, emissions were split into these activity sectors in order to evaluate the individual contribution of each source sector to the air quality in the selected urban areas through the PSAT application, defined as receptor areas. An overview of the annual emissions structure of each case study, for the main pollutants (NOx, PM, SO_2, NH_3 and non-methane VOC), can be seen in Figure 3.5 of supplementary material (section 3.8). The contribution of the three main sectors to the urban area emissions are very different among the case studies. The industrial combustion and processes have a great impact in CIRA and the residential and commercial combustion in LJB, while in the other cases, the three main sectors have relevant contributions to the emissions of all pollutants. As air pollution has become a growing concern in urban areas, where most people live and work and therefore are more exposed to atmospheric pollution, the source apportionment analysis focuses on grid cells with the highest population density, those corresponding to consolidated urban areas. So, for each case study, the receptor area was defined as a set of cells, with similar results, in the 0.05º resolution domain, which cover the respective urban area. The number of selected cells, depending on the size of the urban area, was 3 for LIG, SOS and CIRA, and 4 for AMS, BRS and LJB.

3.4. AIR QUALITY MODELLING PERFORMANCE

The CAMx performance was evaluated through the application of conditional quantiles, following the approach of Carslaw and Ropkins (2012), and performance statistics, based on the monitored PM10 and NO_2 concentrations during 2010, from the Air Quality e-Reporting database (EEA 2019b). PM10 and NO_2 datasets were composed by 9 and 15 urban/suburban background air quality monitoring stations, with more than 75% of data collection capture. The performance of CAMx for PM2.5 was not evaluated due to the lack of observed data.

Figure 3.2 shows the conditional quantile plots comparing CAMx output and AQ e-Reporting data for the hourly average concentrations of NO_2 and the daily average concentrations of PM10, for the case studies ensemble. The conditional quantile plot splits the predicted values into evenly spaced bins and calculates median values of the predictions and corresponding observations (red line), as well as 25/75th and 10/90th percentiles (yellow and orange shadings, respectively). Additionally, the histograms show the bins' counts for observed values (blue-contoured) and the predicted values (grey). A perfect CAMx performance would lie on the blue line and have a percentile spread as small as possible (Wilks 2006).

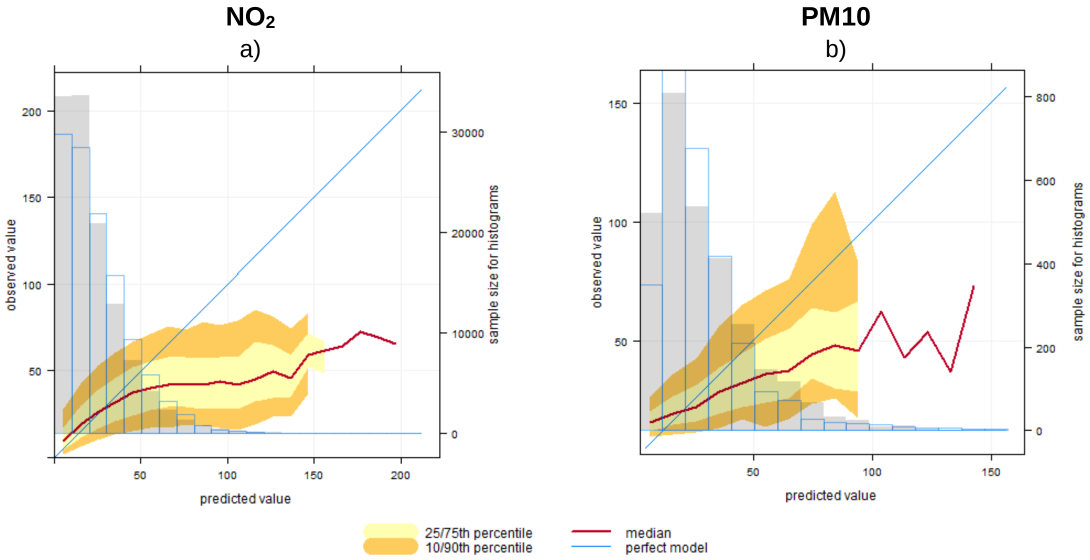

Figure 3.2. *Comparison between CAMx output bins (predicted value, blue bars) and Air Quality e-Reporting bins (observed value, grey bars) through the application of conditional quantiles, and median values of the predictions and corresponding observations (red line), as well as 25/75th and 10/90th percentiles (yellow and orange shadings, respectively), for (a) hourly average concentrations of NO_2 and (b) daily average concentrations of PM10, in 2010, for the case studies ensemble.*

From Figure 3.2, the analysis of the histograms shows that about 80% of the observations are recorded for bins between 0 and 40 µg.m^{-3}, for both pollutants. It is for this range of values that CAMx presents its best performance. The poorest CAMx performance is in reproduction of peak concentrations, especially for PM10, and it can be clearly highlighted by the increasing percentiles spreads, that become larger in the upper tail of the distribution. In general, CAMx tends to underestimate NO_2 concentrations, while PM10 concentrations tend to be overestimated.

To analyse the quantitative CAMx performance the following statistical parameters were used: (i) correlation coefficient; (ii) mean bias; (iii) mean error; (iv) index of agreement; (v) root mean square error (RMSE); and (vi) modelling quality indicator (MQI), considering a deviation between modelled and measured concentrations as twice the measurement uncertainty, as defined by Janssen and Thunis (2022). Tables 3.3 and 3.4 of supplementary material (section 3.8) summarize the values of the CAMx performance statistics, for NO_2 and PM10, respectively, for the case studies ensemble and for each one separately.

For the case studies ensemble, mean bias for NO_2 and PM10 is -2.55 µg.m^{-3} and 1.73 µg.m^{-3}, respectively, showing the general underestimation of NO_2 and the overestimation of PM10, already identified in the conditional quantile analysis. For both NO_2 and PM10, mean error and RMSE are up to 13.91 µg.m^{-3} and 19.89 µg.m^{-3}, respectively. Correlation and index of agreement for both NO_2 and PM10 concentrations cover a range from 0.49 to 0.56, pointing out some troubles of CAMx in reproducing the patterns of the observed concentrations. When each case study is analysed separately, the statistical parameters vary from those obtained for the set of case studies. In general, for NO_2, AMS is the one with the highest agreement between observed and predicted values, while for PM10 the best performance is obtained for BRS. On the other hand, the poorest CAMx performance was obtained for LIG and LJB, for NO_2 and PM10, respectively. For the modelling quality indicator, except for PM10 for LJB, values below 1 are obtained for all case studies and pollutants. According to the FAIRMODE guidelines (Janssen and Thunis 2022), 90th percentile MQI values below 1 indicate that the modelling quality objective, defined as the minimum level of quality to be achieved by a model for policy use, is fulfilled.

CAMx performance can be attributed to different factors. Pepe et al. (2016) studied the origin of NOx underestimation that could be probably explained by the potential overestimation of the vertical mixing. Although the purpose of their study was focused on assessing the influence of meteorological parameters on NO_X, similar conclusions were drawn for NO_2. This must be the case of LIG and LJB, where the complexity of the terrain (LJB is situated in a basin and LIG is located between sea and steep hills) could lead to an overestimation of vertical transport, as reported by Emery et al. (2011). Baker and Bash (2012) and Itahashi et al. (2018) evaluated the wet deposition processes in both CAMx and CMAQ CTM models, concluding that this process is underestimated in the CAMx model. Also, the WRF model performance for precipitation can be an important factor in the CAMx performance for wet deposition. It should be noted that an underestimation of precipitation will result in a underestimation of wet deposition (Itahashi et al. 2018, 2020) and consequent overestimation of PM10 concentrations. As pointed out by Oikonomakis et al. (2018) the CAMx underestimation/overestimation could be also a consequence of real emissions underestimation/overestimation in the emission inventory database. Additionally, Pepe et al. (2019) show that CAMx performance improves at rural stations, when compared to urban and suburban stations, suggesting that some of the discrepancies observed in urban and suburban sites can be attributed to sources of local scale, which influence is difficult to reproduce at this spatial resolution. A similar conclusion can be made for this study. Although there are no rural stations for any of the 6 case studies, when urban and suburban stations are analysed separately, it is possible to verify a better performance of the model for suburban stations, for NO_2 concentrations. However, in some case studies, the low number of air quality monitoring stations and consequent less observed data available to compare with predicted data, can also lead to a misinterpretation of the model's performance.

3.5. AIR QUALITY AND SOURCE APPORTIONMENT ANALYSIS

The air quality characterization was based on the spatial distribution concentration maps (section 3.5.1) and on a source contribution analysis (section 3.5.2). The spatial analysis was done for the average concentrations of NO_2, PM10 and PM2.5 for the following periods: (i) annual; (ii) a typical winter month (February); and (iii) a typical summer month (August). The source contribution analysis was provided to estimate the contribution to the modelled NO_2, PM10 and PM2.5 concentrations, from transboundary transport (TBD) and from specific source groups previously defined – residential and commercial combustion (RES), industrial combustion and processes (IND), road transport (TRP) and all the remaining sources (OTH). The results were analysed in terms of the relative contribution of those groups to the NO_2, PM10 and PM2.5 concentrations simulated for each receptor area previously defined in the PSAT application.

3.5.1. Concentration fields

The NO_2 and PM10 concentration fields, obtained from the WRF-CAMx application for the three periods previously defined, is presented in Figure 3.3. PM2.5 concentration fields are not presented as they are similar to those shown for PM10, but with different magnitudes. For each case study, the results show similar spatial patterns for the different periods and air pollutants analysed. In addition, the dispersion of air pollutant patterns follows the predominant wind direction for the periods analysed.

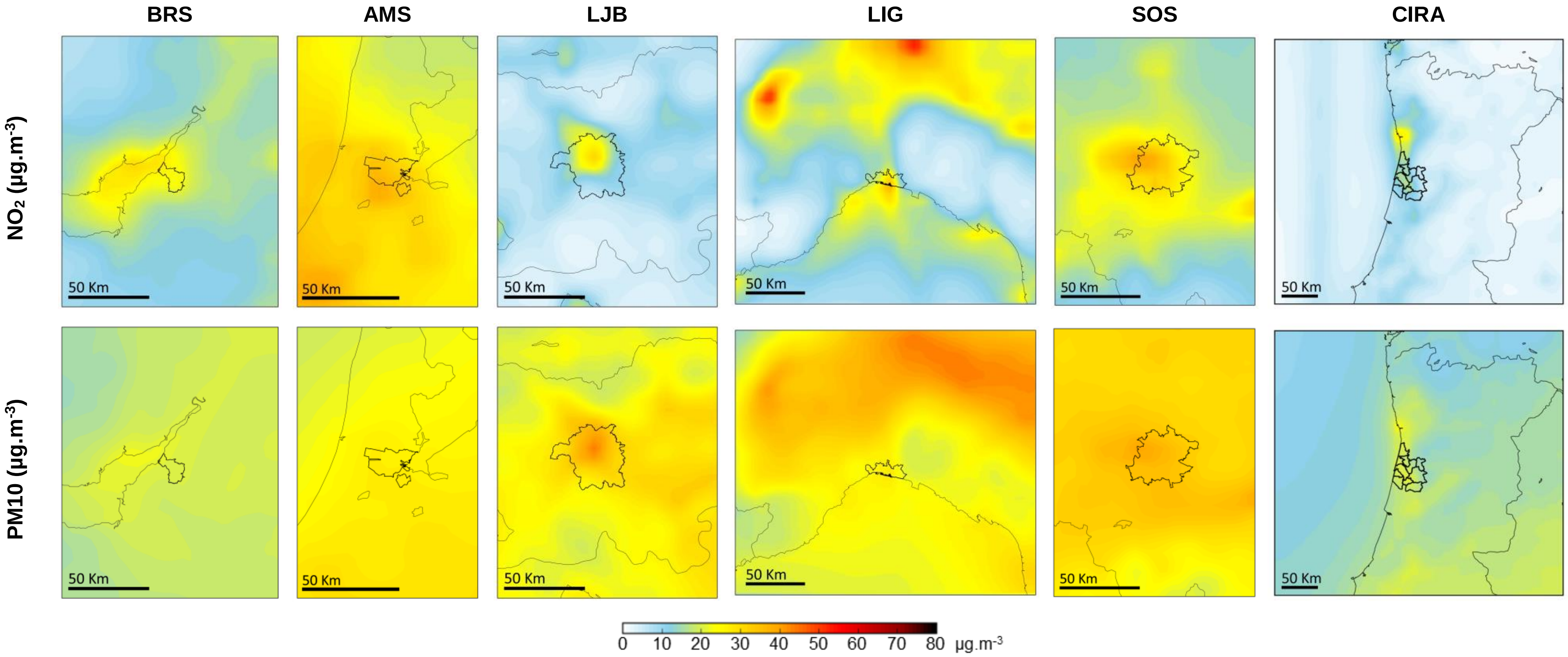

Figure 3.3. *Spatial distribution of NO_2 and PM10 annual average concentrations for each case study.*

Focusing on the analysis of annual concentration fields, the higher values of NO_2 and PM10 concentrations are found in the urban centres of the case studies. In some case studies, high values of pollutant concentrations are also observed in other urban areas, which although not belonging to the city/region under analysis, are within the defined domains, such as: (i) Cardiff, for the BRS case study; (ii) Milan and Turin, for LIG case study; and (iii) Porto, for CIRA case study. Comparing the different case studies among them, for NO_2, LIG is the one with the highest values, with maximum reaching 50 $\mu g.m^{-3}$, next to the large urban areas and the seaport. Following are the case studies of AMS and SOS, where the maximum values are around 40 $\mu g.m^{-3}$. Finally, the remaining case studies do not present values greater than 35 $\mu g.m^{-3}$. For PM10, the case studies with the highest concentrations are LJB, LIG and SOS, with maximum values around 45 $\mu g.m^{-3}$. Secondly, are AMS and CIRA case studies, with values up to 35 $\mu g.m^{-3}$ and, finally, BRS case study with maximum values close to 20 $\mu g.m^{-3}$. For PM2.5, the analysis is similar to that of PM10, but with concentration values between 5% to 15% lower than those of PM10.

Regarding the analysis of seasonal concentration fields (Figures 3.6 and 3.7 of supplementary material (section 3.8)), results show that, for all case studies, the maximum values are found in winter, while the minimum values are recorded in summer. This seasonal variability is observed for all pollutants but is most noteworthy for PM10 and PM2.5. In the results it is also possible to verify that the case studies with greater amplitude of values, both for NO_2 and for PM10 and PM2.5, are LJB, followed by SOS and LIG. AMS is the case with lower amplitude of NO_2 concentration values, ranging from 33 $\mu g.m^{-3}$ in summer to 43 $\mu g.m^{-3}$ in winter. For PM10 and PM2.5, CIRA is the case study where the amplitude between winter and summer concentrations is smaller, with differences of about 5 $\mu g.m^{-3}$, for both pollutants.

For all case studies, and for both annual and seasonal concentration fields, results show higher spatial variability for NO_2 concentrations than for PM10 and PM2.5. These results point out that NO_2 concentrations have a greater dependence on local sources, when compared with those of PM10 and PM2.5, as suggested by Eeftens et al. (2015) and Guo et al. (2019).

3.5.2. Source contribution analysis

The contribution of each source group for NO_2 and PM10 concentrations, in the receptor area of each case study, for the three periods previously defined, are analysed in Figure 3.4. PM2.5 contributions are not presented as they are similar to those shown for PM10.

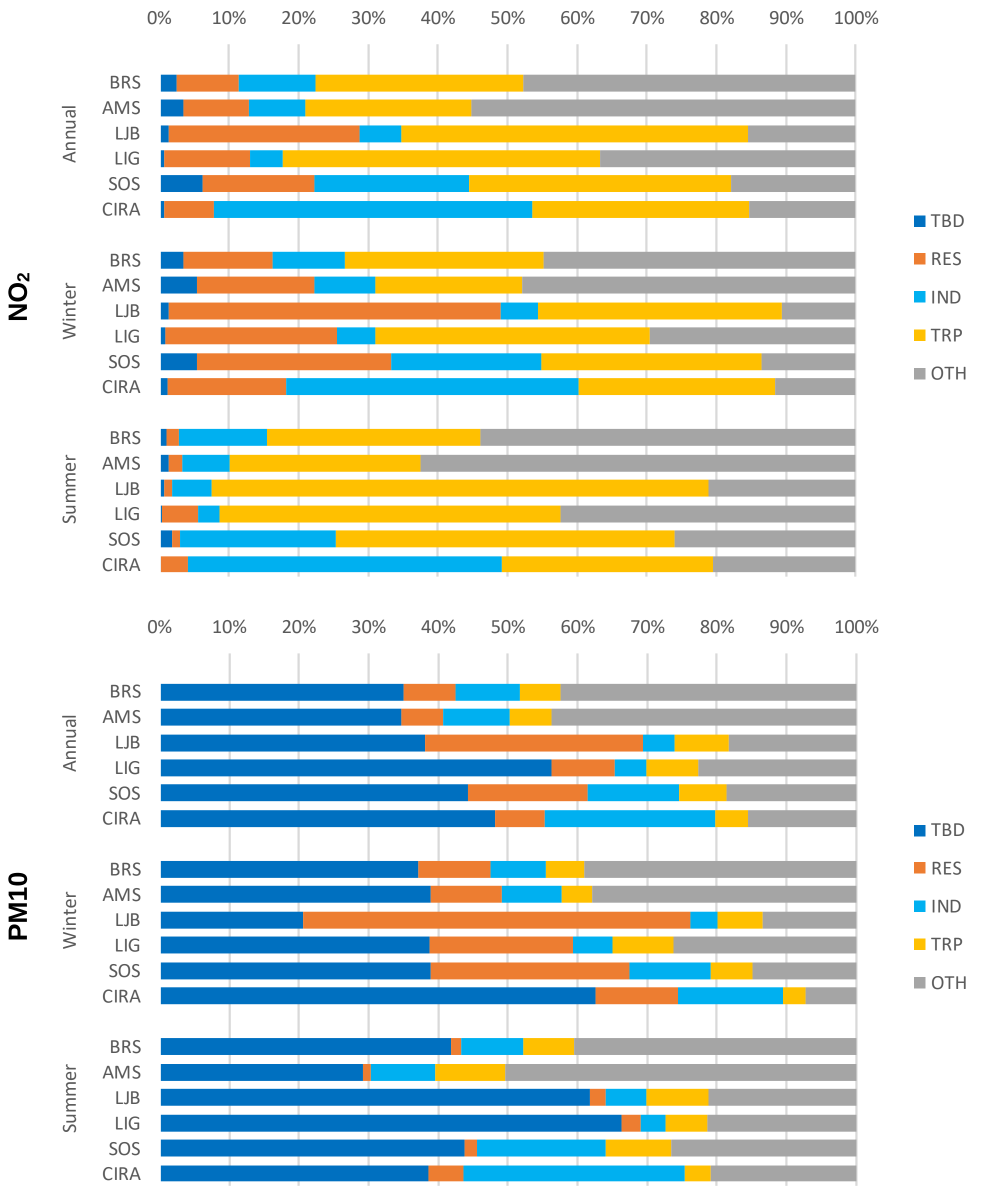

Figure 3.4. *Annual, winter and summer contributions of each source group for NO_2 and PM10 concentrations, for each case study; (TBD- transboundary transport, RES - residential and commercial combustion, IND - industrial combustion and processes, TRP - road transport and OTH - all the remaining sources).*

For NO_2, the annual average contributions of each source group reveal that the major contribution is from road transport, for almost all case studies, with values up to 50%. These results are in line with what was observed in the analysis of the annual emissions structure of each case study (see Figure 3.5 of supplementary material (section 3.8)), where, for most of the case studies, NOx emissions are mainly resulting from road transport sector. According to Degraeuwe et al. (2019), in the European Union, road transport is the largest contributor to NO_2 pollution, ahead of the energy, commercial, institutional and household sectors. Kiesewetter et al. (2014) attributes the contribution of road traffic to NO_2 concentrations to the high shares of diesel cars in European fleets, but expects a declining in this contribution if currently approved legislation is successfully implemented. Conversely, in the CIRA case study road transport presents only a contribution of 30%, and, the industrial combustion and processes represent a contribution of 45% to the NO_2 concentrations in the region, as already expected according to other studies (Figueiredo et al. 2013; Coelho et al. 2018). In the winter period, the contribution of residential and commercial combustion increases in all case studies, but with special importance for LJB, due to the widespread use of wood for domestic heating in outdated boilers and stoves (ARSO 2015; Slingerland et al. 2018).

On the other hand, during summer, the contribution of residential and commercial combustion is quite small, with industrial combustion and processes and road transport having the most relevant contributions. For the three periods under analysis, transboundary transport presents contributions up to 5%, highlighting the importance of local sources for NO_2 concentrations as suggested by Eeftens et al. (2015), Degraeuwe et al. (2019) and Guo et al. (2019). Regarding the remaining sources, for CIRA, LJB and SOS, these present a relatively low contribution, with an annual average contribution up to 18%. However, for the remaining case studies, their contribution is more important, reaching a contribution greater than 50% for AMS. In general, these values are due to emissions associated with seaport and airport activities, mainly in AMS and LIG (Rodrigues et al. 2021), but also due to electricity production plants, more relevant in BRS case study (Wakeling et al. 2018). Although these emission sources are not classified as large combustion sources in the national emission inventory and, therefore, were not previously identified as point sources, at the local level they are of great relevance for some case studies, highlighting the importance of local sources for NO_2 concentrations (Eeftens et al. 2015; Degraeuwe et al. 2019; Guo et al. 2019).

For PM10 and PM2.5, the analysis of the source's contribution is very similar, so it is reported only for PM10. For PM10, for all case studies, the major contribution is from transboundary transport, with values ranging from 35%, in AMS and BRS, to 56% in LIG. These results highlight the importance of long-range transport of PM10, as stated by Kiesewetter et al. (2015) and Garg and Sinha (2017). This transboundary transport effect increases, in some case studies, during summer, and is smaller during winter. Residential and commercial combustion also show a great contribution, especially during winter. This effect can be linked especially to residential combustion for domestic heating, since it is observed in the case studies where the lowest average temperature was obtained, e.g. in LJB with 56% and SOS with 29%. From the analysis of the annual emissions of these two case studies (see Figure 3.5 of supplementary material (section 3.8)), 41% of PM10 emissions in SOS, and 35% in LJB, result from the residential and commercial combustion sector. Additionally, LJB is in one of the European countries with the highest levels of PM10 concentrations and emissions per capita and per area due to the outdated domestic heating systems together with the unfavourable basin geography that causes unfavourable temperature inversions, particularly in winter, favouring pollutant accumulation (ARSO 2015; Slingerland et al. 2018). In turn, during summer, these values decrease to less than 5% in all case studies, with residential and commercial combustion being the one that, during this period, presents the smallest contribution. The industrial combustion and processes also make a relevant contribution, especially for CIRA, as this is a very industrialized area with industrial centres very close to the urban centre (Figueiredo et al. 2013; Coelho et al. 2018), as it can be seen in the annual emissions structure of Figure 3.5 (supplementary material, section 3.8), where this sector is responsible for 44% of PM10 emissions. Regarding road transport, contributions vary between 3% and 10%, throughout the year, demonstrating a reduced importance for PM10 concentrations, in contrast to its relevance for NO_2 concentrations. For the remaining sources, AMS and BRS are those where the highest values are obtained. When compared to the other case studies, they present higher relative contributions of PM10 emissions in agriculture (OTH10), solvent’s use (OTH6) and in seaport and airport activities (OTH8), as shown in Figure 3.5 (supplementary material, section 3.8), and in line with the findings discussed by Wakeling et al. (2018) and Wever et al. (2018).

The results above show that although some case studies face similar air quality issues, their causes can be very different and are closely linked to the specificities of each case study, such as: (i) location (e.g. LJB is located in a basin, reducing the pollutants dispersion); (ii) climate (e.g. lowest average temperature of SOS leads to an increase in residential combustion for domestic heating); or (iii) main activity sectors (seaport and airport activities in AMS and LIG; electricity production plants in BRS; chemical and manufacturing companies in CIRA). For BRS and AMS, road transport should be considered as being the most prominent contributor for air pollution. For LJB, LIG and SOS, in addition to road transport, residential and commercial combustion must also be considered. In turn, for CIRA, critical sectors are industrial combustion and processes and, once again, road transport. These results are in line with other studies. Pommier et al. (2020) and Kiesewetter et al. (2015) stated that contributions vary significantly between regions. However, in Pommier et al. (2020) it is possible to identify road traffic, industrial activities, and the burning of domestic fuel as being the most prominent contributors. However, due to the high relevance of transboundary transport in PM10 concentrations, this must also be taken into account when analysing air pollution sources. Thus, air pollution issues extend beyond the geographical and political frontiers of the city, across different cities, regions and countries. This idea is corroborated by Kiesewetter et al. (2015) who highlighted the transboundary transport of PM as an issue in many European Union Member States, and the need for a synchronized wide action.

3.5.3. Limitations

Despite its novelty and relevance in the context of CTM-based SA studies, this study has some limitations. Although PSAT analysis gives valuable insights about the sources and their relative contributions to PM10, PM2.5 and NO_2 for each case study, caution should be taken in using these results to support air quality policies. Due to the non-linearity of the atmospheric processes, the source apportionment contributions calculated with PSAT will not reflect the impact of emission changes of concentrations. To get insight on these potential impacts on concentration levels, brute force-based apportionments or dedicated CTM scenarios must be performed.

In addition, seasonal results based on two typical months (typical winter month - February; typical summer month - August) are indicative of seasonal patterns and contributions, but they should not be linearly extrapolated to other specific months, since they do not fully describe the role of transboundary transport and the defined source groups in each case study.

As for all CTM-based studies, uncertainties in meteorology, emission inventories, and formulations and parameterizations, as well as the spatial resolution used can affect the model performance and also the accuracy of source apportionment results. Thus, such model performance issues should be considered in the interpretation of results. These issues can be of relevance in assessing compliance with air quality legislation. For NO_2, the identified underestimation can lead to erroneous conclusions that no air quality management measures are required. On the other hand, opposite conclusions can be made for PM10, as it is overestimated.

Despite these limitations, the results presented in this work provide valuable information regarding the sources and their relative contributions to PM10, PM2.5 and NO_2 in the six cities/regions under analysis.

3.6. CONCLUSIONS

The modelling system based on the combination of the WRF meteorological model and the CAMx chemical transport model with the PSAT tool was applied over six European urban areas, which are distinguished by their location, population density and air pollution context, to quantify the contribution of multiple source categories to PM and NO_2 concentrations.

CAMx's performance was first assessed by comparing its results with the AQ e-Reporting data. An underestimation of the NO_2 concentrations and an overestimation of the PM concentrations were verified. It is of note that the CAMx performance is variable city by city, with a high dependence on meteorological and air quality measurements availability. Such result states that CAMx, with the PSAT tool, can represent a powerful tool to provide useful information about pollutants concentrations as well as estimate its source contribution of the different emission categories.

Results indicate, in general, the higher annual NO_2 and PM concentrations in the urban centres of the case studies. A comparison between the different cities shows that LIG is the region with highest NO_2 annual mean concentrations, while LJB, LIG and SOS are the case studies with the highest PM annual mean concentrations. For almost all case studies, the main activity source is the road transport, followed by residential and commercial combustion or industrial combustion and processes. For NO_2, results reveal a negligible contribution of the transboundary transport, highlighting the relevance of local sources, while, for PM, transboundary transport appears as the biggest contributor for all six case studies.

The air quality and source apportionment assessment provided in this paper should be the first step for a detailed diagnosis of the status of the air quality. The comprehensive methodology of this work made it suitable to be applied for any air quality assessment that may be performed for other urban areas. Also, the main findings of this paper highlight two major challenges that must be considered when working with different case studies: (i) similar air quality issues can have different causes for each case study, as they depend on emission sources and specific characteristics of each location; and (ii) the need for establishing collaborative actions to reduce air pollution due to long-range and transboundary air pollution transport.

As future work, this modelling system should be applied with a higher spatial resolution, allowing a more accurate identification of the air pollution hotspots and their sources contributions. Emission reduction scenarios must also be tested. According to Thunis et al. (2020), due to the complexity of atmospheric processes, the reduction of emissions may not show a linear reduction in the concentrations of pollutants. Under the framework of the ClairCity project, the next step will be the apportionment of air pollution emissions and concentrations by citizens' behaviour and day-to-day activities in order to make these challenges relevant to how people chose to live, behave and interact within their city environment. Consequently, this will empower them to better understand their contribution to air pollution, accelerating the transition to clean air future.

3.7. REFERENCES

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3.8. SUPPLEMENTARY MATERIAL

Case studies characterization

Bristol is a major city in the Southwest of England with a population of around 450 000. It is the main hub of the economy of the southwest England. Working in the city and some key suburbs and commuting to these from the wider region is common. Bristol has a port and airport, but these are not major influences on emissions in the city. Situated in north-western Europe, Bristol has a maritime climate and predominantly south-western winds. Air quality in Bristol is mainly affected by regional background concentrations, by urban background concentrations and of local concentrations at certain hotspots in the city. The key pollutant of interest is NO_2.

Amsterdam is the capital and the largest city of The Netherlands with around 835 000 inhabitants, and one of the main urban and economic centres of the country. The Amsterdam metropolitan area is an important centre of international enterprise and industry. Amsterdam harbour is the largest petrol import and storage port and the second largest coal port of Europe. Amsterdam economy is also highly dependent on tourism. Situated in north-western Europe and near to the North Sea, Amsterdam has a maritime climate and predominantly south-western sea winds. Amsterdam city policy in the field of air quality is successful in the sense that in most places, it manages to meet the European limit values.

Ljubljana is the capital and the largest city of Slovenia. It is situated on an alluvial plain in central Slovenia. It lies at the crossroad of four European regions: The Alps, Central Europe, South-Eastern Europe and the Mediterranean. With around 289 000 inhabitants and its central geographic location within Slovenia, it is considered the economic hub of the country, with industry, services, a university, and tourism contributing to its economic activity. Many commuters also have an important contribution to air pollutant emissions in the city. Ljubljana’s climate is south Alpine with a moderate continental influence. The city is situated in a basin between the Ljubljana Plain and the Ljubljana Marshes, which has a negative impact on the city’s air quality.

The Liguria Region, in Italy, is located in between the sea and the steep hills along the coast of the Gulf of Genoa. The territory is overall hilly, with an extensive forest area. The population density is 289 inhabitants per km^2, above the Italian average. The population of the region is concentrated on the coast, mainly in Genoa, along with its industry and large harbour, both of which are unusually close to urban dwellings. The sea and harbour play a critical role in Liguria’s economy. Today Genoa ranks as the first port in Italy in terms of total movement of goods. The Liguria region is characterised by a Mediterranean climate and a mountainous geography. Predominant wind direction helps to reduce air pollution, but the limited possibilities for land-use due to the geographical situation lead to a concentration of port, industry, traffic, and housing in the coastal area, thus contributing to air quality problems. Exceedances of NOx emissions are still problematic.

Sosnowiec is a medium sized city located in the Upper-Silesia region, in southern Poland. Its population is around 200 000 people. Sosnowiec retains a strong industrial sector, with production continuing in some specific industries, especially in automotive parts. Coal mines still play an important role in the regional economy. Sosnowiec is situated in a mainly flat area and has a continental climate, with cold winters and warm summers. Winds in Sosnowiec are not particularly strong, blowing primarily from West. The cold winters, with consequent high heating demand, combined with the low winds can contribute to pollution remaining over the city, leading to high air pollution levels on cold winter days. Furthermore, the city is experiencing urban congestion problems typical of any metropolitan zone.

The Aveiro Region is in the Centre Region of Portugal close to the Atlantic Ocean and consists of eleven relatively small municipalities with a great diversity of geography and economy. Every municipality is influenced by the presence of Ria de Aveiro, with more than six thousand hectares of the estuary permanently covered with water. Aveiro Region has around 370 000 inhabitants with the population density varying substantially in each municipality. Rural municipalities are typically situated inland, while industrial municipalities are along the coast, closer to the port, one of the most important ports in Portugal. The Aveiro Region has a high economic and social importance in Portugal due to various chemical and manufacturing companies active in the region. Air quality in the Aveiro region is relatively good, due to an open landscape in a temperate maritime climate. However, NO_2 and PM exceedances do occur occasionally.

- **Modelling application**

Table 3.2. *WRF-CAMx domains' coordinates an dimensions.*

	European Domain			*Case Study Domain*		
	Lon [º]	***Lat [º]***	***Number of cells***	***Lon [º]***	***Lat [º]***	***Number of cells***
BRS	-19.00 to 21.00	30.50 to 55.50	160 x 98	-3.50 to -1.75	50.75 to 52.25	35 x 30
AMS				4.25 to 5.50	51.75 to 53.00	25 x 25
LJB				13.75 to 15.50	45.25 to 46.75	35 x 30
LIG				7.25 to 10.50	43.50 to 45.50	65 x 40
SOS				18.25 to 20.00	49.50 to 51.00	35 x 30
CIRA				-10.75 to -6.00	39.25 to 42.50	95 x 65

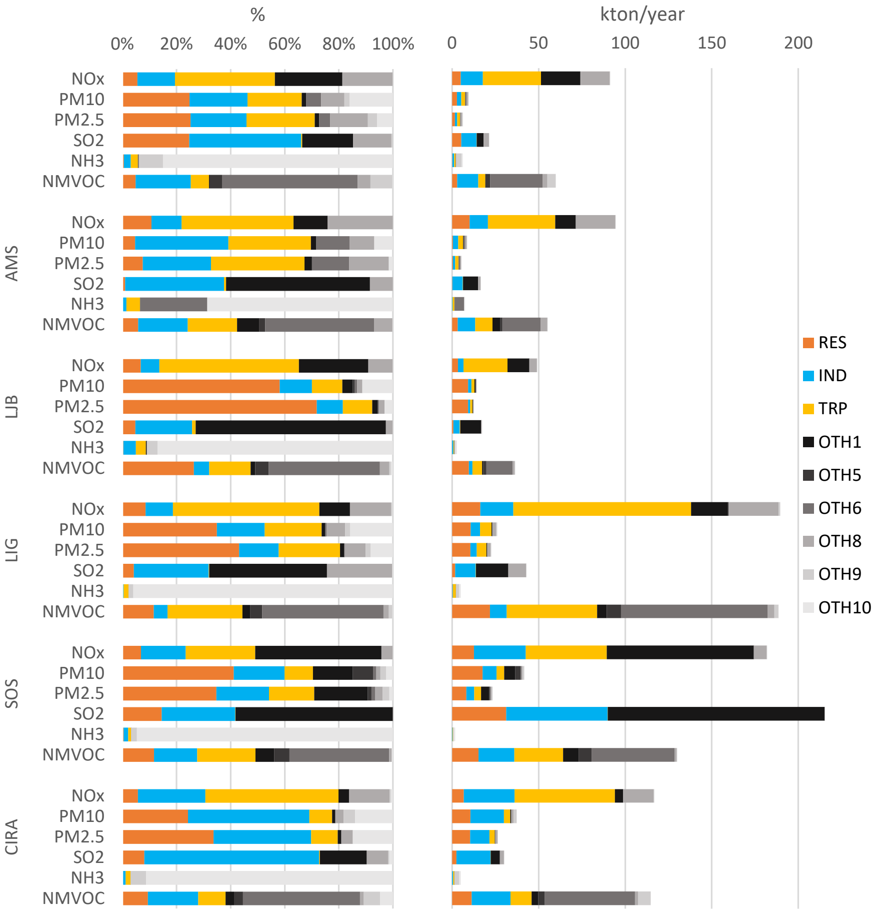

Figure 3.5. *Total annual emissions of NOx, PM10, PM2.5, SO_2, NH_3, NMVOC, for the 0.05º resolution domain of each case study, by source group (TBD- transboundary transport, RES - residential and commercial combustion, IND - industrial combustion and processes, TRP - road transport, and OTH - all the remaining sources, divided as: energy industries (OTH1), extraction and distribution of fossil fuels (OTH5), solvent use (OTH6), non-road transport and other mobile sources (OTH8), waste treatment (OTH9) and agriculture (OTH10)).*

Air quality modelling performance

Table 3.3. *CAMx performance statistics for NO_2 hourly concentrations, computed for 2010, for the case studies ensemble and for each one separately.*

	All cases	***BRS***	***AMS***	***LJB***	***LIG***	***SOS***	***CIRA***
Nº Observations [-]	117758	40849	8406	24493	21551	16211	6248
Nº Air Quality Sites [-]	15	5	1	3	3	2	1
Correlation [-]	0.50	0.55	0.59	0.54	0.19	0.54	0.58
Mean Bias [µg.m^{-3}]	-2.57	-4.93	1.5	-4.65	1.96	-4.01	3.60
Mean Error [µg.m^{-3}]	13.34	11.99	13.94	13.62	18.23	12.03	6.73
Index of Agreement [-]	0.56	0.61	0.60	0.58	0.30	0.53	0.39
RMSE [µg.m^{-3}]	19.47	17.65	18.52	20.81	24.8	17.26	10.19
MQI [-]	0.71	0.68	0.68	0.75	0.96	0.68	0.49

Table 3.4. *CAMx model performance for PM10 daily concentrations, computed for 2010, for the case studies ensemble and for each one separately.*

	All cases	***BRS***	***AMS***	***LJB***	***LIG***	***SOS***	***CIRA***
Nº Observations [-]	2948	359	639	945	0	688	317
Nº Air Quality Sites [-]	9	1	2	3	0	2	1
Correlation [-]	0.50	0.68	0.66	0.49	-	0.60	0.29
Mean Bias [µg.m^{-3}]	1.57	0.44	2.02	11.78	-	-8.97	-5.64
Mean Error [µg.m^{-3}]	13.91	7.87	10.46	17.47	-	16.39	11.72
Index of Agreement [-]	0.49	0.42	0.46	0.29	-	0.57	0.33
RMSE [µg.m^{-3}]	19.89	10.48	14.09	24.47	-	22.91	15.12
MQI [-]	0.98	0.83	0.87	1.30	-	0.98	0.94

- **Air quality concentration fields**

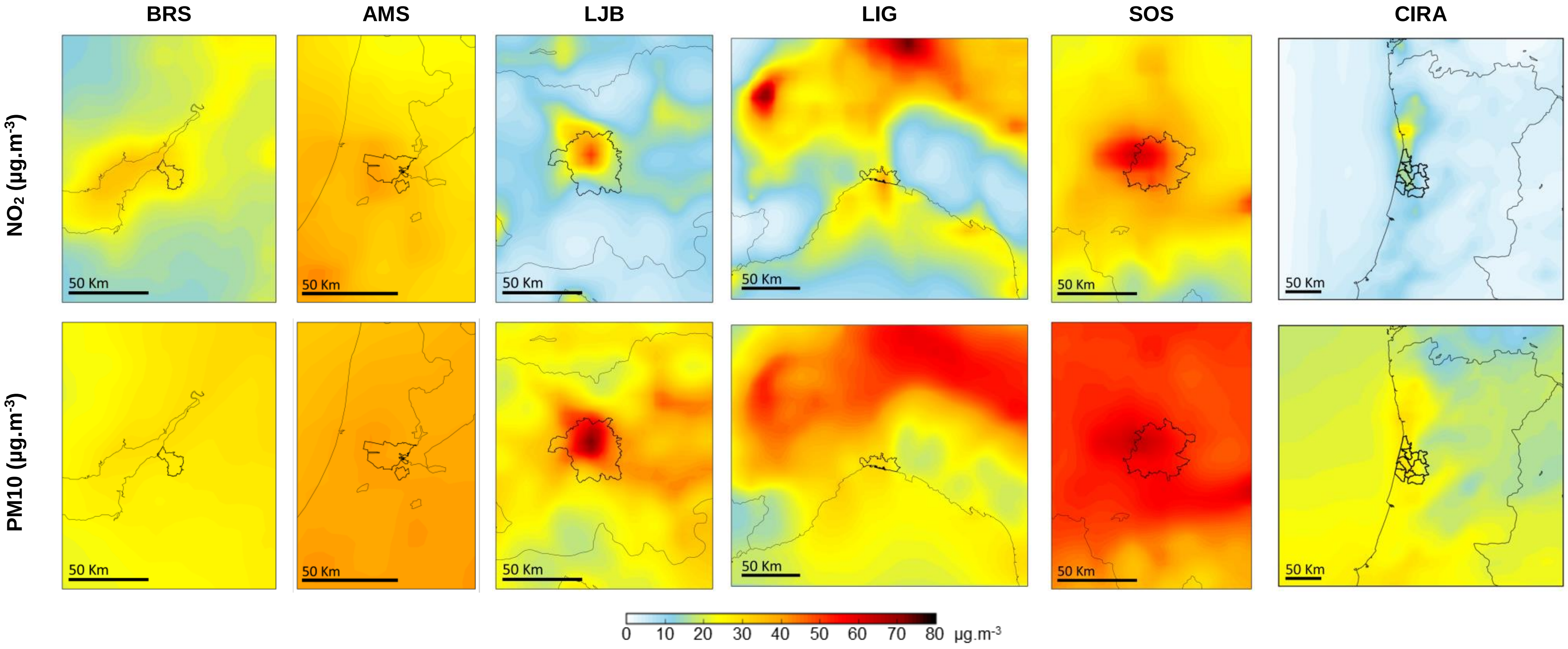

Figure 3.6. *Spatial distribution of NO_2 and PM10 winter average concentrations for each case study.*

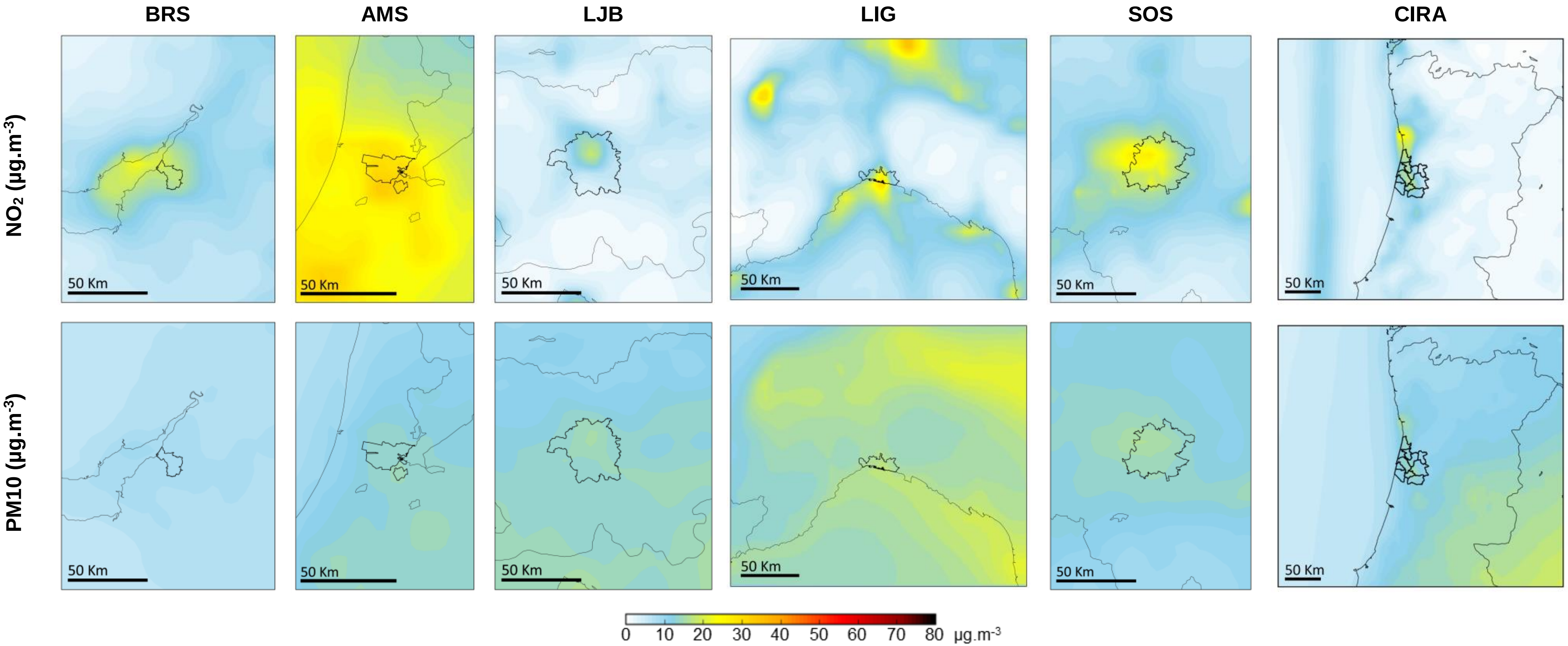

Figure 3.7. *Spatial distribution of NO_2 and PM10 summer average concentrations for each case study.*

CHAPTER 4

This Chapter has been published as:

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4. HEALTH IMPACT ASSESSMENT OF AIR POLLUTION UNDER A CLIMATE CHANGE SCENARIO: METHODOLOGY AND CASE STUDY APPLICATION

Abstract

The World Health Organization estimates that every year air pollution kills seven million people worldwide. As it is expected that climate change will affect future air quality patterns, the full understanding of the links between air pollution and climate change, and how they affect human health, are challenges for future research. In this scope, a methodology to assess the air quality impacts on health was developed. The WRF-CAMx modelling framework was applied for the medium-term future climate (considering the SSP2 4.5 scenario) and for the recent past (considered as baseline). Following the WHO recommendations, mortality health indicators were used to estimate the health impacts of long-term exposures. For that, the Aveiro Region, in Portugal, was considered as a case study. Future climate results indicate the occurrence of higher temperatures, and lower total precipitation. Despite that, improvements in the main pollutants' concentrations, and consequently in the reduction of the related premature deaths are foreseen, mainly due to the reduction of pollutants emissions imposed by the European legislation for the upcoming years. The applied approach constitutes an added value in this research field, being crucial to anticipate the effects of climate change on air quality and evaluate their impacts on human health.

Keywords: SSP (Shared Socio-economic Pathway) scenarios, air quality, WRF-CAMx, numerical modelling, urban areas, health impact assessment, premature deaths

4.1. INTRODUCTION

Nowadays, air pollution and climate change are two of the biggest environmental and health threats, with increasing concern among people worldwide (Grossberndt et al. 2021), being expected to continue for the next decades (Von Schneidemesser et al. 2015). More than 70% of the European Union (EU) population live in urban areas (Eurostat 2016), where economic activities cause high levels of air pollution (EEA 2022). Citizens' exposure to air pollutants, like particulate matter with a diameter of 10 microns or less (PM10) and with a diameter of 2.5 microns or less (PM2.5), nitrogen dioxide (NO_2), and ozone (O_3), has been threatening human health. The EU Air Quality Directive (Directive 2008/50/EC) (EC 2008) aims to protect health, vegetation, and natural ecosystems, through the definition of limit and target values for those air pollutants, among others. In 2020, less than 1% of the EU urban population lived in zones with PM2.5 and NO_2 concentrations above the EU limit values, while 12% of citizens were exposed to O_3, and 11% to PM10 levels above EU standards

(EEA 2022). However, when considering the new 2021 World Health Organization (WHO) air quality recommendations (WHO 2021), in 2020, the EU urban population exposure raised to 96% for PM2.5, 89% for NO_2, 71% for PM10, and 93% for O_3 (EEA 2022). These data have become more relevant since the EU Clean Air Programme (EC 2013) and the European Green Deal (EC 2019) proposed a revision of the ambient air quality directives, aiming to closely align the EU air quality standards with the WHO recommendations (EEA 2022). The WHO estimates that every year, ambient air pollution kills around 4.2 million people worldwide, due to stroke, heart disease, lung cancer, lower respiratory infections, and chronic obstructive pulmonary disease (WHO 2022a). In 2019, in the EU alone, the estimated premature deaths attributable to long-term exposure to PM2.5, NO_2, and O_3, concentrations were around 307000, 40400, and 16800, respectively (EEA 2021).

As air pollution is strongly dependent on meteorological conditions (Seo et al. 2017; Coelho et al. 2021), it is sensitive to climate change. According to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) (IPCC 2021), it is expected that climate change will have complex effects on chemistry, transport, and the deposition of local air pollutants. IPCC also refers to urban areas as the ones that must respond to climate change risks (Revi et al. 2014; IPCC 2021), mostly due to the complex interactions between social, economic, and environmental stressors (Radhakrishnan et al. 2017, 2018; Coelho et al. 2022b). Indeed, climate change impacts in urban areas have recently become an important and urgent research topic (Jiang et al. 2017). The full understanding of the links between climate change and air pollution in urban areas, and how they affect human health, are nowadays research challenges. Moreover, future emission scenarios should be considered due to their higher impact on air quality patterns, in line with the European Green Deal and the EU Clean Air Programme.

In the light of the above, the main aim of this work is to assess air pollution effects on human health at urban scale in the future, considering a given climate change scenario, through the application of a numerical modelling system, and using the Aveiro Region, in central Portugal, as a case study. This work will focus on the medium-term future (around 2050), following the Portuguese Roadmap for Carbon Neutrality in 2050 (Monjardino et al. 2019), under which Portugal has committed to prevent the increase of the global average temperature to well below 2°C above pre-industrial levels, and to make efforts to limit the increase to 1.5°C, in line with the Paris Agreement.

The Aveiro Region is located in the central part of Portugal, close to the Atlantic Ocean, with an area of 1 693 km^2 and around 370 000 inhabitants. The Aveiro Region includes 11 municipalities, which are distinguished by their geography, economy, activity sectors, and population density (Rodrigues et al. 2021; Coelho et al. 2022b). The major economic sector of the region is the manufacturing industry, accounting for 50% of the region's income (Coelho et al. 2018), followed by tourism, and services (Rodrigues et al. 2021). The most industrialized municipalities are typically located along the coast, closer to one of the most important seaports in Portugal. Rural municipalities, where agriculture plays an important role, are mainly situated in the inland parts of the region (Slingerland et al. 2018). According to the Köppen climate classification (Peel et al. 2007), the Aveiro Region is classified as a warm-summer Mediterranean climate (Csb). Despite the climate and the open landscape characteristic of many municipalities in the Aveiro Region, which favour pollutants dispersion, PM and NO_2 exceedances occur occasionally in some areas of the region (Rodrigues et al. 2021; Coelho et al. 2022b).

This work, easily applicable to other case studies, will fulfil the need for further studies that assess the future climate effects on air quality, with a high-resolution level, to support the identification of early climate and air pollution adaptation strategies. Additionally, several scientific communities, policymakers, and citizens, may benefit from the advances in the coordination between climate change, air quality, and health impact assessments for urban areas.

4.2. DATA AND METHODS

This section presents the data and methodology used in this work, which can be divided in two parts, namely: (i) the WRF-CAMx modelling framework applied to assess the medium-term future climate and its impacts on air quality over the Aveiro Region (section 4.2.1); and (ii) the health impact assessment (section 4.2.2), based on WHO recommendations and considering the CAMx outputs.

4.2.1. Climate and air quality modelling

To assess the future climate and air quality patterns in the study area, the WRF - Weather Research and Forecasting Model (Skamarock et al. 2019) together with the CAMx - Comprehensive Air Quality Model with Extensions (ENVIRON 2020) modelling framework was applied. These models have been widely used to assess air pollution in different case studies worldwide, and more specifically in Portugal, with reliable and realistic results (Rafael et al. 2017; Ferreira et al. 2020; Coelho et al. 2021, 2022b).

Three nested domains with increasing resolution at a downscaling ratio of five were used, with the outermost domain of 30 km horizontal resolution centred over the Iberian Peninsula, and the innermost domain of 1.2 km horizontal resolution, with 75 × 75 horizontal grid cells, focusing on the Aveiro Region (Figure 4.1).

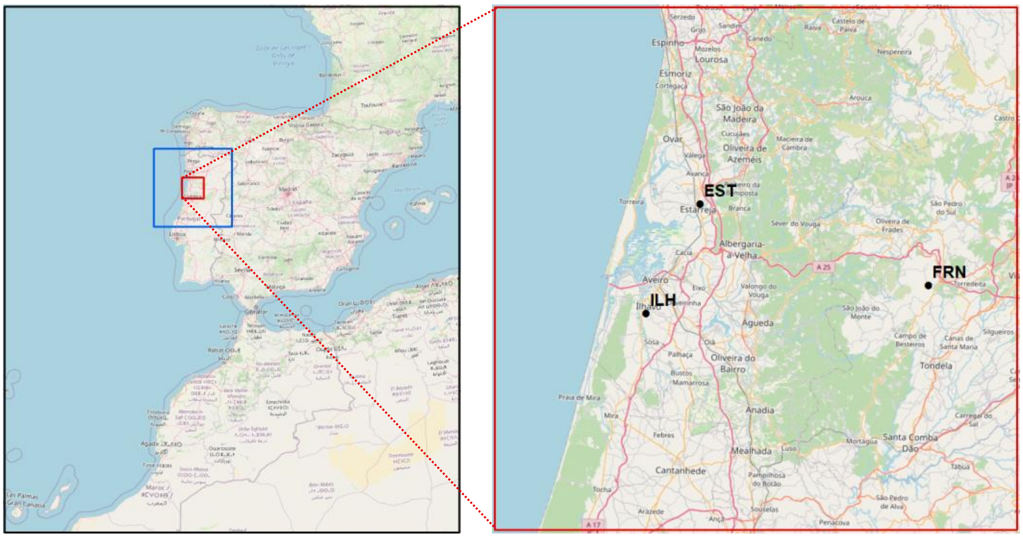


Figure 4.1. *WRF-CAMx modelling system domains with 30 (D1 - within the black rectangle), 6 (D2 - within the blue square) and 1.2 (D3 - within the red square) km resolution; and the location of the air quality monitoring stations (black points; EST: Estarreja - industrial background; ILH: Ílhavo - suburban background; and FRN: Fornelo do Monte - rural background).*

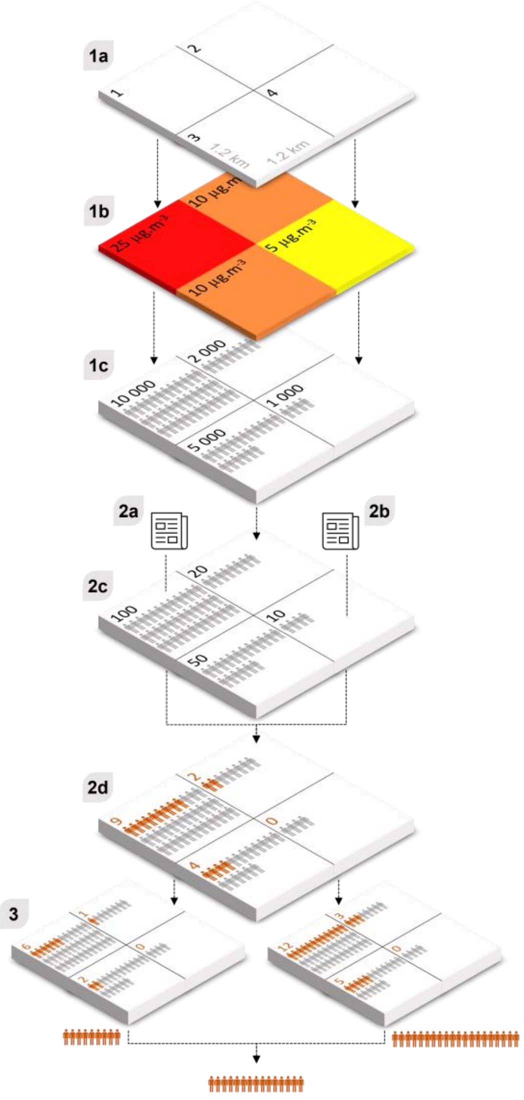
The WRF model was forced by the Max Planck Institute for Meteorology Earth System Model version 1.2 (MPI-ESM1.2-HR) (Mauritsen et al. 2019), with 0.938° horizontal resolution and 95 vertical levels. Other works (Brands et al. 2013; Marta-Almeida et al. 2016) show that MPI-ESM is considered one of the best global models to predict the climate in Europe. Previous studies (Russo et al.; Marta-Almeida et al. 2016; Rafael et al. 2020) also evaluated the use of MPI-ESM data as WRF forcing, thus ensuring the confidence in this model configuration for climate studies. The WRF physical configuration, selected based on previous studies (Coelho et al. 2020, 2021, 2023a; Marta-Almeida et al. 2016; Rafael et al. 2017), was as follows: microphysics - WRF Single-Moment 6-class scheme (Hong and Lim 2006); longwave and shortwave radiation - RRTMG (Iacono et al. 2008); surface layer - MM5 (Jiménez et al. 2012); land surface - Noah (Tewari et al. 2004); planetary boundary layer - YSU (Hong et al. 2006); cumulus - Grell-Freitas (Grell and Freitas 2014); and sea surface temperature 6-hourly update. To keep the WRF downscaling consistent with the large-scale atmospheric dynamics of the forcing data, spectral nudging was used in the outermost domain for atmospheric waves larger than 1000 km in latitude and longitude.

The modelling system was applied for two years, one statistically representative of the recent past (2014), considered as a baseline, and the other statistically representative of the medium-term future climate (2055). As one of the objectives of this work is to focus on urban areas with a high-resolution level, the selection of representative years was a compromise to guarantee WRF-CAMx outputs with high spatial resolutions, avoiding excessively high computational efforts. Based on previous studies methodologies (Coelho et al. 2021, 2023a; Ascenso et al. 2021), the selection of the representative years was supported by a climatological analysis of every year of the periods 1995-2014 (for the recent past) and 2046-2065 (for the medium-term future), provided by coarser simulations, and compared with the average of each period, to find the year with the lowest climate anomaly. This analysis was performed for temperature, precipitation, wind direction, and solar radiation, as they are considered by the scientific community as fundamental for describing the climate (IPCC 2021) and are particularly important in air pollutants transport and deposition (Aw and Kleeman 2003; Jacob and Winner 2009; Wang and Wang 2014; Kayes et al. 2019). For the medium-term future climate, the new SSP2-4.5 (Shared Socio-Economic Pathway) scenario (Fricko et al. 2017) was applied. As stated by Riahi et al. (2017), SSP2-4.5 is part of the "*middle of the road*" socio-economic pathway, considering medium challenges for both climate change adaptation and mitigation and a "*medium pollution control*" scenario, with a nominal 4.5 $W.m^{-2}$ radiative forcing level by 2100.

The CAMx initial and boundary (every 6 hours) conditions for the outermost domain were provided by the Community Atmosphere Model with Chemistry (CAM-Chem) (Emmons et al. 2020) global chemical model, with 0.9° x 1.25° spatial resolution. For the recent past climate, anthropogenic emissions were taken from the EMEP (European Monitoring and Evaluation Programme) emission inventory (EMEP 2017) and were spatially disaggregated according to the methodology described in Ferreira et al. (2020) and speciated into the Carbon Bond 6 (CB6) chemical mechanism species considered in the CAMx simulation (Yarwood et al. 2010). For the medium-term future climate, emissions were estimated by adapting the methodology defined by Sá et al. (2015) and considering the emission projections from the Portuguese roadmap for carbon neutrality (Monjardino et al. 2019), in line with the SSP2-4.5 scenario considered in the WRF simulation. According to this methodology, medium-term future climate emissions, when compared to the recent past, show an average emission reduction of approximately 33% for PM10, 38% for PM2.5, 68% for NOx, 27% for NMVOC, 70% for CO, 15% for NH_3 and 35% for SOx. Note that these are average emission reductions for all source activities despite the emission projections foreseeing an increase for some activity sectors (e.g., NMVOC and PM10 in industrial combustion and processes sector, CO and NMVOC in agriculture, see Figure 4.6 of supplementary material (section 4.6)).

4.2.2. Health impact assessment

The effects of PM2.5, NO_2, and O_3 long-term exposure on mortality have been reported in several epidemiological and toxicological studies (WHO 2013a). Several studies (Næss et al. 2007; Brook et al. 2010; Amnuaylojaroen et al. 2022) indicate that the exposure to PM2.5 is associated with damaged lung function, systemic inflammation, and alteration of the electrical processes of the heart, as well as a causal association with cardiovascular and cardiorespiratory mortality. The associations of long-term NO_2 with respiratory and cardiovascular mortality, with effects on natural and cause-specific mortality, are similar to those estimated for PM2.5, according to several cohort studies (Brunekreef 2007; Cesaroni et al. 2013; WHO 2013a; Marmett et al. 2022). Regarding O_3 long-term exposure, several cohort analysis suggested an effect on respiratory or cardiorespiratory mortality, especially in people with potential predisposing conditions (Lipfert et al. 2006; Marmett et al. 2022; Soares and Silva 2022). For these pollutants, following the approach usually applied in the EEA (European Environment Agency) annual health assessments, described in Soares et al. (2019), mortality health outcomes were analysed since it is the most serious health impact and the one with the most robust scientific evidence (EEA 2018). For O_3, mortality due to respiratory diseases was considered, while for PM2.5 and NO_2, all-cause (natural) mortality was considered, both at ages above 30 years. The health outputs were traduced in the number of premature deaths, for the population affected by air pollution living within the study area. According to the WHO (2016), the health impact assessment can be performed in three major steps, namely: (i) population exposure assessment; (ii) health risk estimation; and (iii) uncertainty calculation. Figure 4.2 shows the health impact assessment scheme, with an example for PM2.5, long-term exposure, all-cause (natural) mortality, using WHO 2013 methodology and hypothetical concentration and population values.



Population exposure assessment:

1a) Grid: the area for which the health risk assessment will be calculated consists of a 4 cells grid with 1.2x1.2 km^2 horizontal resolution.

1b) Concentration Map: the air quality modelling results, for cells 1 to 4, show PM2.5 annual concentrations that vary between 5 and 15 $\mu g.m^{-3}$, for the year under analysis.

1c) Population/Exposure: the population is distributed across the grid and the exposure is calculated. For example, in cell 1, 10000 inhabitants are exposed to 15 $\mu g.m^{-3}$ of PM2.5.

Health risk estimation:

2a) Baseline Concentration: the baseline concentration for PM2.5 is 0 $\mu g.m^{-3}$, meaning that, for instance, for grid 1 the effect of the whole range of 25 $\mu g.m^{-3}$ of PM2.5 will be estimated.

2b) Relative Risk: in the case of PM2.5, the concentration-response function used for total (all-cause) mortality in people above 30 years of age implies a relative risk of 1.062 per 10 $\mu g.m^{-3}$. Thus, assuming linearity, an increase of 10 $\mu g.m^{-3}$ of PM2.5 is associated with a 6.2% increase in total mortality in the total population considered.

2c) Mortality: the total mortality (incidence base) in the country for the year under analysis and the population over 30 years of age is 10 deaths per 1000 inhabitants, so the number of deaths per grid is as shown.

2d) Premature Deaths: the number of deaths attributed to exposure to PM2.5 in each cell is obtained from:

*Relative Risk (RR) = exp (β * concentration – baseline concentration) = exp (0.00602 * concentration - 0)*

For cell 1: 1.162281

Population Attributable Fraction (PAF) = (RR-1) / RR

For cell 1: 0.139623

*Premature Deaths (PD) = PAF * mortality * population*

For cell 1: 13.96 ≈ 14

Uncertainty calculation:

3) Uncertainty: the uncertainty range is calculated using the lower (1.040) and upper (1.083) limits of the relative risk of PM2.5, instead of 1.062.

The total mortality is then expressed as 18 premature deaths, with a 95% confidence interval between 12 and 24.

Figure 4.2. *Health impact assessment scheme. Example for PM2.5, long-term exposure, all-cause (natural) mortality, using WHO 2013 methodology (adapted from EEA (2018)).*

The main objective of the first step, the population exposure assessment, is to cross gridded pollutant concentration maps with gridded population data, at the same spatial resolution, resulting in a map with the population exposed, by grid cell, to the selected pollutant. For that, in the present work, the following data were used:

(i) CAMx surface concentrations results for PM2.5, NO_2 and O_3, by grid cell, for each period analysed (steps 1a and 1b in Figure 4.2 scheme);

(ii) Most recent population data stratified by age and sex, from Census 2021, by the Portuguese national institute for statistics (INE) (INE 2021), for the recent past, and INE projection for 2050 (INE 2017) for the future, spatially disaggregated according to the CAMx grid (step 1c in Figure 4.2 scheme).

In the second step, health risk estimation, relative risk data from epidemiological studies and incidence mortality statistics are used, together with the population exposed from step 1, to assess the number of premature deaths, by grid cell, as shown in steps 2a to 2d in Figure 4.2 scheme. To apply it to the current case study, the following input data were used:

(i) Portuguese annual total number of deaths broken down by age, cohort and sex, from the European mortality database (WHO 2022b) (step 2c in Figure 4.2 scheme);

(ii) Baseline concentration (C_0) and concentration-response functions (*CRF*) were used for the estimation of the relative risk (*RR*) (2a and 2b from Figure 4.2 scheme) as follows:

$$RR = e^{\beta \cdot (C_i - C_0)}$$

where, C_i is the concentration level the population is exposed to in grid cell i and β is the coefficient of the *CRF*. Assuming an exponential behaviour, β can be estimated based on the *CRF*:

$$CRF/UC = e^{\beta \cdot UC}$$

where *CRF* applied are described in Table 4.1 and *UC* = 10 µg.m^{-3}.

For that, due to the recent update of the WHO air quality guidelines, two different methodologies were used: (i) the so-called WHO 2013, according to the recommendations of HRAPIE (WHO 2013b) and REVIHAAP (WHO 2013a) projects, considering *CRF* from Jerrett et al. (2009) for O_3 and from Hoek et al. (2013) for PM2.5 and NO_2; and (ii) the so called WHO 2021, according to the recommendations of new WHO air quality guidelines (WHO 2021), considering *CRF* from Chen and Hoek (2020) for PM2.5 and Huangfu and Atkinson (2020) for O_3 and NO_2. Table 4.1 presents detailed information on the *CRF* used, as well as a source of mortality data and C_0. From Table 4.1, major differences between the WHO 2013 and the WHO 2021 *CRF* methodologies can be found for mortality health outcomes associated with PM2.5 and NO_2 long-term exposure. For PM2.5, WHO 2021 considers a higher relative risk than WHO 2013. According to the WHO 2021 methodology, an increase in 10 µg.m^{-3} of PM2.5 is associated with an 8% increase in total mortality (Hoek et al. 2013), while in WHO 2013 the increase in total mortality was only 6.2% (Chen and Hoek 2020). However, WHO 2021 assumes a PM2.5 baseline concentration of 5 µg.m^{-3}, below which no health effects are expected, while WHO 2013 assumes expected health effects no matter the magnitude of PM2.5 concentrations. For NO_2 the opposite happens, with a decrease in the relative risk from 5.5% (Hoek et al. 2013) in WHO 2013 to 2% (Huangfu and Atkinson 2020) in WHO 2021, and the assumption that lower concentrations (above 10 µg.m^{-3}, rather than above 20 µg.m^{-3} in WHO 2013) may have health impacts. The new baseline concentrations were determined by the WHO (2021), using the average of the five lowest 5th percentile levels measured in five selected studies for PM2.5 (Weichenthal et al. 2014; Villeneuve et al. 2015; Pinault et al. 2016, 2017; Cakmak et al. 2018) and NO_2 (Hart et al. 2011, 2013; Tonne and Wilkinson 2013; Carey et al. 2013; Turner et al. 2016).

Table 4.1. *CRF used according to the WHO 2013 (WHO 2013a, b) and WHO 2021 (WHO 2021) recommendations.*

Pollutant	**RR per 10 µg.m^{-3} (95% CI)**		**Baseline Concentration (C_0)**		**Source of mortality data**	**Health outcome**
	WHO 2013	**WHO 2021**	**WHO 2013**	**WHO 2021**		
PM2.5, annual mean	1.062 (1.040; 1.083) (Hoek et al. 2013)	1.08 (1.06; 1.09) (Chen and Hoek 2020)	> 0 µg.m^{-3}	> 5 µg.m^{-3}	European mortality database, ICD-10: A-R (WHO 2022b)	Mortality, all-cause (natural), age 30+ years
NO_2, annual mean	1.055 (1.031; 1.080) (Hoek et al. 2013)	1.02 (1.01; 1.04) (Huangfu and Atkinson 2020)	> 20 µg.m^{-3}	> 10 µg.m^{-3}		
O_3, SOMO35[1]	1.014 (1.005; 1.024) (Jerrett et al. 2009)	1.01 (1.00; 1.02) (Huangfu and Atkinson 2020)	> 70 µg.m^{-3}	> 70 µg.m^{-3}	European mortality database, ICD-10: J00-J99 (WHO 2022b)	Mortality, respiratory diseases, age 30+ years

[1] Summer months (April–September), average of daily maximum 8-hour mean over 35 ppb.

The contribution of a risk factor to a premature death can be estimated by means of population attributable fraction (*PAF*), that can be calculated from the *RR*, for every grid cell i, as follows:

$$PAF = (RR - 1)/RR$$

Thus, the premature deaths (*PD*) can be estimated from *PAF*, assuming the baseline incidence as the crude death rates by age a and sex s ($CDR_{a,s}$) and the population at grid cell i (pop_i), as follows:

$$PD = \sum_{a,s} CDR_{a,s} \cdot pop_i \cdot PAF$$

In step three, the uncertainty associated with the health risk estimation is calculated. Most of the uncertainty is related to the CRF used, since it derives from the assumptions made in epidemiological studies that take into account other confounding factors that can also have an impact on mortality (e.g., smoking, diet, lifestyle) (WHO 2016). Thus, following the EEA approach (Soares et al. 2019), the uncertainty is calculated using a 95% confidence interval, indicating that there is a 95% probability that the true value lies in the range defined by the interval. Finally, after the population exposure assessment, the health risk estimation and the uncertainty calculation, the total mortality due to PM2.5, NO_2 and O_3 long-term exposure was assessed.

Additionally, following the EEA approach (Soares et al. 2019), the years of life lost (*YLL*) indicator was also estimated as a mortality-related health outcome. *YLL* is defined as the years of potential life loss due to premature death. It is an estimate of the average number of years that a person would have lived if the person would not have died prematurely. *YLL* considers the age at which death occurs and is greater for deaths at a younger age and lower for deaths at an older age (Murray 1996). It gives, therefore, more nuanced information than the number of *PD* alone. *YLL* is determined by relating *PD* with life expectancy (*LE*) by sex s and age a, for every grid cell i:

$$YLL = \sum\nolimits_{a,s} LE_{a,s} \cdot PD$$

where $\mu_{a,s}$ is the average time a person is expected to live, based on the year of their birth, their current age and sex (available in Census 2021 (INE 2021)).

4.3. RESULTS AND DISCUSSION

This section presents the analysis and discussion of the obtained results for climate and air quality (section 4.3.1), and for the health impact assessment (section 4.3.2), for the reference and medium-term future.

4.3.1. Climate and air quality modelling

For both climate and air quality assessment, results obtained for the medium-term future climate were compared with the results for the recent past. The comparison was made for the following periods: (i) annual; (ii) spring (March to May); (iii) summer (June to August); (iv) autumn (September to November); and (v) winter (December to February).

For the climate change assessment, mean temperature and total precipitation were analysed. Figure 3 shows the annual average of mean temperature and total precipitation, for the medium-term future climate, recent past climate, and the difference between them. Seasonal results can be found in the supplementary material (section 4.6), in Figures 4.7 and 4.8.

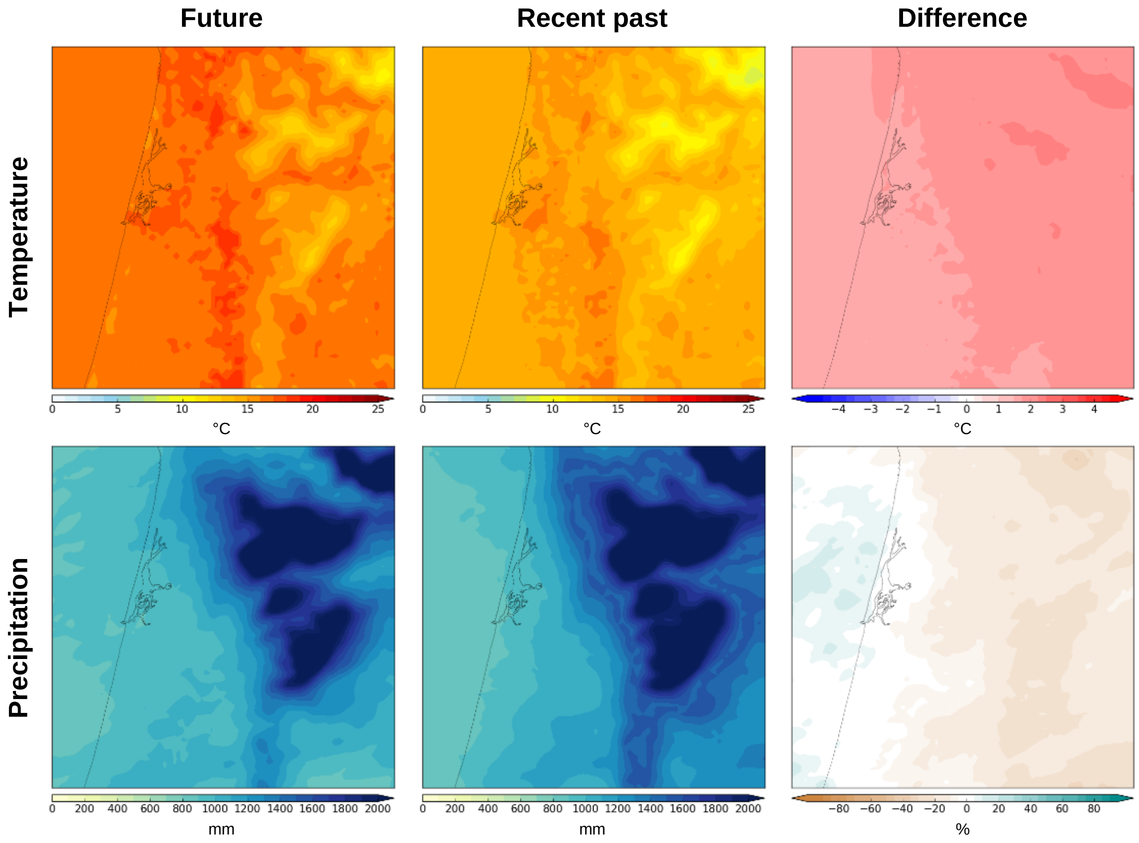


Figure 4.3. *Annual mean temperature and total precipitation, for the medium-term future climate (2055), recent past climate (2014) and the difference between them (2055 – 2014).*

The differences between the medium-term future climate and the recent past climate for the annual mean temperature, show an increase in future temperature between 1 and 2°C, with lower differences estimated closer to the coastline and in the lower altitude areas. For the annual total precipitation, a decrease of up to 40% is expected in some areas of the region. Over the coastline, no differences are projected in total precipitation and an increase, between 10 and 20%, is predicted over some parts of the Atlantic Ocean. Notwithstanding, in agreement with what has been shown in other studies (IPCC 2021), a trend toward reduced precipitation in the medium-term future is clear. This precipitation trend, together with the higher temperatures projected, will lead to drier conditions, as has been already observed in southern Europe countries (Vicente-Serrano et al. 2014). Regarding the seasonal analysis, the results show different variations. The highest temperature differences are projected for the summer, where the mean temperature can increase up to 4°C, and the lowest differences are projected for autumn, with some parts of the study region for which differences are not expected. Moreover, for none of the seasons, is a temperature decrease expected in the future. Differences in precipitation also exhibit several variations across the seasons. For summer and autumn, a precipitation reduction of around 80% and 45%, respectively, is expected in almost the entire study region. However, in winter and spring, the precipitation decrease projected for the medium-term future is only foreseen for the inland regions of the domain, with an expected increase over the Atlantic Ocean and coastal areas. These results are in line with previous studies (Jacob et al. 2014; Marta-Almeida et al. 2016; Rafael et al. 2017; Coelho et al. 2020, 2021), which also project higher temperatures and lower total precipitation, even though they are applied to other case studies, using different climate change scenarios, and/or considering distinct future periods. Greater warming over land will change key features of water cycle, affecting the precipitation variability (IPCC 2021). According to other studies (MedECC 2020; IPCC 2021), it is expected a precipitation increase over high latitudes and oceans, and a decrease over large parts of the subtropics in response to greenhouse gas-induced warming. Thus, coastal areas will be the zones with the smaller changes, as it is where the climate change signals change the sign (Jacob et al. 2014). The projected decrease in precipitation over land, amplified by the high evapotranspiration related to strong warming, could also lead to more frequent and intense periods of drought (Vicente-Serrano et al. 2014; MedECC 2020; Coelho et al. 2020).

These future climate patterns will have complex effects on chemical reactions, transport, dispersion, and the deposition of air pollutants (Kayes et al. 2019). Temperature affects the chemical reaction rates as well as the dispersion and deposition of chemical compounds (Aw and Kleeman 2003; Jacob and Winner 2009). The precipitation washout also plays an important role in removing pollutants from the atmosphere, by wet deposition (Duhanyan and Roustan 2011; Guo et al. 2016). Figure 4.4 shows the annual average concentration of NO_2, PM10, and PM2.5, and the annual average concentration of the maximum daily 8 h mean O_3, for the medium-term future climate, recent past climate, and the difference between them. Seasonal results can be found in section 4.6 (supplementary material), Figures 4.9 to 4.12.

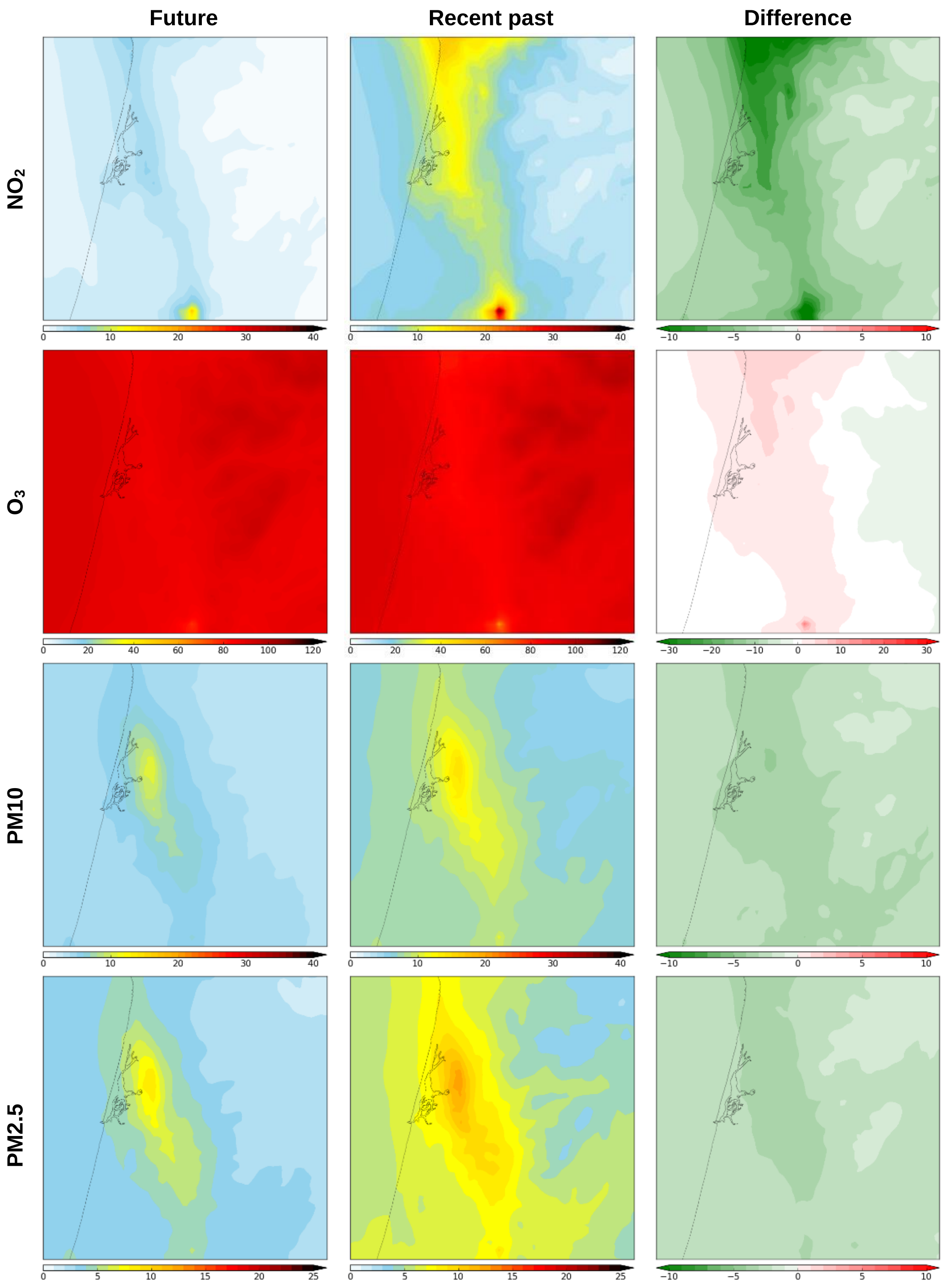


__Figure 4.4.__ Annual average concentration of NO_2, PM10, and PM2.5, and the annual average concentration of the maximum daily 8 h mean O_3, in $\mu g.m^{-3}$, for the medium-term future climate (2055), recent past climate (2014) and the difference between them (2055 – 2014). For future and recent past maps, the maximum value of the colour scale represents the EU limit value.

For both periods analysed, medium-term future and recent past climate, all pollutants show similar spatial patterns, but with different magnitudes of the concentrations. The differences between the medium-term future climate and the recent past climate, result in a decrease in the annual average concentration of NO_2, PM10, and PM2.5 up to 10, 4, and 3 $\mu g.m^{-3}$, respectively. For the medium-term future climate, the differences in the annual average concentration of the maximum daily 8-h mean O_3 show decreases (up to 4 $\mu g.m^{-3}$) and increases (up to 10 $\mu g.m^{-3}$), depending on the area. When analysed seasonally, O_3 shows variations along the seasons, while the remaining pollutants maintain a similar behaviour to the annual one. For NO_2, major differences are found in the same areas where higher concentration values are obtained for the recent past, namely: (i) in the north of the domain, where part of the Porto metropolitan area (the second largest urban area in Portugal) is located, and that influences Aveiro Region air quality due to the pollutants transport by the north/northwest dominant winds; and (ii) in a small area in the south, the site of one of the major cement factories in Portugal. The 68% of NOx emission reduction projected for the future, mostly related to transportation, industry and energy sectors (see Figure 4.6 of supplementary material, section 4.6), will lead to this reduction in the NO_2 concentrations. In contrast, in the locations with higher NO_2 concentration reductions, an air quality deterioration due to O_3 pollution will be expected, due to the reduction of NOx values, but also due to the projected temperature increase, favouring photochemical phenomena. Concerning PM10 and PM2.5 results, higher concentrations were estimated over the most industrialized areas of the Aveiro Region, where major concentration reductions are also projected for the future. Although a decrease in total precipitation is expected in the Aveiro Region, which could lead to an increase in both PM10 and PM2.5 concentrations (Duhanyan and Roustan 2011; Guo et al. 2016), this does not happen due to the reduction of the future PM emissions considered. These results are in line with previous studies applied for Portuguese case studies (e.g., Sá et al. 2016; Coelho et al. 2022a, 2023b), as well as for other countries/regions (e.g., Markakis et al. 2014, 2016; Lacressonnière et al. 2014). Similarly to what was verified in the current study, Sá et al. (2016) recorded a decrease of the NO_2 (around 15%), and an increase of O_3 (up to 3%) annual mean concentrations in Portuguese coastal regions. Lacressonnière et al. (2014) projected a decrease in PM10 and NOx annual mean concentration, around 2.1 $\mu g.m^{-3}$ for both pollutants, and an increase, up to 21.6 $\mu g.m^{-3}$, for O_3, for continental Europe in 2030.

For the medium-term future, the projected decrease in NO_2, PM10, and PM2.5 annual concentrations will ensure that the limit values imposed by the European Union Air Quality Directive (EC 2008) will not be exceeded. For the same period, despite the projected increase in the maximum daily 8 h mean O_3 concentrations, they will also be below the defined target values for the protection of human health. However, if the new WHO air quality guidelines (WHO 2021) are considered, the future annual concentration of PM2.5 and maximum daily 8 h mean O_3 will be above the recommended air quality guideline levels (5 and 60 $\mu g.m^{-3}$, respectively), in some areas of the Aveiro Region.

To give an insight into the accuracy of the presented results, the CAMx performance was evaluated through the application of performance statistics, based on the monitored NO_2, O_3, and PM10 concentrations during 2014, from the Air Quality e-Reporting database (EEA 2019), for the three air quality monitoring stations identified in Figure 4.1. In general, the CAMx tends to underestimate NO_2 and PM10 concentrations, while O_3 concentrations tend to be overestimated. However, according to the FAIRMODE guidelines (Janssen and Thunis 2022), the obtained results fulfil the modelling quality objective, defined as the minimum level of quality to be achieved by a model for policy use. For a detailed quantitative analysis of the CAMx performance, see supplementary material (section 4.6), Table 4.3.

4.3.2. Health impact assessment

Premature deaths, due to PM2.5, NO_2, and O_3 long-term exposure, for the mid-term future climate, the recent past climate, and the difference between them, considering both the WHO 2013 and the WHO 2021 *CRF* methodologies, are shown in Table 2.

Table 4.2. *Premature deaths and YLL, due to PM2.5, NO_2, and O_3 long-term exposure, for the medium-term future climate (2055), recent past climate (2014) and the difference between them (2055 – 2014), considering both the WHO 2013 and the WHO 2021 CRF methodologies. Premature deaths and YLL considering the 95% confidence interval are shown in brackets. Note that negative values mean avoided premature deaths and YLL.*

Pollutant	**Health outcome**	**WHO 2013**			**WHO 2021**		
		Future	**Recent past**	**Differences**	**Future**	**Recent-past**	**Differences**
PM2.5, annual mean	Premature deaths	3297 (1914; 4836)	6945 (3864; 9767)	-3648 (-1950; -4931)	93 (49; 109)	2234 (1579; 2546)	-2141 (-1530; -2437)
	YLL	34029 (19124; 50281)	73823 (40185; 105728)	-39794 (-21061; -55446)	891 (472; 1 044)	22699 (15706; 26260)	-21808 (-15234; -25216)
NO_2, annual mean	Premature deaths	0 (0; 0)	13 (6; 21)	-13 (-6; -21)	0 (0; 0)	241 (83; 560)	-241 (-83; -560)
	YLL	0 (0; 0)	124 (58; 213)	-124 (-58; -213)	0 (0; 0)	2472 (794; 6113)	-2472 (-794; -6113)
O_3, SOMO35	Premature deaths	0 (0; 0)	0 (0; 0)	0 (0; 0)	0 (0; 0)	0 (0; 0)	0 (0; 0)
	YLL	0 (0; 0)	0 (0; 0)	0 (0; 0)	0 (0; 0)	0 (0; 0)	0 (0; 0)

When considering the WHO 2013 *CRF* methodology, PM2.5 long-term exposure led to 3297 (95% CI: 1914 to 48369) and 6945 (95% CI: 3864 to 9767) premature deaths in the medium-term future and recent past climate, respectively, representing around 53% of premature deaths avoided in the future. These premature deaths represent 73823 (95% CI: 40185 to 105728) *YLL* in the recent past climate and 34029 (95% CI: 19124 to 50281) in the medium-term future climate. For NO_2, 13 premature deaths, with a 95% CI between 6 and 21, are estimated for the recent past climate, accounting for 124 *YLL*, with a 95% CI between 58 and 213. For the future no premature deaths and *YLL* are expected. If the WHO 2021 *CRF* methodology is considered, PM2.5 long-term exposure led to 93 (95% CI: 49 to 109) premature deaths and 891 (95% CI: 472 to 1044) *YLL* in the medium-term future, meaning around 96% of premature deaths and *YLL* avoided, when compared to the recent past values. For NO_2, 241 premature deaths and 2472 *YLL* are estimated in the recent past climate, with a 95% CI between 83 and 560, and between 794 and 6113, respectively, and, as for the WHO 2013 *CRF* methodology, no premature deaths and *YLL* are expected for the future. For both *CRF* methodologies and periods considered, O_3 long-term exposure will not result in premature deaths and *YLL*. Regardless of the *CRF* methodology used, there will be a reduction in the number of premature deaths and the associated *YLL*, related to PM2.5 and NO_2 long-term exposure, due to the projected air quality improvement for the medium-term future. However, premature deaths and *YLL* will also continue to occur in the future, due to long-term exposure to PM2.5 pollution, even if in a smaller number. From Table 4.2 it can be also concluded that the use of different *CRF* methodologies could substantially impact the estimated number of premature deaths and *YLL*. When considering the WHO 2021 methodology, the changes in the relative risk and baseline concentration used will impact the estimation of the health risk and, consequently the number of premature deaths and *YLL*.

For PM2.5, although there is an increase in the relative risk, the reduction of the considered range concentrations from all to only above 5 µg.m^{-3}, associated with the baseline concentration used, led to a reduction in the number of premature deaths and the associated YLL. However, for NO_2, the opposite is verified, with an increase in premature deaths, and consequently on YLL, mainly due to the change in the baseline concentration. Additionally, the confidence interval shows some changes, with a smaller range between the lower and upper values, due to the changes in the lower and upper relative risk considered.

To better understand where premature deaths are expected to occur, maps of mortality due to NO_2 and PM2.5 long-term exposure were also produced and are presented in Figure 4.5. For the two periods and the CRF methodologies considered, O_3 mortality maps were not drawn as no premature deaths were obtained.

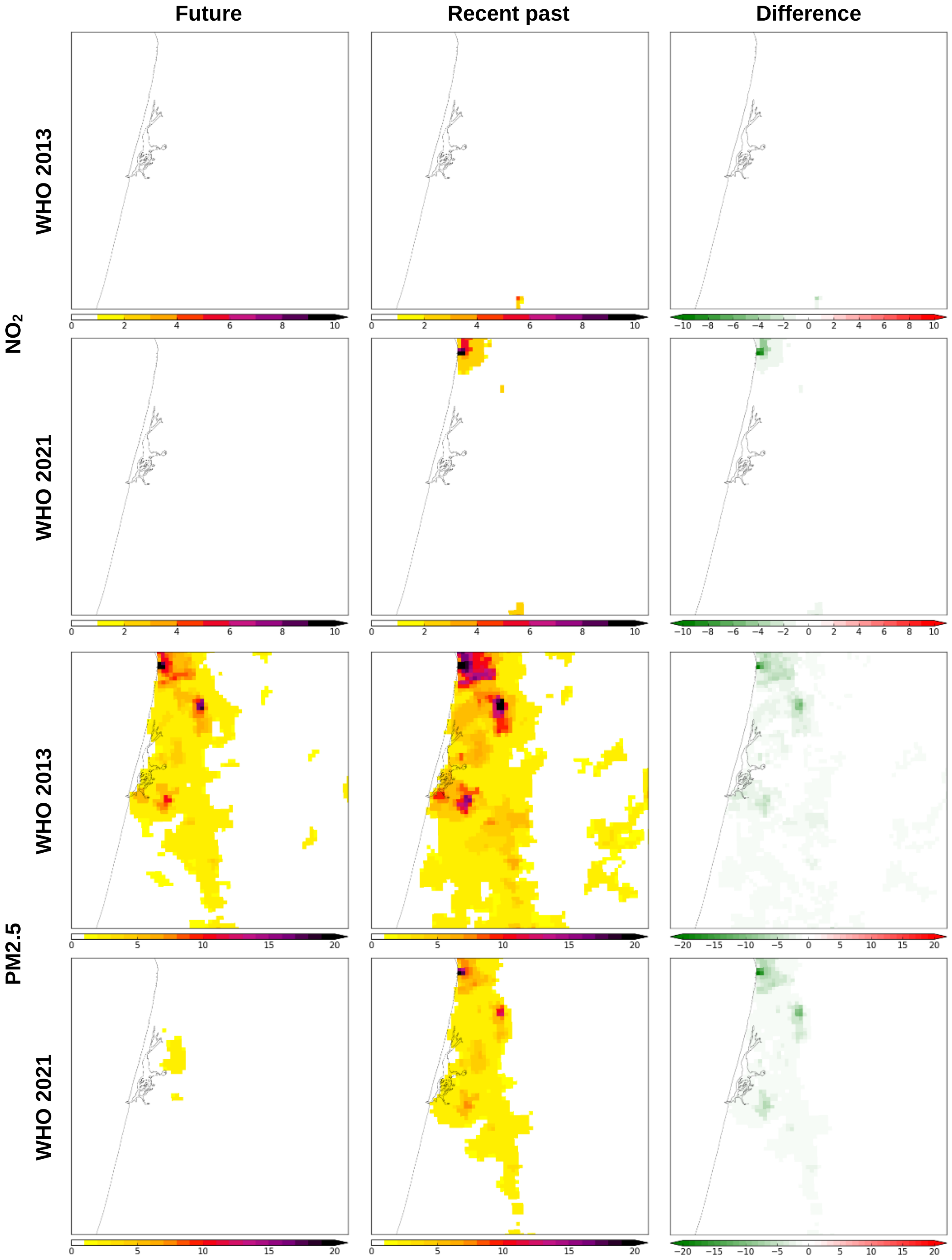


Figure 4.5. *Mortality due to NO_2 and PM2.5 long-term exposure, expressed in number of premature deaths, for the medium-term future climate (2055), recent past climate (2014) and the difference between them (2055 – 2014), considering both the WHO 2013 and the WHO 2021 CRF methodologies. Note that negative values mean avoided premature deaths.*

The projected premature deaths associated with NO_2 and PM2.5 show similar spatial patterns for medium-term future climate and recent past climate, but with different magnitudes of values. According to Figure 4.5, premature deaths are mostly obtained near the coastline, where the population density is higher, with special relevance in the Porto metropolitan area, in the north of the domain, but also highly industrialized areas, such as the one in the Aveiro Region, where higher pollutant concentrations were previously obtained. Regarding the effects of the CRF methodology used in the spatial distribution of premature deaths, this is more evident for NO_2. When using the WHO 2021 methodology, with the reduction of the baseline concentration value, the relevance of NO_2 concentrations in urban areas is highlighted, as shown in Figure 4.5, where premature deaths are foreseen in the Porto metropolitan area. The same is not true when the WHO 2013 methodology is used. As already analysed in Table 4.2, regardless of the CRF methodology used, in the medium-term future climate there will be a reduction in the number of premature deaths related to both NO_2 and PM2.5 long-term exposure.

For the recent past climate, these results are in line with those obtained by EEA (2017). Although the reference and future years are different, as well as the climate change scenario and the applied case study, the studies from Silva et al. (2016), Partanen et al. (2018), and Likhvar et al. (2015), also projected a decrease in the number of premature deaths due to air pollutants long-term exposure, in the future.

4.4. CONCLUSIONS

In this work, the WRF-CAMx modelling system, followed by a health impact assessment, was applied to the Aveiro Region urban area, in Central Portugal. Climate change results for the medium-term future climate, when compared with the recent past climate, project higher mean temperature and lower total precipitation for the Aveiro Region. Despite these results, general improvements in air quality are foreseen, with no exceedances to the EU air quality limited values, mainly due to the reduction in future emissions imposed by European legislation. Nonetheless, some concerns regarding PM2.5 and O_3 concentrations remain when the new WHO air quality recommendation levels are considered. Following the air quality trend, mortality due to pollutants' long-term exposure will also decrease, with only premature deaths related to PM2.5 pollution being expected, regardless of the CRF methodology used (WHO 2013 or WHO 2021).

Although this joint analysis of climate change, air quality, and health impacts give valuable information on these research fields, some limitations can be identified, and caution should be taken in using these results. As for all studies based on numerical modelling, uncertainties resulting from the modelling formulations and parameterizations, the input data (e.g., meteorology, emission inventories, population data), the CRF methodology, as well as the spatial resolution used, can affect the model results accuracy and also the reliability of the health impact assessment. Additionally, land use patterns will be influenced by climate change, and will impact emissions and, consequently, air quality, thus land use changes should be considered in future works.

Moreover, this study allowed to conclude that the use of different CRF methodologies in the health impact assessment could substantially impact the number of premature deaths. This evidences the magnitude of uncertainty related to the estimation of health impacts of air pollution and reinforces the need for an adequate awareness when communicating this type of scientific results to stakeholders and the general population.

Notwithstanding, the methodology applied in this work constitutes an added value in this research field, providing valuable information for policymakers and helping citizens increase awareness about climate change, air pollution, and human health impacts. Furthermore, both modelling and epidemiologist scientific communities may apply the developed methodology in other areas and benefit from advances in the harmonization between climate change, air quality, and health impact assessments for urban areas.

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4.6. SUPPLEMENTARY MATERIAL

Figure 4.6 shows the medium-term future emission projections from the Portuguese roadmap for carbon neutrality (Monjardino et al. 2019), expressed in % of the 2014 total emission, for CO, NMVOC, NH_3, NOx, PM10, PM2.5, and SO_2, by source activity.

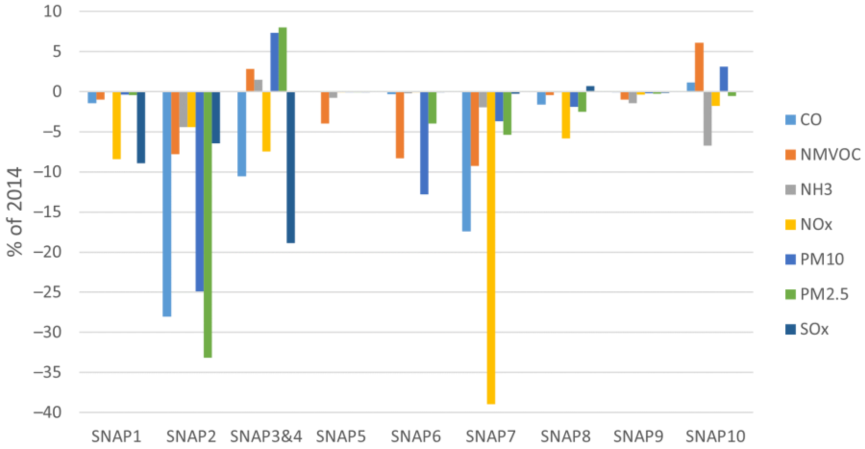

Figure 4.6. *Medium-term future emission projections from the Portuguese roadmap for carbon neutrality (Monjardino et al. 2019), expressed in % of the 2014 total emission, for CO, NMVOC, NH_3, NOx, PM10, PM2.5, and SO_2, by source activity (SNAP1 – energy industries; SNAP2 - residential and commercial combustion; SNAP 3&4- industrial combustion and processes; SNAP5 - extraction and distribution of fossil fuels; SNAP6 - solvent use; SNAP7 - road transport; SNAP8 - non-road transport and other mobile sources; SNAP9 - waste treatment; and SNAP10 – agriculture).*

Figure 4.7 shows the seasonal mean temperature, in °C, for the medium-term future climate, recent past climate and the difference between them.

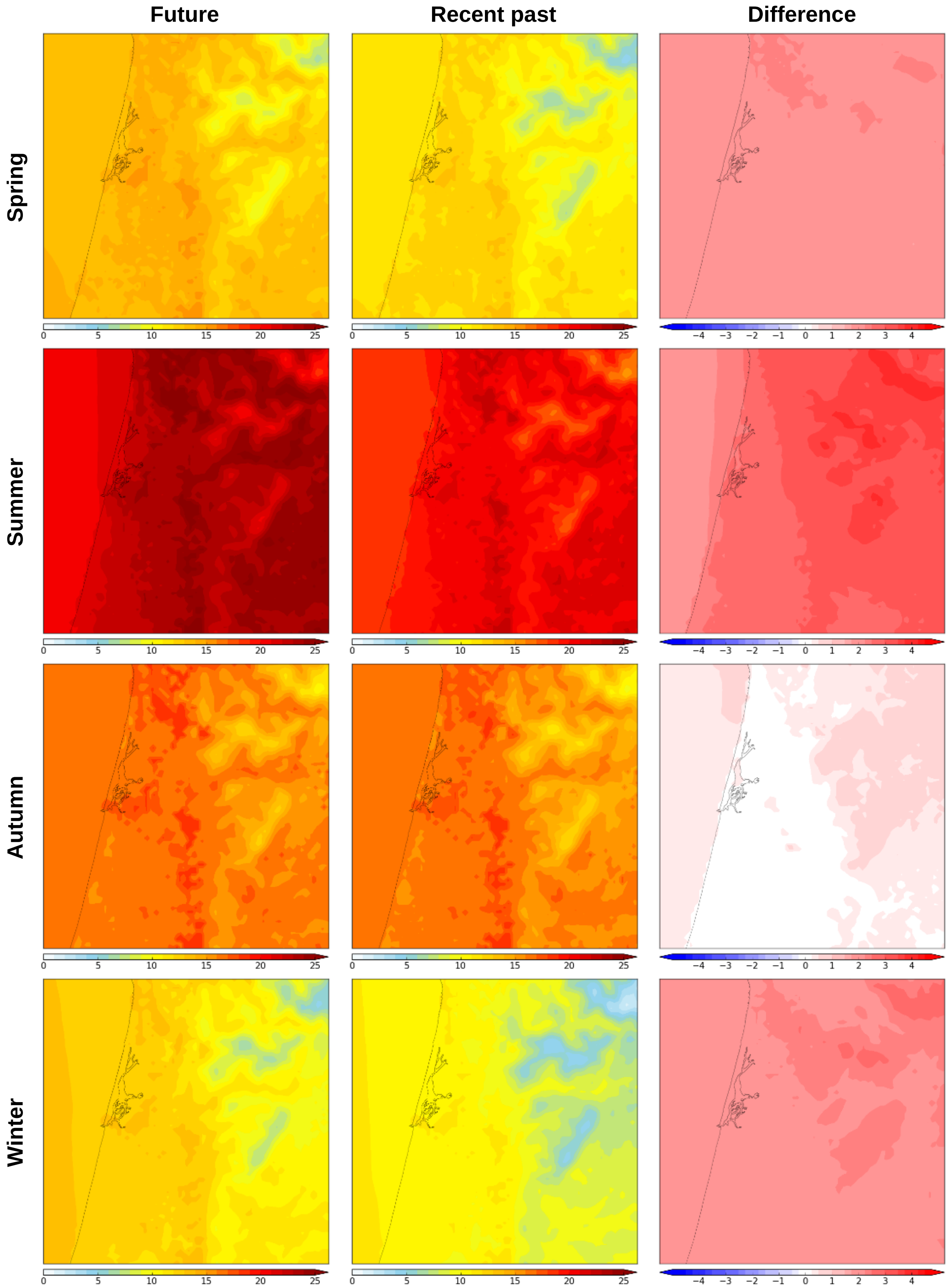

Figure 4.7. *Seasonal average of mean temperature, in °C, for the medium-term future climate (2055), recent past climate (2014), and the difference between them (2055 – 2014).*

Figure 4.8 shows the seasonal total precipitation, for the medium-term future climate, recent past climate, and the difference between them.

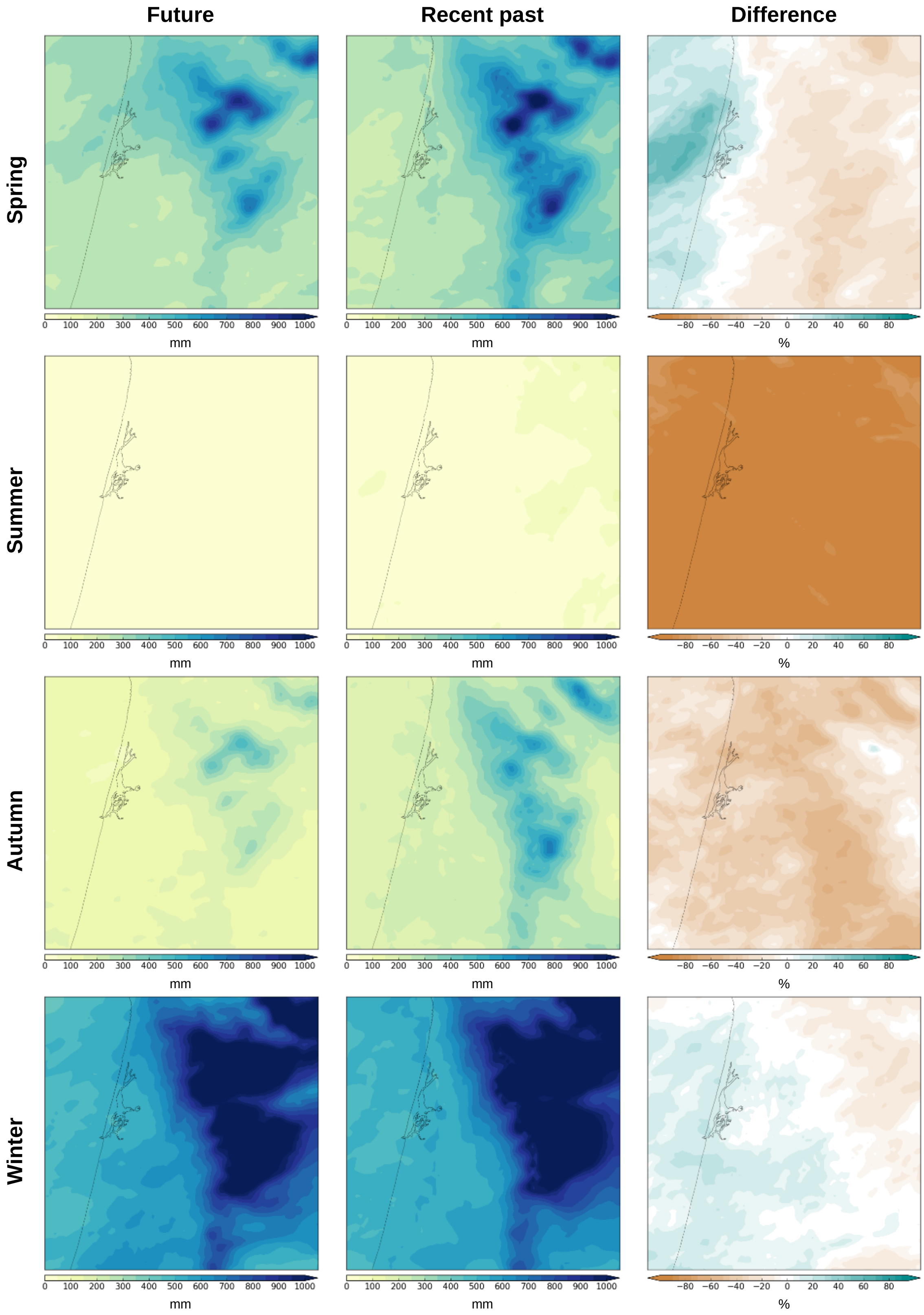

Figure 4.8. *Seasonal total precipitation, for the medium-term future climate (2055), recent past climate (2014), in mm, and the difference between them (2055 – 2014), in %.*

Figure 4.9 shows seasonal average concentrations of NO_2, for the medium-term future climate, recent past climate, and the difference between them.

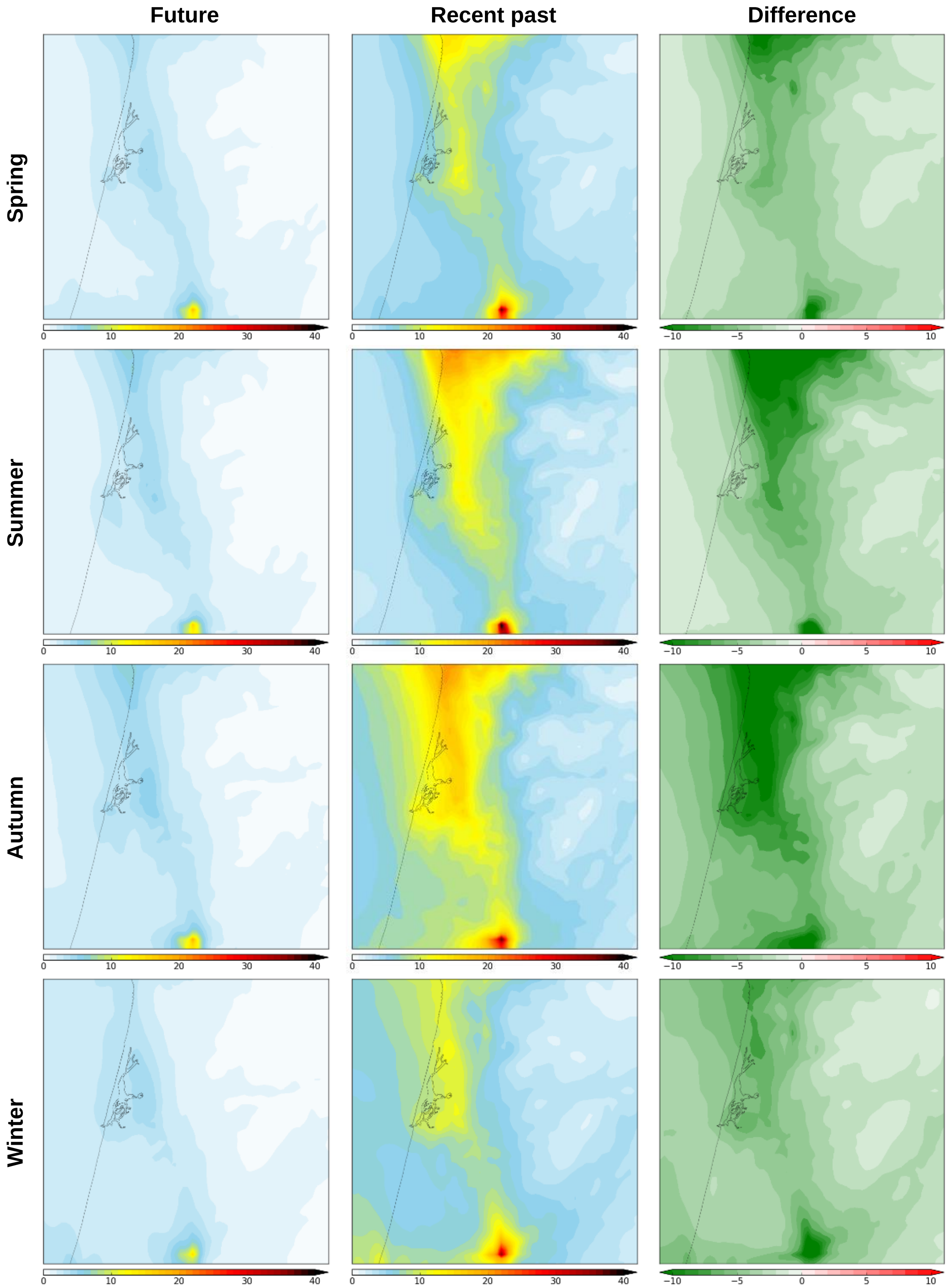

Figure 4.9. *Seasonal average concentrations of NO_2, in µg.m^{-3}, for the medium-term future climate (2055), recent past climate (2014), and the difference between them (2055 – 2014). For future and recent past maps, the maximum value of the colour scale represents the EU limit value (40 µg.m^{-3}).*

Figure 4.10 shows seasonal average concentrations of the maximum daily 8 h mean O_3, for the medium-term future climate, recent past climate, and the difference between them.

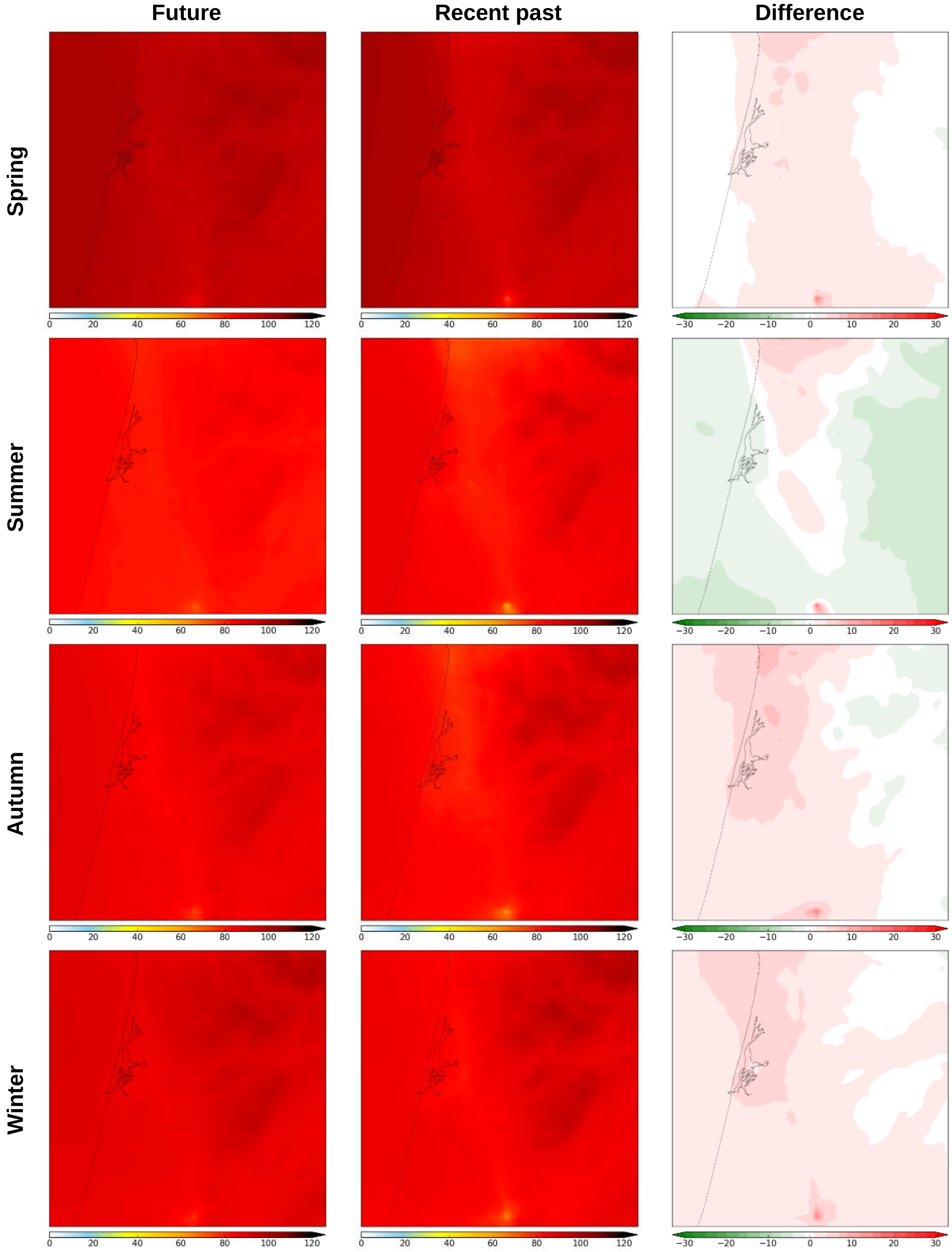

Figure 4.10. *Seasonal average concentrations of the maximum daily 8 h mean* O_3*, in* $\mu g.m^{-3}$*, for the medium-term future climate (2055), recent past climate (2014), and the difference between them (2055 – 2014). For future and recent past maps, the maximum value of the colour scale represents the EU target value (120* $\mu g.m^{-3}$*).*

Figure 4.11 shows seasonal average concentrations of PM10, for the medium-term future climate, recent past climate, and the difference between them.

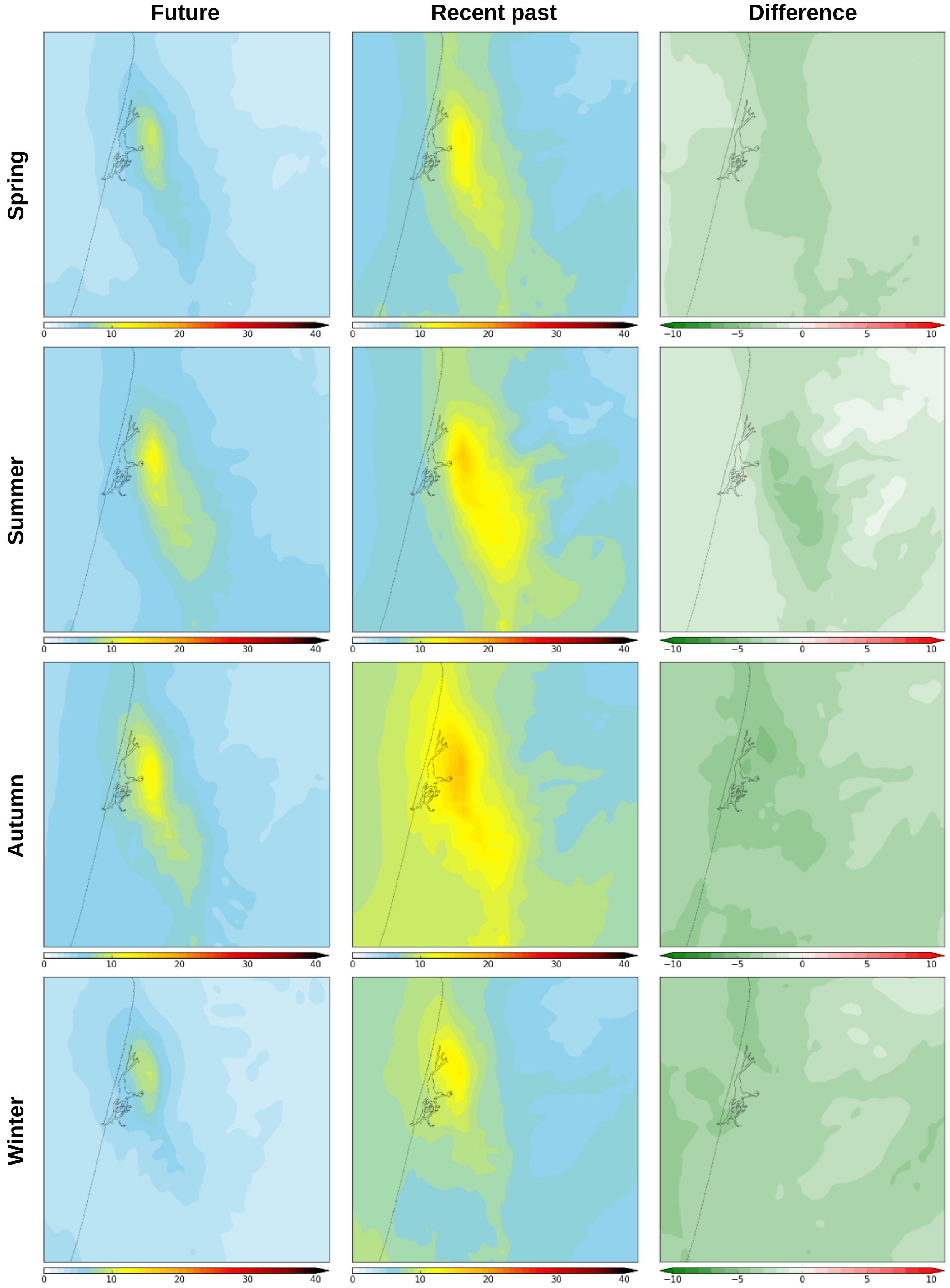

Figure 4.11. *Seasonal average concentrations of PM10, in µg.m^{-3}, for the medium-term future climate (2055), recent past climate (2014), and the difference between them (2055 – 2014). For future and recent past maps, the maximum value of the colour scale represents the EU limit value (40 µg.m^{-3}).*

Figure 4.12 shows seasonal average concentrations of PM2.5, for the medium-term future climate, recent past climate, and the difference between them.

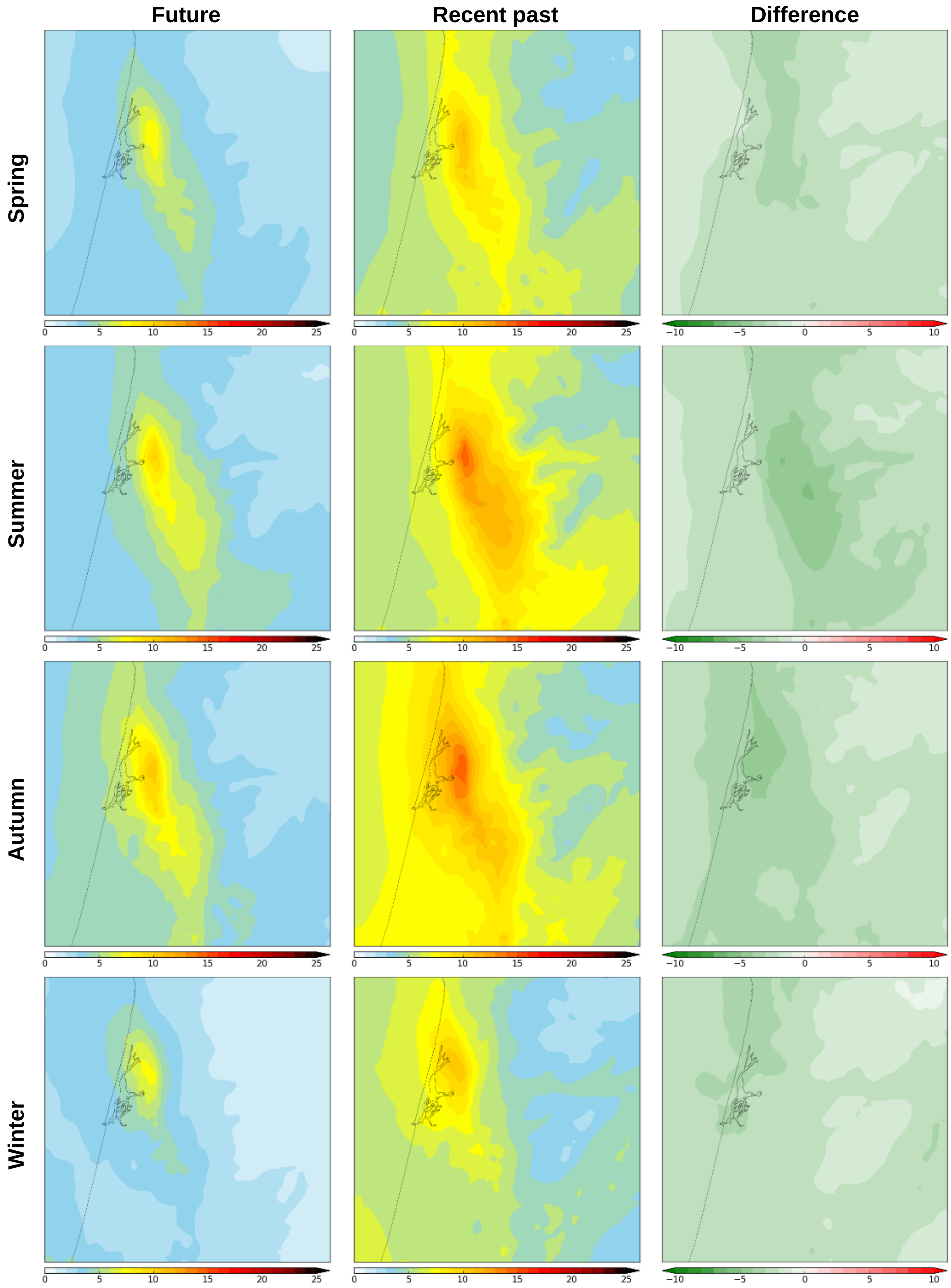

Figure 4.12. *Seasonal average concentrations of PM2.5, in µg.m^{-3}, for the medium-term future climate (2055), recent past climate (2014), and the difference between them (2055 – 2014). For future and recent past maps, the maximum value of the colour scale represents the EU limit value (25 µg.m^{-3}).*

To analyse the quantitative CAMx performance, the following statistical parameters were used: (i) correlation coefficient; (ii) mean bias; (iii) mean error; (iv) index of agreement; (v) root mean square error (RMSE); and (vi) modelling quality indicator (MQI), considering a deviation between modelled and measured concentrations as twice the measurement uncertainty, as defined by Janssen and Thunis (Janssen and Thunis 2022). Table 4.3 summarize the values of the CAMx performance statistics, for NO_2, O_3, and PM10.

Table 4.3. *CAMx performance statistics for NO_2 hourly concentrations, O_3 8 hour mean concentrations, and PM10 daily mean concentrations, computed for the recent past climate (2014), for three air quality monitoring stations (EST – Estarreja, industrial background; ILH – Ílhavo, suburban background; and FRN – Fornelo do Monte, rural background).*

	NO_2			**O_3**			**PM10**		
	EST	**ILH**	**FRN**	**EST**	**ILH**	**FRN**	**EST**	**ILH**	**FRN**
Nrº Observations [%]	100	97	100	344	356	365	100	99	100
Correlation [-]	0.39	0.32	0.68	0.52	0.50	0.54	0.33	0.54	0.52
Mean Bias [µg.m^{-3}]	-1.56	-0.26	-0.72	1.72	2.50	3.22	-6.79	-1.10	-5.65
Mean Error [µg.m^{-3}]	5.67	0.99	3.76	15.95	12.87	13.93	10.67	3.53	7.10
Index of Agreement [-]	0.62	0.64	0.89	0.70	0.68	0.68	0.73	0.64	0.48
RMSE [µg.m^{-3}]	7.31	1.27	5.24	20.43	16.86	17.36	14.72	5.57	9.18
MQI [-]	0.35	0.07	0.26	0.26	0.21	0.22	0.90	0.93	0.86

For the three air quality stations, mean bias for NO_2, O_3 and PM10 range from -1.56 to -0.26 µg.m^{-3}, from 1.72 to 3.22 µg.m^{-3}, and from -6.79 to -1.10 µg.m^{-3}, respectively, showing a general underestimation of NO_2 and PM10, and the overestimation of O_3. Higher RMSE are found for O_3, with values up to 15.95 µg.m^{-3} and 20.43 µg.m^{-3}, respectively. Correlation and index of agreement for the three pollutants cover a range from 0.32 to 0.68. For the modelling quality indicator, values below 1 are obtained for all air quality monitoring stations and pollutants. According to the FAIRMODE guidelines (Janssen and Thunis 2022), 90th percentile MQI values below 1 indicate that the modelling quality objective, defined as the minimum level of quality to be achieved by a model for policy use, is fulfilled.

The CAMx performance can be attributed to different factors. Some studies (Emery et al. 2011; Pepe et al. 2016; Coelho et al. 2022b) stated that NO_2 and PM10 underestimations could be explained by a potential overestimation of the vertical mixing. Additionally, a potential underestimation/ overestimation in the emission inventory database could lead to the CAMx underestimation/ overestimation (Oikonomakis et al. 2018; Coelho et al. 2022b). Further, the CAMx performance could also be affected by the WRF meteorological fields. As one of the objectives of this work is to compare future climate results with those of the recent past, the same methodology had to be adopted for both periods. Thus, the WRF simulation was forced by the climate model MPI-ESM and not with data from observations/reanalysis, which may affect the WRF performance, and consequently the CAMx results. Nonetheless, other studies (Russo et al.; Marta-Almeida et al. 2016; Rafael et al. 2020) evaluated the use of MPI-ESM data as WRF forcing, obtaining a high level of confidence in this model configuration for climate studies.

CHAPTER 5

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5. FACING THE CHALLENGES OF AIR QUALITY AND HEALTH IN A FUTURE CLIMATE: THE AVEIRO REGION CASE STUDY

Abstract

Air pollution and climate change are the most important environmental issues for European citizens. Despite the air quality improvements achieved in recent years, with most pollutants' concentrations below the European Union legislated values, it is necessary to understand whether this will continue in the future due to expected climate changes impacts. In this context, this work tries to answer two main questions: (i) What is the relative contribution of emission source regions/activities to air quality, now and in the future, considering a climate change scenario?; and (ii) What additional policies are needed to support win-win strategies for air quality and climate mitigation and/or adaptation, at urban scale? For that, a climate and air quality modelling system, with source apportionment tools, was applied to the Aveiro Region, in Portugal. Main results show that in the future, due to the implementation of carbon neutrality measures, air quality in the Aveiro Region may improve, with reduction up to 4 $\mu g.m^{-3}$ for particulate matter (PM) concentrations and 22 $\mu g.m^{-3}$ for nitrogen dioxide (NO_2), and consequently, the premature deaths due to air pollution exposure will also decrease. The expected air quality improvement will ensure that, in the future, the limit values of the European Union (EU) Air Quality Directive will not be exceeded, but the same will not happen if the proposed revision of the EU Air Quality Directive is approved. Results also shown that, in the future, industrial sector will be the one with higher relative contribution for PM concentrations and the second one for NO_2. For that sector, additional emission abatement measures were tested, showing that, in the future, it is possible to comply with all the new limit values proposed by the EU.

Keywords: urban areas; source apportionment; emission abatement strategies; numerical modelling

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5.1. INTRODUCTION

In the last years, air pollution has been considered as the biggest environmental risk factor, threatening human health and climate (WHO 2022a). Densely populated urban areas are facing air pollution problems, exposing their inhabitants to air pollutants such as particulate matter (PM) and nitrogen dioxide (NO_2), increasing morbidity and mortality, thus affecting population's health. According to the World Health Organization (WHO), air pollution (outdoor and indoor) is responsible for around 7 million premature deaths annually, across the world (WHO 2016).

Air quality management plans for urban areas, which include emission abatement strategies, have been used as a quick solution to air pollution problems worldwide (Morawska et al. 2021; Goyal et al. 2022). The air quality plans, applied from European (e.g., Miranda et al. 2015), to regional (e.g., Borrego et al. 2016; Relvas et al. 2022), and urban (e.g., Duque et al. 2016; Silveira et al. 2023) areas, have successfully improved the air quality over the years. Although, air pollution hotspots remain in some regions, like Po-valley and Eastern Europe due to PM, and in most European urban areas due to NO_2 (Thunis et al. 2019; EEA 2021).

Miranda et al. (2015) and Williams et al. (2016) have reported the success of the emission abatement strategies, highlighting the importance of key components in the entire decision-making process, such as the need of comprehensive case study assessments, stakeholder participation and consultation procedures, constant improvement in the defined measures and tools for cost benefit analysis. Other studies (e.g., Thunis et al. 2019; Coelho et al. 2022a, b) have also identified the understanding of pollution sources as one of the key issues in defining air quality improvement strategies. Additionally, changes in weather patterns due to climate change should also be considered as they play an important role in the atmospheric processes responsible for the transport, chemical transformation, and deposition of air pollutants, leading also to air quality changes (Jacob and Winner 2009; Sá et al. 2016; Coelho et al. 2022c).

Several studies regarding emission abatement strategies have already been done, however some research gaps remain unfulfilled. For example, East et al. (2021) applied a modelling approach able to identify the main sources of air pollution in the Bogotá metropolitan area, and examined the potential impacts of emissions policies targeting these sources. Similarly, Bai et al. (2021) examined the contributions of different sectors to several key criterion air pollutants across Shanxi province of China, and performed sensitivity analysis to quantify the mitigation potentials for each sector emissions in the city. However, both studies were only applied for past years, without considering the expected climate change impacts on the future air quality. Contrary, other studies, such as San José et al. (2016) for Madrid, Antwerp, Milan, Helsinki and London, and Monteiro et al. (2018) for Portugal, studied the impacts of the global climate change on urban climate and air quality, considering different climate change scenarios, but without providing a full understanding of the main pollution sources or defining future emission abatement strategies for air quality management. Despite this important knowledge gaining, some research gaps remain unfilled, namely the integration of the climate change impacts in the methodology applied to support air quality management strategies.

In Portugal, aligned with the Directive 2001/81/EC (EC 2001) on national emission ceilings (NEC), the Portuguese Government has defined its strategies and commitments aimed to further reduce air pollutants emissions until 2030. The NEC Directive requires Member States to develop National Air Pollution Control Programmes (NAPCP), including the evaluation of compliance not only of emission ceilings, but also of air quality objectives, and their impacts on health. Nonetheless, the guidance for designing emission reduction strategies for the NAPCP neglects the influence of climate change (EC 2019). On the other hand, to tackle climate and energy challenges, three possible carbon neutrality trajectories, with distinct implications on air pollutant emissions, were defined in the Portuguese Roadmap for Carbon Neutrality in 2050 (RNC2050), in which Portugal is committed to reduce its greenhouse gases emissions so that the balance between emissions and removals from the atmosphere will be zero by 2050 (Monjardino et al. 2019, 2021). However, RNC2050 was defined based only on climate targets, ignoring how changes in pollutant emissions could impact air quality. Thus, there is a lack of integration of emission control needs and climate change mitigation and adaptation strategies towards the improvement of air quality and the protection of human health for the future.

In this context, there are two questions that needs to be explored to fulfil the research gaps previously identified, namely: (i) What is the relative contribution of emission source regions/activities to air quality, now and in the future, considering a climate change scenario?; and (ii) What additional policies are needed to support win-win strategies for air quality and climate mitigation and/or adaptation, at urban scale? Therefore, the main goal of this study is to apply a modelling approach able to assess the impacts of future climate and projected emissions, including the relative contribution of different source regions and sectors, on air quality at urban scale, as well as their impact on human health. To achieve the proposed goal and answer the two research questions, an air quality modelling system, with source apportionment tools, was applied to the Aveiro Region, considering a climate change scenario.

This work will contribute to enrich the knowledge in the air quality and climate change fields, identified by the European citizens as the two most important environmental issues (EC 2020). This research is pertinent, complex and multidisciplinary and it is in accordance with the Horizon Europe Cluster 5 - Climate, Energy and Mobility (EC 2021). The objectives and the approach defined are also aligned with two goals of the 2030 Agenda for Sustainable Development (UN 2015), namely: Goal 11 - make cities and human settlements inclusive, safe, resilient and sustainable; and Goal 13 - take urgent action to combat climate change and its impacts. This work will overcome the existing gaps in this scientific domain and support decision makers to define the best win-win strategies to reduce climate change impacts on air quality.

5.2. CASE STUDY CHARACTERIZATION

Aveiro Region, the selected case study, is a multi-urban area located in the central coastal area of Portugal (see Figure 5.1), with a great diversity of geography, economy, activity sectors, and population density (Rodrigues et al. 2021; Coelho et al. 2022a). The Aveiro Region includes 11 municipalities, namely Águeda, Albergaria-a-Velha, Anadia, Aveiro, Estarreja, Ílhavo, Murtosa, Oliveira do Bairro, Ovar, Sever do Vouga and Vagos, and covers a total area of 1693 km^2. All municipalities are influenced by the presence of Ria de Aveiro, a lagoon with more than six thousand hectares (Coelho et al. 2022a).

Regarding its demography, the Aveiro Region accounts for 367 455 inhabitants (INE 2021), which represents about 3.5% of the national population. The number of inhabitants per municipality varies significantly. For example, Sever do Vouga has a population density below 100 inhabitants.km^{-2} (considerably lower than the national level of 112 inhabitants.km^{-2}), while Ílhavo exceeds 500 inhabitants.km^{-2}. Figure 5.1 shows the spatial distribution of the population density in the Aveiro Region, highlighting Ílhavo, Aveiro, and Ovar municipalities as those with the highest population density.

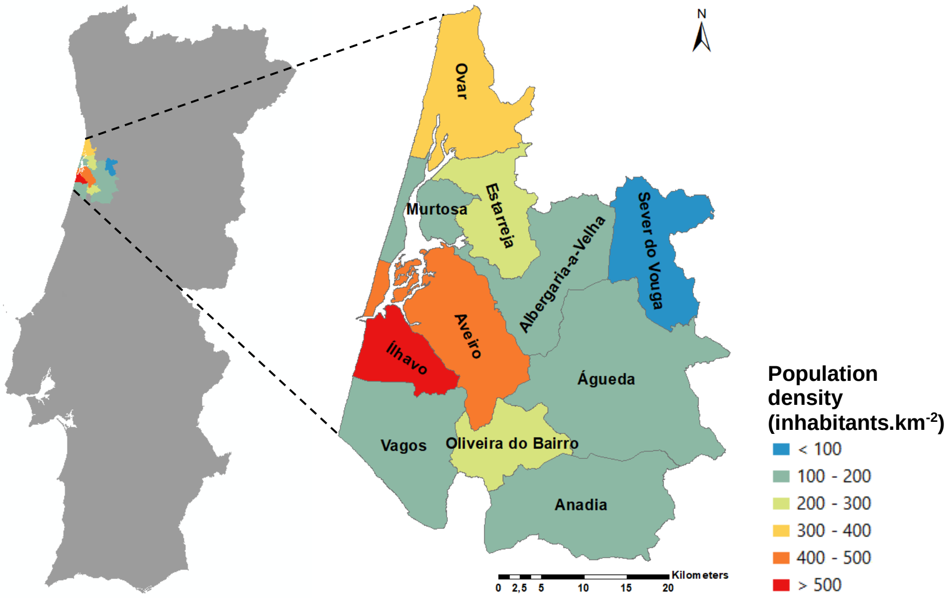

Figure 5.1. *Location of the Aveiro Region in Continental Portugal (right) and distribution of the municipalities that comprise it (left).*

Rural municipalities, where agriculture plays an important role, are mainly situated inland and are the ones with lower population density (Slingerland et al. 2018). On the other hand, urban and industrial municipalities (with higher population density) are placed along the coast, closer to one of the most important seaports of Portugal, for both fishing and commercial sectors (Coelho et al. 2018; Slingerland et al. 2018).

Manufacturing industry, accounting for 50% of the region’s turnover, is the predominant economic sector of the region (Coelho et al. 2018). Several chemical and manufacturing industries are located in the region, such as chemical facilities mainly located in Estarreja municipality, and one of the major Portuguese pulp and paper producers (with a turnover of ~1% of the national gross domestic production - GDP), located in Aveiro municipality (Coelho et al. 2018).

According to the Iberian Climatic Atlas (AEMET-IM 2011), the Aveiro Region territory has a temperate climate with dry or temperate summers, being classified as CSB - warm-summer Mediterranean climate, according to the Köppen-Geiger scale (Peel et al. 2007). The orography of the region is characterized by low altitudes along the coast (< 100 m), increasing towards inland, with the maximum altitude of 841 m in the Sever do Vouga municipality (Coelho et al. 2018). The Aveiro Region's climate, associated with the open landscape characteristic of most of its territory, favours the pollutants dispersion. However, PM and NO_2 exceedances to the legislated values still occur in some areas of the region (Rodrigues et al. 2021; Coelho et al. 2022a).

5.3. MODELLING FRAMEWORK

In this section, the methodology followed to assess the future climate and air quality over Aveiro Region is presented (section 5.3.1), as well as the future health impacts (section 5.3.2).

5.3.1. Medium-term Future Climate and Air Quality

A modelling system, which includes the Weather Research and Forecasting (WRF) model (Skamarock et al. 2019) and the Comprehensive Air Quality Model with Extensions (CAMx) (ENVIRON 2020), was applied to the Aveiro Region, using a three-nested approach based on a coarser domain with 30 km horizontal resolution and the domain of interest centred in the Aveiro Region, with 75 x 75 horizontal grid cells, at 1.2 km of horizontal resolution (see Figure 5.2). Both numerical models have been applied to case studies all over the world, and more specifically in Portugal, with reliable and realistic results (Rafael et al. 2017; Ferreira et al. 2020; Relvas et al. 2022; Coelho et al. 2022a, c, 2023).

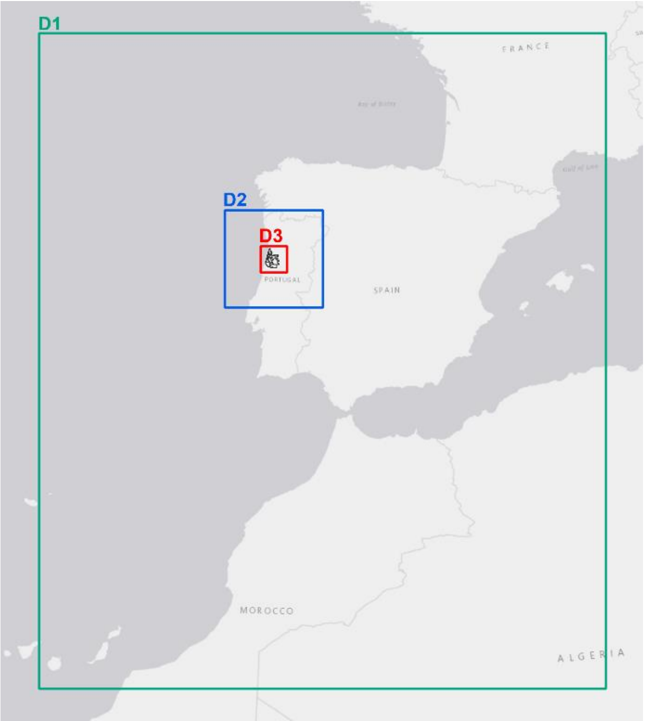

Figure 5.2. *Model domains with horizontal resolutions of 30 (D1), 6 (D2), and 1.2 (D3) km.*

The WRF model, forced by the Max Planck Institute for Meteorology Earth System Model version 1.2 (MPI-ESM1.2-HR) (Mauritsen et al. 2019), was used to downscale global climate simulations for the three defined domains. The WRF physical configuration is detailed in Coelho et al. (2022c).

Two simulation periods were considered, 2014 and 2055, the first statistically representative of the recent-past and used as reference, and the second statistically representative of the medium-term future climate. The selection of the statistically representative years, fully described in Coelho et al. (2022c), was supported by a climatological analysis of temperature, precipitation, wind direction, and solar radiation data provided by coarser simulations, for the periods 1995–2014 (for the recent past) and 2046–2065 (for the medium-term future). For each year of the periods 1995–2014 and 2046–2065, the selected variables were analysed and compared with the average of the period, to find the year with the lowest climate anomaly. For the medium-term future climate, the intermediate Shared Socio-Economic Pathway with a nominal 4.5W·m^{-2} radiative forcing level by 2100 (SSP2-4.5; Fricko et al. 2017) was applied, considering medium challenges for both climate change adaptation and mitigation and a medium pollution control scenario (Riahi et al. 2017).

WRF outputs were used as meteorological inputs to the CAMx model. CAMx initial and boundary conditions for the first domain were provided by the global chemical model CAM-Chem (Emmons et al. 2020), with 0.9° x 1.25° spatial resolution, every 6 hours. For the recent-past simulation, anthropogenic emissions from the European Monitoring and Evaluation Programme (EMEP) emission inventory (EMEP 2017) were spatially disaggregated by proxies, following previously defined and applied methodologies (Morán et al. 2016; Silveira et al. 2017; Ferreira et al. 2020), and speciated into the Carbon Bond 6 chemical mechanism (CB6) chemical mechanism species considered in CAMx (Yarwood et al. 2010). For the future climate simulation, anthropogenic emissions were estimated according to Sá et al. (2015) methodology, and, as explained in Coelho et al. (2022c), considering the emission projections of the RNC2050 intermediate scenario (Monjardino et al. 2019, 2021), which are in line with SSP2-4.5 scenario considered in WRF simulation. Thus, medium-term future climate emissions will have an average reduction of 70% for CO (carbon monoxide), 68% for NOx (nitrogen oxides), 38% for PM2.5 (particulate matter with a diameter of 2.5 microns or less), 33% for PM10 (particulate matter with a diameter of 10 microns or less), 36% for SOx (sulphur oxides), 27% for NMVOC (non-methane volatile organic compounds), and 15% for NH_3 (ammonia), relative to the recent-past. Biogenic emissions were obtained from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al. 2012).

The main differences between the two simulations are summarized in Table 5.1.

Table 5.1. *Differences between the two simulations.*

	Recent-past	**Future**
Simulation period	2014	2055
Climate change scenario	None	SSP2-4.5
Anthropogenic emissions	EMEP	RNC2050 projections
Biogenic emissions	MEGAN (for 2014)	MEGAN (for 2055)

For both simulation periods, the CAMx Particulate Source Apportionment Technology (PSAT) (Yarwood et al. 2007) was applied to quantify the contributions of multiple source areas and sectors to ambient pollution of the innermost domain. The PSAT tool requires the definition of source areas and sectors to be tracked in some selected receptor points. The definition of the source sectors was based on the national emission inventory (Pereira et al. 2018) and previous works applied for Aveiro Region (Coelho et al. 2022a, b). The emission sectors (Selected Nomenclature for Air Pollution – SNAP - categories) identified as the most relevant for the Aveiro Region were the following: (i) residential and commercial combustion (SNAP2); (ii) industrial combustion and processes (SNAP3&4), (iii) road transport (SNAP7); and (iv) other mobile sources (SNAP8). All the remaining sectors (energy production, extraction and distribution of fossil fuels, solvents use, waste treatment and agriculture) were grouped and treated as one (OTH). Biogenic emissions were also considered as a sector (BIOGENIC). For the source areas definition, 12 distinct areas were defined, one for each of the eleven municipalities of the Aveiro Region, and an additional source area comprising the entire surrounding area (OUT), as shown in Figure 5.3. As air quality is a big concern in urban areas, where there is more human exposure to air pollution, 7 receptor points were defined in consolidated urban areas with higher population density (see Figure 5.3).

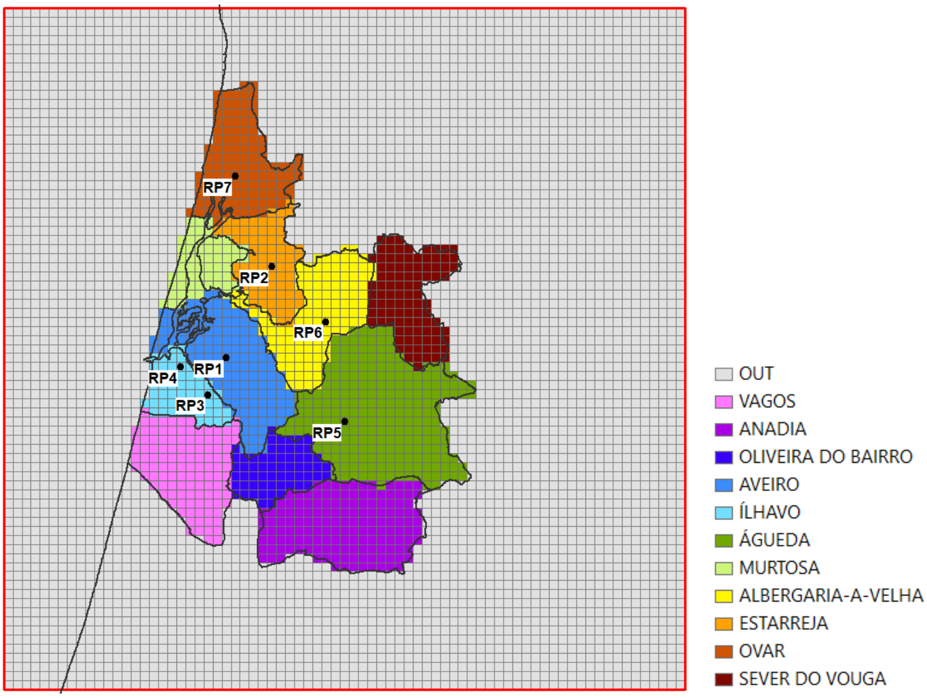

Figure 5.3. *PSAT receptor points (RP) and source area map for the innermost domain (D3).*

5.3.2. Health Impact Assessment

Long-term health impacts of PM2.5 and NO_2 exposures were estimated, following the methodology applied by the European Environment Agency (EEA) in their annual health assessments (Soares et al. 2019), and considering the CAMx concentrations by grid cell and pollutant, together with population data stratified by age and sex (INE 2021). According to the EEA (2018), PM2.5 and NO_2 are the pollutants with the most serious health impacts and with the most robust scientific evidence. For both PM2.5 and NO_2, premature deaths in ages above 30 years for all cause (natural) mortality were considered. The concentration-response functions (CRFs) methodologies from Chen and Hoek (2020) for PM2.5 and Huangfu and Atkinson (2020) for NO_2, were used, according to the up to date recommendations of WHO air quality guidelines (WHO 2021a). Detailed information on the health impact assessment can be found in Coelho et al. (2022c). Table 5.2 presents detailed information on the CRFs, relative risk (RR) and 95% confidence interval (CI), range of concentration and source of health data used.

Table 5.2. *CRF used according to the WHO (2021a) recommendations.*

Health outcome	Pollutant metric	Baseline concentration (C_0)	RR (95% CI) per 10 $\mu g.m^{-3}$	Source of CRF	Source of health data
Mortality, all-cause (natural), age 30+ years	PM2.5, annual mean	> 5 $\mu g.m^{-3}$	1.08 [1.06; 1.09]	Chen and Hoek (2020)	European mortality database, ICD-10, codes A–R (WHO 2022b)
	NO_2, annual mean	> 10 $\mu g.m^{-3}$	1.02 [1.01; 1.04]	Huangfu and Atkinson (2020)	

5.4. RESULTS AND DISCUSSION

In this section, the obtained results for the future air quality (section 5.4.1), and for the health impact assessment (section 5.4.2) are analysed and discussed.

5.4.1. Medium-term Future Air Quality

The PM2.5, PM10 and NO_2 concentration fields, obtained from the WRF-CAMx modelling system, are shown in Figure 4. The analysis focuses on the annual averages since these are the indicators used for the health impact assessment of the long-term exposure to air pollution. In both medium-term future and recent-past climates, the results show similar spatial patterns for the three air pollutants analysed, with the air pollutants dispersion driven by the prevailing northwest winds.

The accuracy of the presented results was assessed in Coelho et al. (2022c), where the CAMx performance was evaluated through the application of performance statistics. According to Coelho et al. (2022c), the CAMx performance fulfils the modelling quality objective of the FAIRMODE guidelines (Janssen and Thunis 2022), defined as the minimum level of quality to be achieved by a model for policy use.

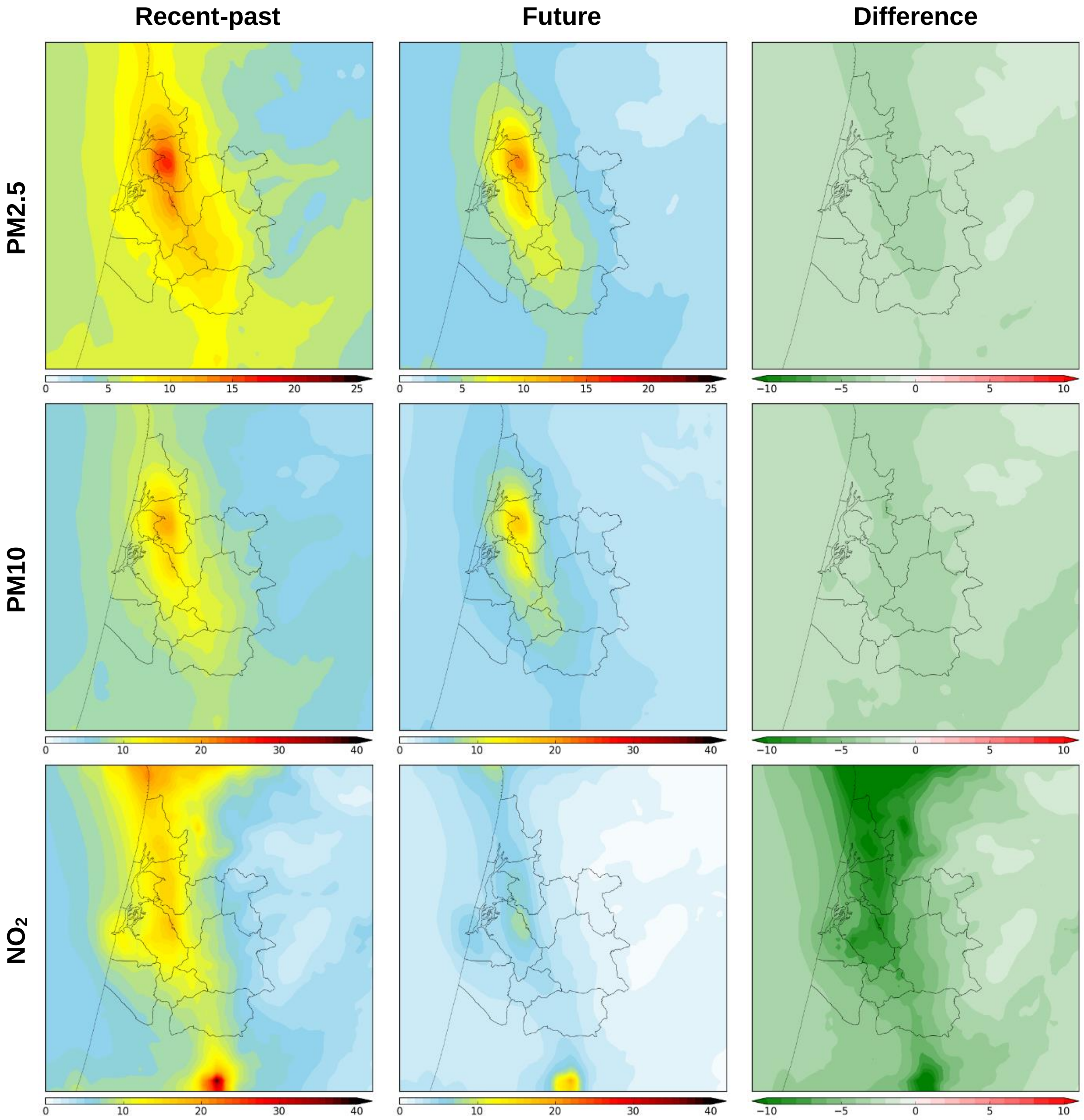

Figure 5.4. *Annual average concentration of PM2.5, PM10 and NO_2, in μg.m^{-3}, for the medium-term future climate (2055), recent-past climate (2014) and the difference between them (2055 – 2014). For future and recent-past maps, the maximum value of the colour scale represents the EU limit value.*

According to Figure 5.4, and as already stated in Coelho et al. (2022c), it is expected that air quality will improve at the middle of the century, in Aveiro Region. Despite the climate changes that will affect the region, with an increase in dry spells due to increased temperature and decreased precipitation (see Coelho et al. (2022c) for more details), the concentrations of the main air pollutants will decrease. For both PM10 and PM2.5, higher concentrations were obtained in Estarreja and Aveiro municipalities, next to the most industrialized areas of Aveiro Region. The differences between medium-term future climate and recent-past climate are between -1 and -4 μg.m^{-3}. Regarding NO_2, differences between medium-term future and recent-past climates range from -1 to -22 μg.m^{-3}. The biggest differences were obtained in the south of the domain, outside the Aveiro Region, where one of the largest cement industries in Portugal is located, and in the north of the domain, where the second largest metropolitan area in Portugal is (Porto metropolitan area), and that influences the Aveiro Region air pollution due to the pollutants dispersion by the north/northwest predominant winds.

The expected reduction in PM2.5, PM10 and NO_2 annual concentrations will ensure that, in the medium-term future, there will be no exceedances to the limit values of the European Union (EU) Air Quality Directive (EC 2008). However, in October 2022, the EC proposed a revision of the EU Ambient Air Quality Directive (EC 2022). If the new directive is approved in its current proposal form, in some areas of the Aveiro Region, the future PM2.5 annual concentration will be above the new limit value of 10 $\mu g \cdot m^{-3}$.

To better understand the origin of the air pollutants' levels, the contribution of each source sector, for PM2.5, PM10 and NO_2 annual average concentrations, for both the medium-term future and recent-past climates, in the seven defined receptor points, was analysed (Figure 5.5).

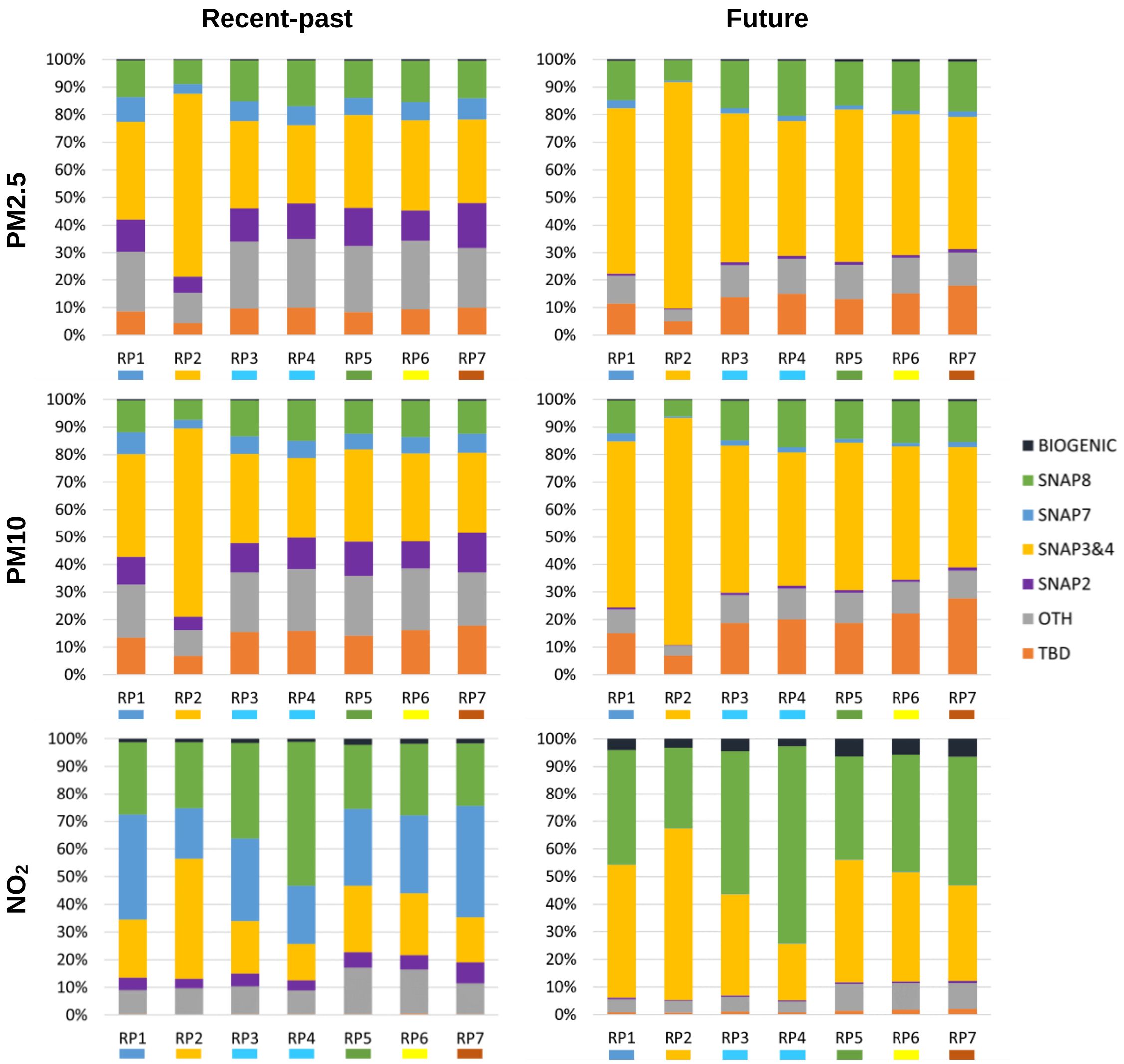

Figure 5.5. *Annual relative contribution of each source sector for PM2.5, PM10 and NO_2 concentrations, for medium-term future climate (2055) and recent-past climate (2014), for each receptor point (RP); (TBD-transboundary transport, SNAP2 - residential and commercial combustion, SNAP3&4 - industrial combustion and processes, SNAP7 - road transport, SNAP8 - other mobile sources, OTH - all the remaining anthropogenic sources, and BIOGENIC – biogenic emissions). Note that the colours below each receptor point correspond to the municipality where it is located, according to the colour scheme presented in Figure 5.3.*

From Figure 5.5, for the recent-past climate, it is clear that SNAP3&4 is the main sector contributing to PM concentration (around 41%). This contribution is even more pronounced in receptor point number 2 (RP2), that is located near the main industrial facilities of Aveiro Region, in Estarreja municipality, where the higher PM concentration are obtained. The industrial sector had already been identified as the one that most contributes to air pollution in the Aveiro Region, not only in other studies (e.g., Coelho et al. 2022a) but also in citizen and stakeholder participation processes (e.g., Coelho et al. 2018). For PM, SNAP8, i.e., aviation, maritime, and off-road transport, also play an important role in the region, with a contribution range from 9% (at RP2) to 17% (at RP4, the receptor point closest to the seaport), for the recent-past climate. Also, for the recent-past, SNAP2 and SNAP7 only account for around 10% and 6% of the PM annual concentrations, respectively. Transboundary transport (TBD), with an average contribution of 13% in the recent-past, highlights the importance of long-range transport of PM10, in agreement with other studies (Garg and Sinha 2017; Coelho et al. 2022a). Regarding NO_2, for the recent-past, the sector with the highest contribution is the SNAP8, with an average of 30%, followed by SNAP7 with 28%, and SNAP3&4 with 24%. SNAP2 represents around 5% of the NO_2 annual concentrations. TBD only accounts for 1% of the NO_2 annual concentration, highlighting the importance of local sources for NO_2 concentrations (Degraeuwe et al. 2021; Coelho et al. 2022a). All the remaining sectors, defined as OTH (i.e., energy production, extraction and distribution of fossil fuels, solvents use, waste treatment and agriculture), account around 20%, 18% and 11% of the PM2.5, PM10 and NO_2 annual concentrations, respectively. For all the pollutants analysed, BIOGENIC only accounts for 1% of de annual concentration in the recent-past period.

For each pollutant, the contributions of each sector differ throughout the year (see supplementary material). For PM2.5 and PM10, SNAP2 is the one that presents the largest variations, with maximum contributions in the cold months, from November to February, and minimum contributions in the remaining months. For PM10, short-lived episodes, usually lasting a few days, where TBD contributions significantly increase are identified. For NO_2 there are no significant variations in the sectors' contributions throughout the year.

For both PM2.5 and PM10, in the medium-term future, when compared to the recent-past, SNAP3&4 will be the only sector to increase its contribution to around 62%; SNAP8, TBD and BIOGENIC will maintain their contributions; and SNAP2, SNAP7 and OTH will decrease to 1%, 1% and 8%, respectively. For NO_2, the contributions of SNAP8, SNAP3&4 and BIOGENIC will increase in the future to 46%, 42% and 4%, respectively; SNAP2, SNAP7 and OTH will decrease to 0%, 0% and 6%, respectively; and the TBD contribution will be maintained.

Most of the changes in the annual contributions could be justified by the changes in the anthropogenic emissions projected in the RNC2050, that were considered in the medium-future climate simulation. Although the total emissions of each pollutant will decrease, when analysed by sector, it is possible to verify that there is an increase in the emissions of some pollutants in specific sectors, namely: PM2.5, PM10, NMVOC and NH_3 in SNAP3&4; and SOx in SNAP8 (see Table 5.3).

Table 5.3. *Medium-term future emission projections from the RNC2050, expressed in % of the recent-past, for CO, NOx, PM2.5, PM10, SOx, NMVOC, and NH_3, by source sector (SNAP2 - residential and commercial combustion, SNAP3&4 - industrial combustion and processes, SNAP7 - road transport, SNAP8 - other mobile sources, OTH - all the remaining anthropogenic sources), and the total of all sectors.*

	CO	**NOx**	**PM2.5**	**PM10**	**SOx**	**NMVOC**	**NH_3**
All sectors	-69.6	-68.0	-38.2	-33.4	-35.5	-27.1	-15.2
SNAP2	-94.8	-97.4	-96.2	-94.8	-100.0	-96.8	-100.0
SNAP3&4	-60.9	-30.8	+18.8	+17.9	-24.9	+10.8	+13.1
SNAP7	-100.0	-100.0	-65.4	-48.7	-100.0	-99.9	-100.0
SNAP8	-76.7	-34.0	-60.4	-60.3	+16.1	-24.3	-100.0
OTH	-8.4	-79.7	-49.7	-46.2	-94.2	-18.1	-11.4

From the analysis of RNC2050 sectorial report on the estimation of atmospheric pollutants (Monjardino et al. 2019, 2021), shown in Table 5.3, it is possible to state that:

(i) For the total emissions of each pollutant there will be an average reduction between 15% and 70%, in relation to the recent-past.

(ii) For residential and commercial combustion (SNAP2), a significant decrease in the consumption of fossil fuels and biomass is considered by 2050, leading to reductions up to 100% in most pollutants.

(iii) In industry (SNAP3&4), the reduction in fossil fuel consumption will lead to a decrease in CO, NOx and SOx emissions. However, there will be an increase in NMVOC, PM and NH_3 emissions due to increased biomass consumption.

(iv) For road transport (SNAP7), due to the electrification of the sector and the adoption of shared mobility models, there will be a significant reduction in emissions of all air pollutants, especially for NOx and CO. By 2050, emissions of most pollutants are expected to reduce by 100%.

(v) For aviation, maritime, and off-road transport (SNAP8), there will be an increase in the demand. In this sector, a decrease in the emission of all air pollutants is expected, except for SOx, which will increase. Maritime transport will be the least decarbonized, with a high rate of use of marine diesel, but already with some introduction of liquefied natural gas and biofuels.

(vi) For the remaining sectors (OTH), a general decrease in emissions of all air pollutants is expected.

To complement this source-oriented approach and get an overview on how the characteristics of each one of the eleven municipalities of the Aveiro Region influence its air quality, the contribution by source area, for PM2.5, PM10 and NO_2 annual average concentrations, for both the medium-term future and recent-past climates, in the seven defined receptor points, was analysed, as shown in Figure 5.6.

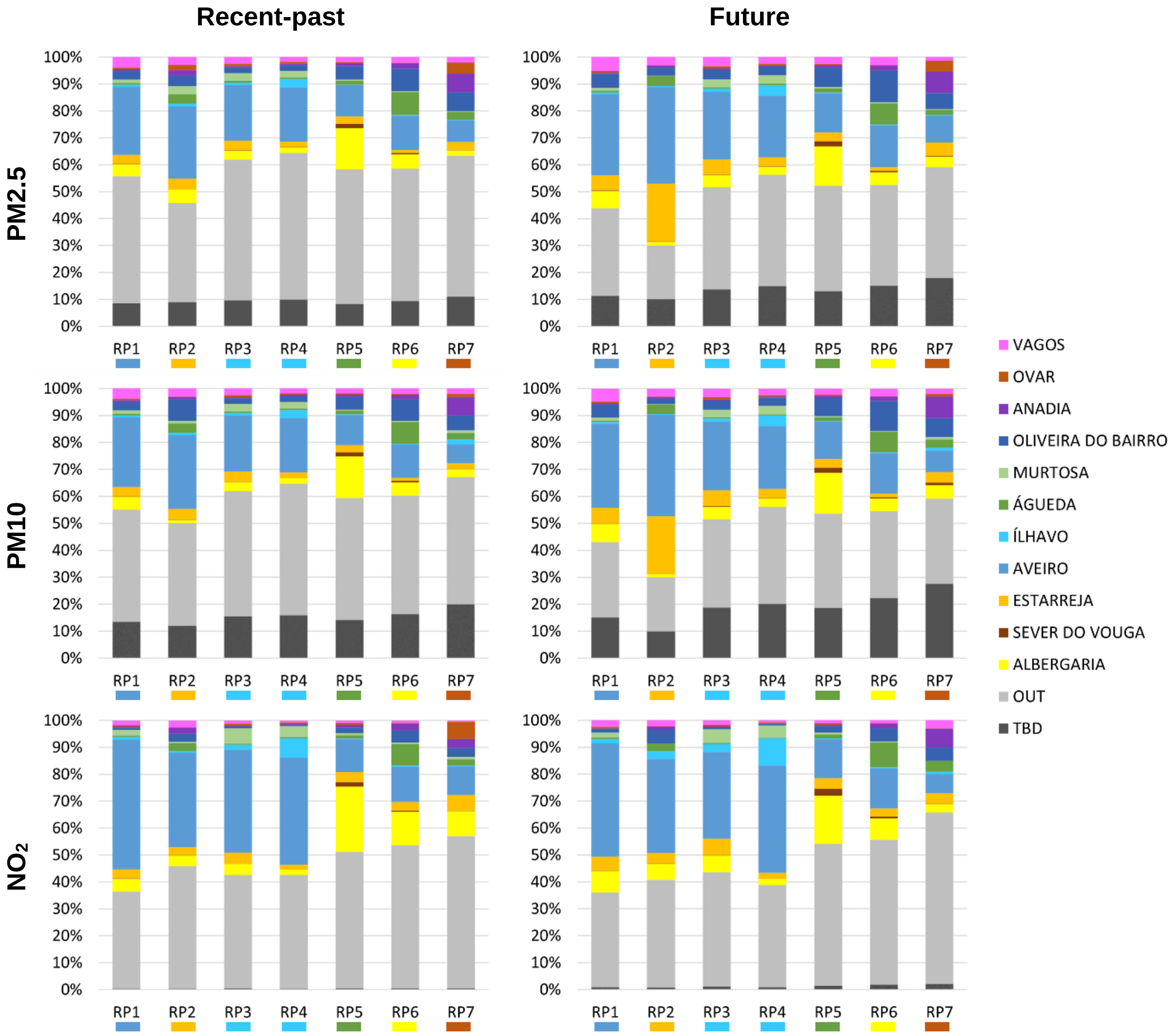

Figure 5.6. Annual relative contribution of each source area for PM2.5, PM10 and NO_2 concentrations, for each receptor point (RP); (TBD- transboundary transport, OUT - Aveiro Region surrounding area, and each one of the eleven municipalities of the Aveiro Region). Note that the colours below each receptor point correspond to the municipality where it is located, according to the colours shown in Figure 5.3.

For both PM2.5 and PM10, Figure 5.6 shows that OUT, i.e., the area inside the innermost domain, but outside the Aveiro Region, is the one that contributes the most for PM concentrations. In average, for the recent-past, OUT contributes 44% for PM2.5 and 38% for PM10 annual concentrations. For the medium-term future, the OUT contribution is expected to decrease, averaging 30% and 26% for PM2.5 and PM10 annual concentrations, respectively. After OUT, the Aveiro municipality area is the one that most contributes to PM annual concentrations, with average contributions of 19% for both PM2.5 and PM10, in the recent-past period. In the medium-term future, these contributions increase to around 25% for both pollutants. As previously mentioned, TBD plays an important role to the PM concentrations, ranking third in this analysis. In general, the remaining areas, i.e., the other 10 municipalities in the Aveiro Region, present less significant contributions (between 1% to 12%), and sometimes only relevant to the PM concentrations recorded in 1 or 2 of the 7 receptor points. For NO_2 concentrations, the analysis is very similar, apart from TBD contributions which are almost zero. For NO_2, OUT contributions continue to be the most significant in all receptor points, even having higher values than those obtained for PM contributions.

The wind fields at the location of each receptor point may play a relevant role in the dispersion and transport of pollutants between the different source areas. The characterization of the wind fields provided by the WRF model for each receptor point, was performed through the representation of wind roses, for both recent-past and medium-term future climate (Figure 5.7).

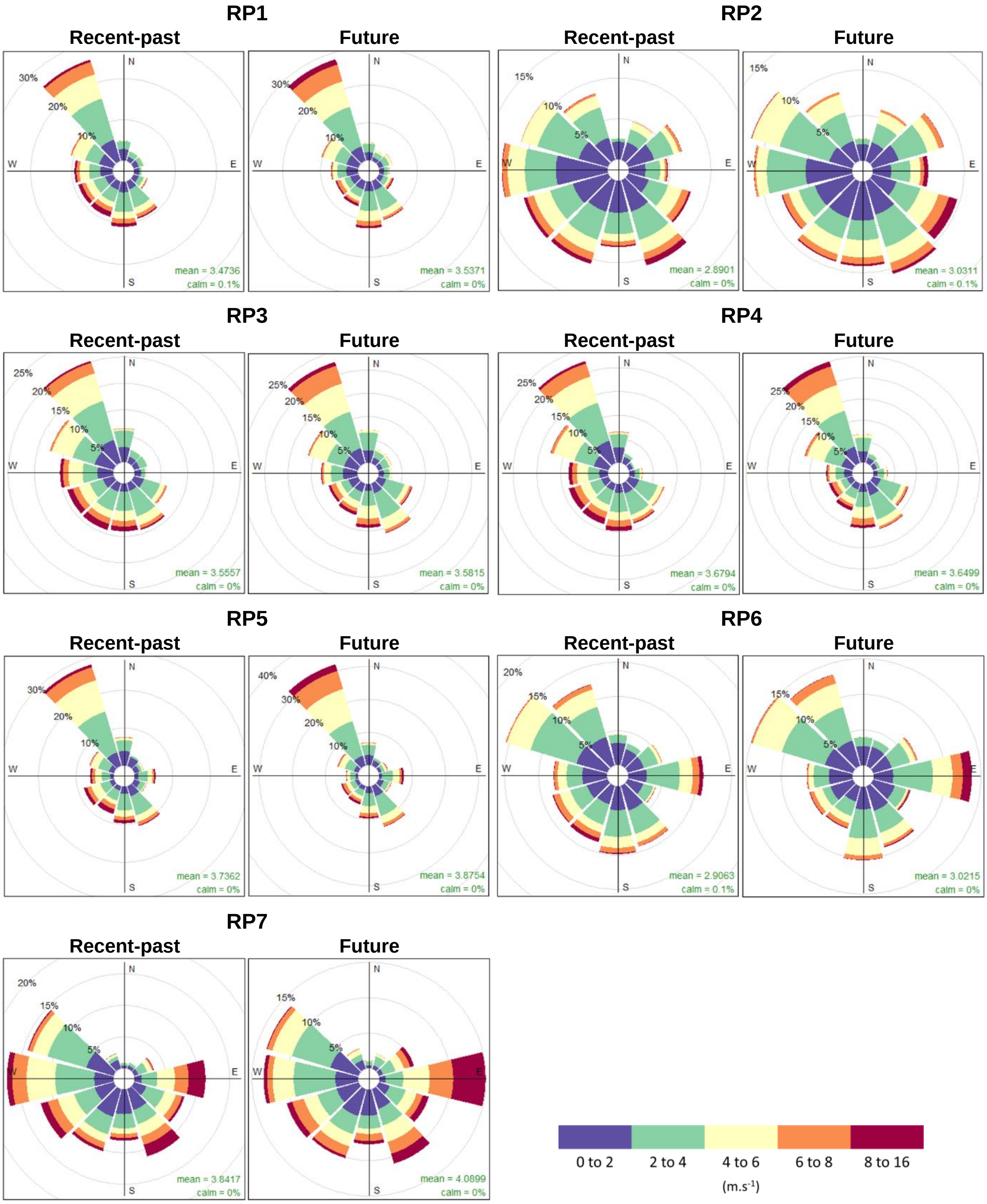

Figure 5.7. *Wind roses of each receptor point, for both medium-term future and recent-past climates.*

The analysis of Figure 5.7 shows that, for the receptor points RP1, RP3, RP4 and RP5, the predominant winds are from northwest, with an occurrence between 25% and 40% of the time. RP1, RP3 and RP4 are located near the coast and the lagoon, in areas with an open landscape and terrain altitudes up to 20 m. On the contrary, RP5 is located inland at around 75 m altitude. For the others, RP2, RP6 and RP7, there is no clear predominance of wind direction due to the complex terrain of their locations. Regarding wind speed, for the recent-past period, average values vary between 3.5 and 3.8 $m.s^{-1}$ for most of receptor points. RP2, located in the area with the highest pollutants concentrations, is the exception, with an average wind speed of 2.9 $m.s^{-1}$. Analysing the difference between medium-term future and recent-past climate, a continuity of the predominant wind directions is expected for all receptor points, but with a slight increase in the average wind speed, thus favouring the transport and dispersion of pollutants.

The analysis of prevailing winds helps to understand the transport of pollutants between municipalities. For each receptor point, the relative contributions of the municipalities in the Aveiro region are similar for all pollutants analysed. For example, RP2, placed in Estarreja municipality, and with predominant winds from the third quadrant, has Aveiro (southeast of the RP2) as the main contributor. The RP5, located in Águeda municipality, has its highest contributions coming from Albergaria-a-Velha and Aveiro, Northwest of the RP5, which is the predominant direction of the winds recorded at that receptor point. The RP7, placed in Ovar, the northernmost municipality of Aveiro Region, has similar contributions from all the 10 municipalities, located South of the RP7, where the majority of the obtained winds come from.

The assessment of the source contributions by sector and area, together with the prevailing winds analysis, proved to be an added value for the characterization of the air quality in the Aveiro Region. With that analysis it can be concluded that, in general, the industrial sector plays an important role in the air quality levels of the Aveiro Region, being the Aveiro municipality the one with the highest contribution. However, the source apportionment analysis also showed the importance of long-range transport for PM concentrations, as well as the influence that the NO_2 pollution levels of bigger metropolitan areas can have in the Aveiro Region air quality.

5.4.2. Health Impact Assessment

The air quality outputs from previous section were used as input to assess the health impact of air pollution in Aveiro Region. Figure 5.8 shows the number of premature deaths due to PM2.5 and NO_2 long-term exposure, for the medium-term future climate, recent-past climate, and the difference between them.

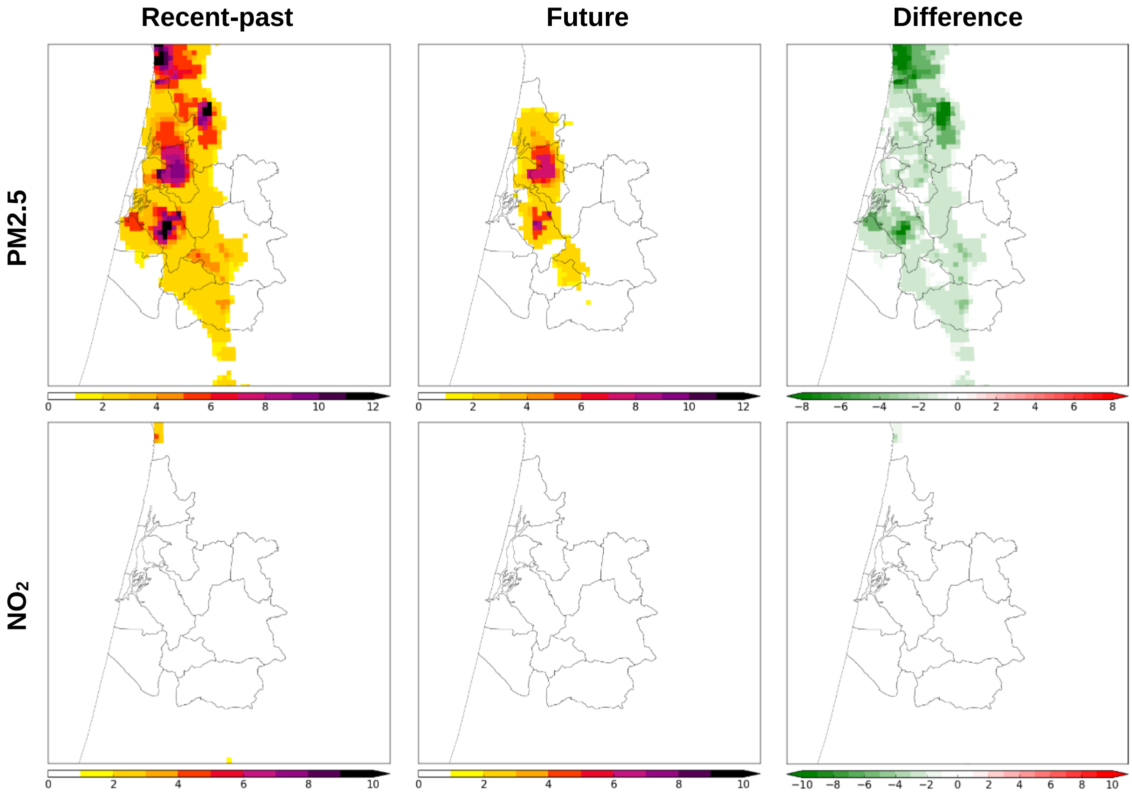

Figure 5.8. *Number of premature deaths due to PM2.5 and NO_2 long-term exposure, for the medium-term future climate (2055), recent-past climate (2014) and the difference between them (2055 – 2014). Note that for the difference maps, negative values mean avoided premature deaths.*

PM2.5 long-term exposure led to 3448 (95% CI: 2512 - 3921) and 829 (95% CI: 559 - 962) premature deaths in the recent-past and medium-term future climate, respectively, representing around 76% of premature deaths avoided in the future. According to Figure 5.8, premature deaths due to PM2.5 long-term exposure are mostly obtained near the coastline, where the population density is higher. For the recent-past, outside of the Aveiro Region, in the north of the domain, Porto metropolitan area is the area with highest number of premature deaths; and in the Aveiro Region, Estarreja, Aveiro, Ovar, and Ílhavo municipalities are the ones with more premature deaths. In the future, despite the 76% of premature deaths avoided due to the implementation of RNC2050 emission reductions, it is expected that premature deaths continue to occur, with special relevance in Estarreja and Aveiro municipalities, where higher pollutant concentrations were previously obtained. For NO_2, 32 (95% CI: 10 - 95) premature deaths, located in the north of the domain, are estimated for the recent-past. For the medium-term future, no premature deaths are expected due to NO_2 long-term exposure. From Figure 5.8 it is clear that the number of expected premature deaths due to the long-term exposure of air pollution relates the magnitude of the pollutant’s concentration and the exposed population. This is evident for NO_2, where the recent-past higher concentrations, obtained in the south of the domain in an area with low population density, do not lead to premature deaths. The same does not happens for PM2.5, with higher premature deaths located in areas with high PM2.5 concentrations and high population density.

5.5. ADDITIONAL EMISSION ABATEMENT STRATEGIES

In the previous section it was possible to verify that in the future, it is expected that the air quality in the region of Aveiro will improve. Although the future emission reductions of the main air pollutants have been defined with the objective of carbon neutrality and the fight against climate change, they will also have a positive impact on air quality. However, PM2.5 pollution will continue to occur in the Aveiro region, exceeding the limit value proposed in the new proposal for the revision of the EU Ambient Air Quality Directive (EC 2022), with negative impact on the health of their inhabitants, leading to the premature deaths. To overcome this problem, additional emission abatement measures must be considered.

From the analysis carried out in the previous section, it was possible to conclude that, in the future, the SNAP3&4 sector will be the main responsible for the concentrations of PM2.5. Although the RNC2050 intermediate scenario already foresees the elimination of fossil fuels use, it predicts an increase in the use of biomass, thus contributing to the increase in PM emissions (Monjardino et al. 2019, 2021). It was also concluded that the PM2.5 concentrations do not originate only in the municipalities of the Aveiro Region, but also in the surrounding areas.

Thus, the authors propose two scenarios with emission abatement strategies additional to those of the RNC2050 intermediate scenario, which should be applied to the SNAP3&4 sector and to the entire territory of mainland Portugal, by 2050. The proposed scenarios are as follows:

(i) Scenario 1 (SCEN1): RNC2050 emissions projections will be maintained for all sectors, except from SNAP3&4 where CO, NOx and SOx emission reductions will be maintained, but increases in NMVOC, PM and NH3 emissions will be reduced by half.

(ii) Scenario 2 (SCEN2): RNC2050 emissions projections will be maintained for all sectors, apart from SNAP3&4 where CO, NOx and SOx emission reductions will be maintained, but NMVOC, PM and NH_3 emissions will not increase, remaining equal to those considered in the recent-past climate (2014).

The future emission projections considered in the scenarios can be consulted in Table 5.4.

Table 5.4. *Medium-term future emission projections considered in RNC2050 and scenarios 1 and 2 (SCEN1 and SCEN2), expressed in % of the recent-past, for CO, NOx, PM2.5, PM10, SOx, NMVOC, and NH_3, for the target sector of SCEN 1 and SCEN2 - SNAP3&4 - industrial combustion and processes, and the total of all sectors. Note that changes in relation to RNC2050 are highlighted in bold.*

	RNC2050		**SCEN1**		**SCEN2**	
	SNAP3&4	All sectors	SNAP3&4	All sectors	SNAP3&4	All sectors
CO	-60.9	-69.6	-60.9	-69.6	-60.9	-69.6
NOx	-30.8	-68.0	-30.8	-68.0	-30.8	-68.0
PM2.5	+18.8	-38.2	**+9.4**	**-42.2**	**0.0**	**-46.2**
PM10	+17.9	-33.4	**+9.0**	**-37.1**	**0.0**	**-40.7**
SOx	-24.9	-35.5	-24.9	-35.5	-24.9	-35.5
NMVOC	+10.8	-27.1	**+5.4**	**-28.5**	**0.0**	**-30.0**
NH_3	+13.1	-15.2	**+6.6**	**-16.0**	**0.0**	**-16.7**

The previously defined air quality modelling methodology (described in section 5.3.1) was applied, considering the two scenarios with additional emission abatement strategies. The modelling results of SCEN1 and SCEN2 were then compared with the medium-term future results of RNC2050 without considering further action, as shown in Figure 5.9.

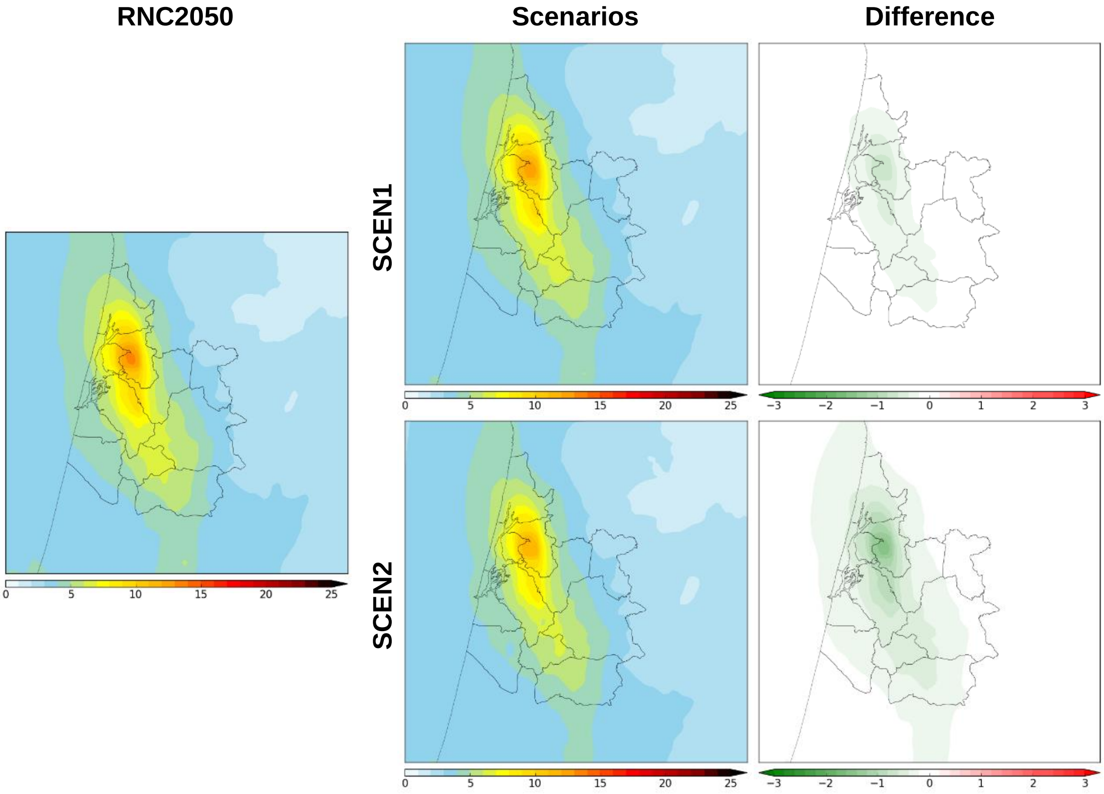

Figure 5.9. *Annual average concentration of PM2.5, in µg.m^{-3}, for the medium-term future climate without considering further action (RNC2050), considering the scenarios 1 and 2 (SCEN1 and SCEN2), and the difference between them (RNC2050 – SCEN). For future maps, the maximum value of the colour scale represents the EU limit value.*

Figure 5.9 shows that both scenarios considered, SCEN1 and SCNE2, will lead to an improvement in the medium-term future air quality in the Aveiro Region. For SCEN1, it is expected that PM2.5 annual concentrations will decrease by up to 0.8 µg.m^{-3}, when compared to the results of the medium-term future climate without considering further action. For SCEN2, greater reductions of up to 1.5 µg.m^{-3} are expected. In both scenarios, major reductions are expected to occur in Estarreja municipality, the most industrialised area in the Aveiro Region and where the PM2.5 annual concentrations are higher, followed by Aveiro municipality, the second municipality with the highest population density and where most of the population lives and works.

From the analysis of Figure 5.9, it is possible to conclude that SCEN2 will be the only one to guarantee a PM2.5 annual concentration below or equal to the limit value defined in the proposal for a new EU Ambient Air Quality Directive (EC 2022).

In terms of health impact due to PM2.5 long-term exposure, both scenarios will continue to lead to premature deaths, although in smaller numbers (see Figure 5.10). When compared to the results of the medium-term future climate without considering further action, SCEN1 will lead to a reduction of about 16%, accounting for a total of 699 (95% CI: 474 - 818) premature deaths. For SCEN2, the reduction will increase to 29%, with a total of 585 (95% CI: 390 - 665) premature deaths due to PM2.5

long-term exposure. In both scenarios, premature deaths will occur mainly in Estarreja and Aveiro municipalities.

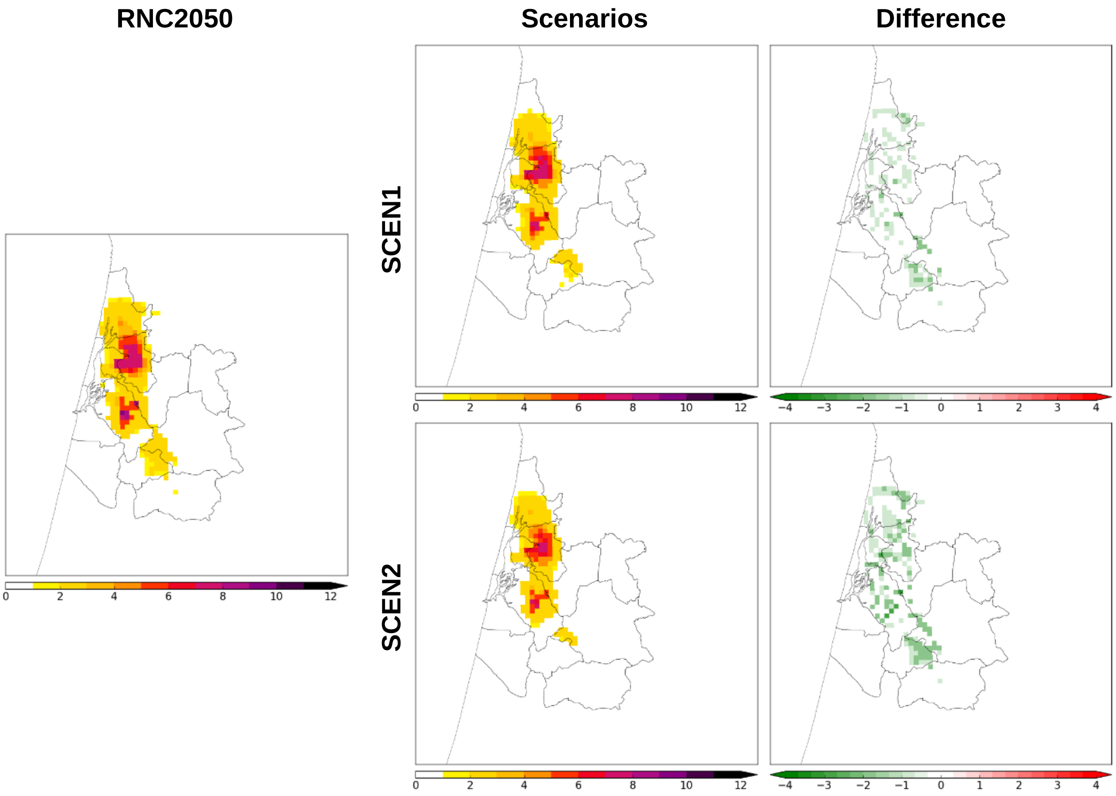

Figure 5.10. *Number of premature deaths due to PM2.5 long-term exposure, for the medium-term future climate without considering further action (RNC2050), considering the scenarios 1 and 2 (SCEN1 and SCEN2), and the difference between them (RNC2050 – SCEN). Note that for the difference maps, negative values mean avoided premature deaths.*

Although SCEN2 PM2.5 annual concentration does not exceed the new proposed EU limit value, it will not prevent premature deaths due to long-term exposure. According to WHO (2021a), only with PM2.5 annual concentrations less than 5 μg.m^{-3}, no health impacts are expected. For this to happen, SNAP3&4 emissions would have to be further reduced to values below those of the recent-past (2014).

A study made for Portugal (Borrego et al. 2012) suggested the improvement of industrial PM retention systems, the reinforcement of the inspection of industry sources, and the establishment of emission standards for industrial clusters and business activities in urban areas, as the main measures to reduce PM emissions. More recently, Monjardino et al. (2021) highlighted the SNAP3&4 as the most relevant sector to tackle air pollution in the future in Portugal, mainly in the industrial processes emissions due to the limited range of technological options to reduce them. Studies conducted for other case studies (Zhang et al. 2019; Cui et al. 2020) concluded that main drivers of air quality improvement would be: (i) strict industrial emission standards; (ii) industrial boilers upgrades; (iii) gradual elimination of outdated industrial units; and (iv) switch industries to the use of cleaner fuels. WHO (2021b) also highlighted the use of clean technologies that reduce industrial smokestack emissions as successful policies to reduce air pollution from the industry sector.

5.6. CONCLUSIONS AND RECOMMENDATIONS

The main purpose of this study was to apply a modelling approach able to assess the impacts of future climate and projected emissions, including the assessment of the relative contribution of different source regions and sectors, on air quality at urban scale, as well as their impact on human health. The WRF-CAMx modelling system, including the PSAT tool, was applied to the Aveiro Region, in Portugal, to quantify the contribution of multiple source sectors and areas to PM2.5, PM10 and NO_2 concentrations, now and in the future.

The main outcomes of the current research are summarized as follows:

(i) In the medium-term future climate, considering the emission projections of the RNC2050 intermediate scenario, the air quality in Aveiro Region will improve, due to the reduction of PM2.5, PM10 and NO_2 concentrations. For that period, SNAP3&4 will be the largest source sector of PM2.5 and PM10 annual concentrations, and the second largest for NO_2 annual concentrations, only preceded by SNAP8. The main source area will be the Aveiro Region surrounding area, although there is a relevant contribution from the Aveiro municipality.

(ii) The improvement of the Aveiro Region air quality, in the medium-term future, will ensure that the limit values of the EU Air Quality Directive will not be exceeded. However, the same will not happen if the proposed revision of the EU Air Quality Directive is approved, with exceedances to the annual limit value of PM2.5 expected to occur. Also, premature deaths due to PM2.5 long-term exposure will be reduced by 76% in the medium-term future.

(iii) The additional emission abatement scenario SCEN2, without the PM emission increase projected in the RNC2050 intermediate scenario, demonstrated that, in the medium-term future, it is possible to comply with the new PM2.5 limit value proposed. However, this will not prevent premature deaths due to PM2.5 long-term exposure, which, according to the WHO, will only happen if PM2.5 annual concentrations are lower than 5 $\mu g.m^{-3}$.

These results show the potential for air quality improvement that can occur as a consequence of measures taken to achieve carbon neutrality. In addition, this study also shows that the definition of measures to fight climate change, as a way of guaranteeing carbon neutrality, may not be sufficient to achieve the future air quality objectives, being necessary to quantify the impacts that projected emissions will have on air quality. Measures that benefit both climate and air quality policies should be promoted, while measures that benefit one over the other should be avoided. As an example, measures to reduce fossil fuel consumption are particularly beneficial for both policies. However, measures that increase the consumption of biomass, if not accompanied by appropriate technologies for reducing emissions, will be detrimental to air quality. More specifically, this study evidenced that the already considered measures for the two main emitting sectors, residential combustion and road transport, are not sufficient to control PM2.5 emissions and their impacts. It was also demonstrated that an additional effort in establishing efficient and win-win strategies in the industrial sector would benefit air quality and health in the future.

In conclusion, the methodology applied in this work is an asset in this research field, providing valuable information for policy makers. This work, by quantifying the contribution of different source sectors and areas, helps to raise awareness of the impact that current lifestyles have on air quality and their effects on health. In addition, the applied methodology, easily applied to other case studies, constitutes an added value to several scientific communities, enhancing scientific advances through harmonizing research on climate change, air quality and health impact in urban areas.

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5.8. SUPPLEMENTARY MATERIAL

Figure 5.11. *Daily contribution of each source sector for PM2.5 concentration, in µg.m^{-3}, for the recent-past climate (2014), for each receptor point.*

Figure 5.12. *Daily contribution of each source sector for PM2.5 concentration, in µg.m^{-3}, for the medium-term future climate (2055), for each receptor point.*

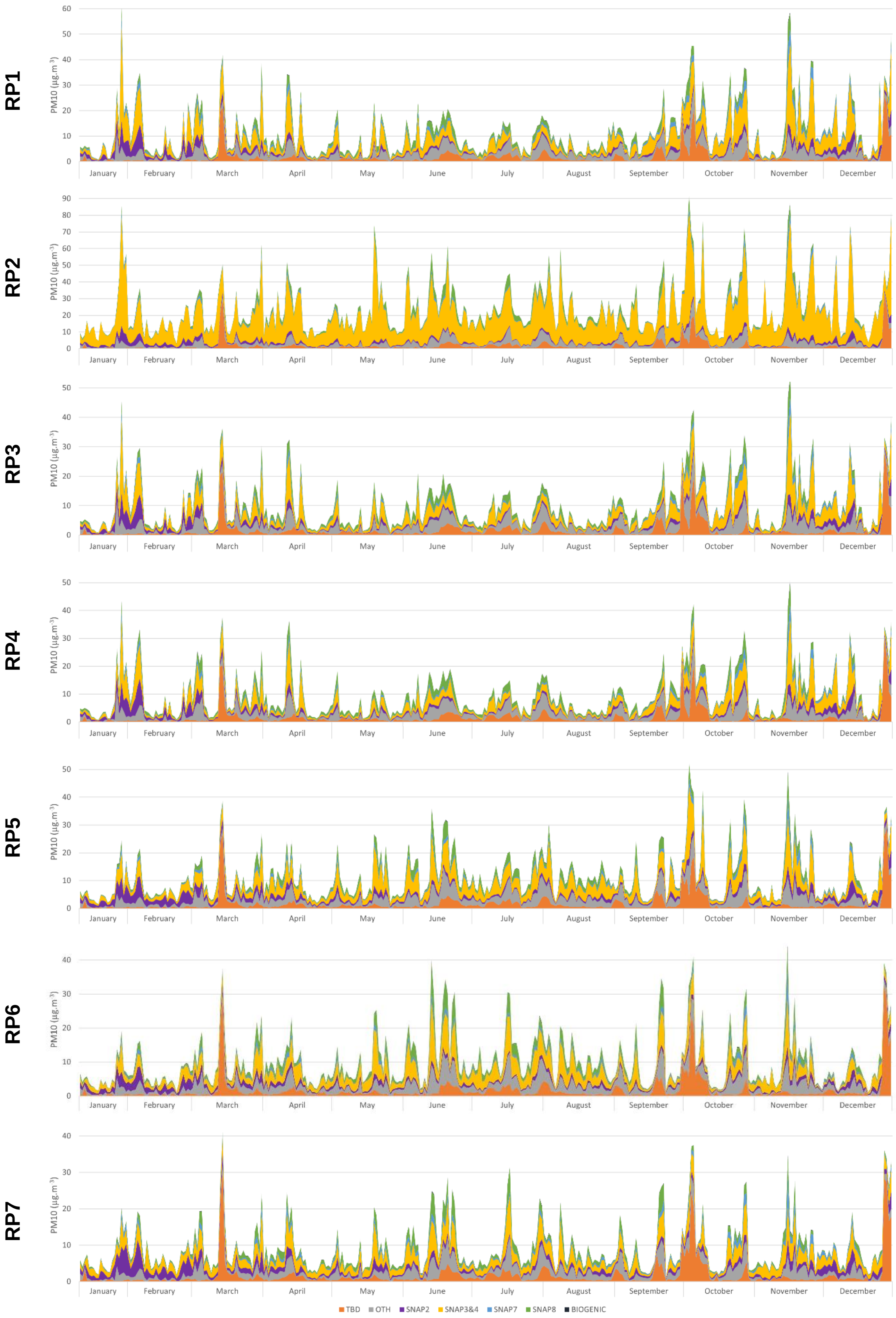

Figure 5.13. *Daily contribution of each source sector for PM10 concentration, in µg.m^{-3}, for the recent-past climate (2014), for each receptor point.*

Figure 5.14. *Daily contribution of each source sector for PM10 concentration, in µg.m^{-3}, for the medium-term future climate (2055), for each receptor point.*

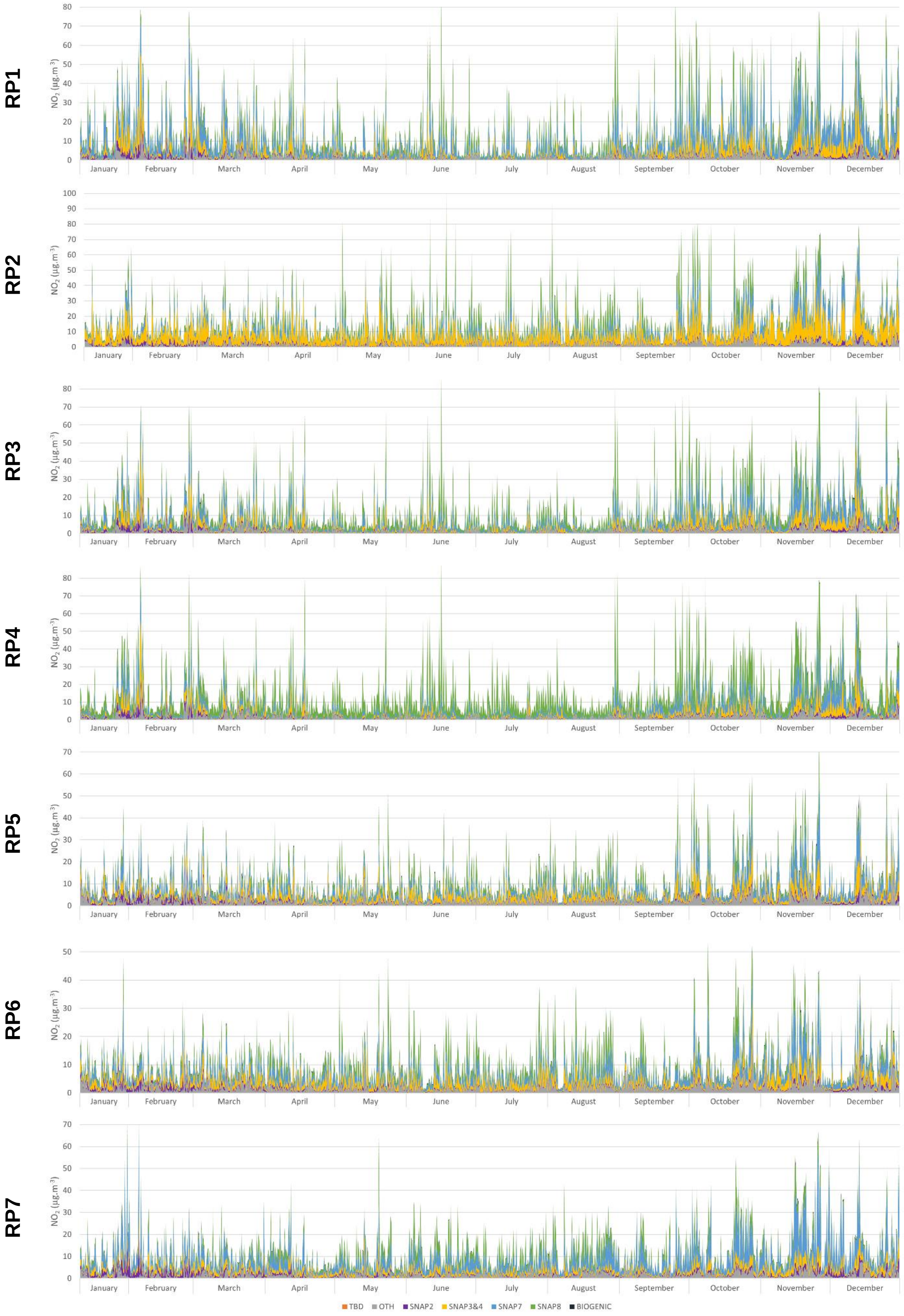

Figure 5.15. *Hourly contribution of each source sector for NO_2 concentration, in $\mu g.m^{-3}$, for the recent-past climate (2014), for each receptor point.*

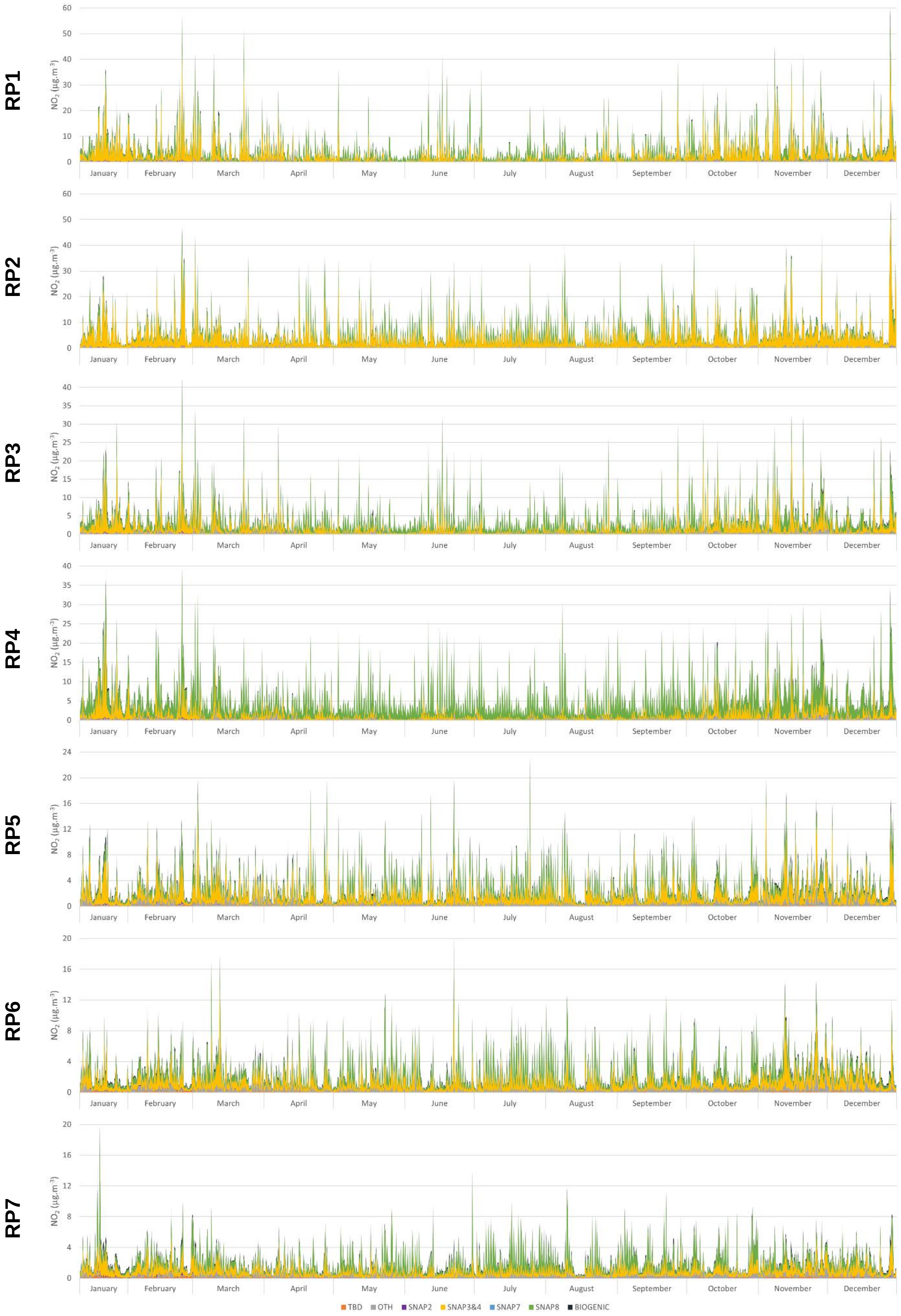

Figure 5.16. *Hourly contribution of each source sector for NO_2 concentration, in µg.m^{-3}, for the medium-term future climate (2055), for each receptor point.*

CHAPTER 6

6. GENERAL CONCLUSIONS

The conclusions and remarks of the PhD work are presented in this Chapter. The answers to the research questions that gave rise to this PhD work are summarized and suggestions of future work are provided.

6.1. SUMMARY OF RESEARCH AND FINDINGS

Integrated approaches tackling air pollution and climate change can play an important role to support urban planning oriented for citizen welfare. Furthermore, win-win strategies on climate and air quality management to be included in national and regional policies, strategies and planning should rely on scientific research support.

In this context, the main goal of this work was to develop a modelling approach able to assess the impacts of future climate and projected emissions, including the relative contribution of different source regions and sectors, on air quality at urban scale.

This thesis contributes to improve the knowledge of integrated measures and policies for both air quality and climate mitigation and/or adaptation, providing valuable information for policymakers. The work developed allows to accomplish the thesis goals and to answer to each of the four formulated research questions.

1. **Which are the best available tools to account for air pollution and climate change impacts on air quality and human health in an integrated way?**

The first step of the scientific investigation necessary to answer to this research question comprised a review on source apportionment models and techniques applied at urban scale, highlighting their purpose and their advantages and disadvantages (Chapter 2). From the literature review, it was concluded that the tagged species method is the most used by the scientific community and the one that provides a more complete assessment of air quality, identifying contributions from both source regions and activity sectors, as well as the transboundary transport. Nevertheless, this method has a disadvantage: it is not able to assess the impact of abatement measures to support the design of air quality plans, requiring the use of additional brute force methods. Consequently, it can be concluded that the two methods, tagged species and brute force, must be used in a complementary way, allowing the assessment of air quality, and supporting the design of air quality management plans in urban areas.

Following that, the tagged species method, using CAMx PSAT tool, was applied to six EU urban areas (including the Aveiro Region), to quantify the contribution of multiple source sectors to PM and NO_2 concentrations, and to assess the CAMx performance (Chapter 3). The obtained results fulfil the FAIRMODE modelling quality objective, ensuring that the minimum level of quality for policy use was achieved by the CAMx model. Main outcomes state that CAMx, with the PSAT tool, can represent a powerful tool to provide useful information about pollutants concentrations as well as estimate the source contribution of the different emission categories.

The health impact assessment methodology, applied to the Aveiro Region for recent-past and medium-term future climates, was defined in Chapter 4. This assessment has as its starting point the methodology applied in the EEA annual health assessments, which considers the CRF and RR defined in HRAPIE and REVIHAAP projects of WHO. Additionally, a new methodology was also considered, based on the recent update of the WHO air quality guidelines, and on the definition of more updated CRF and RR. The health impact main results for the recent-past climate are in line with those obtained in the EEA report, demonstrating the accuracy of the applied methodology.

All previously defined models and methodologies were considered into a final approach, applied to the Aveiro Region, for both recent-past and medium-term future climates (Chapter 5). The source apportionment brute force method, previously defined as having a key role in quantifying the impact of abatement measures to support the design of air quality plans, was also applied to the Aveiro Region. The applied approach, with reliable and realistic results, allowed the definition of an integrated modelling framework capable of accounting for air pollution and climate change impacts on urban air quality and human health, in a comprehensive way.

2. What is the relative contribution of emission source regions/activities to air quality, now and in the future, considering a climate change scenario?

The first attempt to answer to this question was made in Chapter 2, where the WRF-CAMx, with its PSAT tool, was applied to six EU cities and regions, including the Aveiro Region. Using several distinct case studies, which are distinguished by their location, population density and air pollution context, it was also possible to understand the variability of air pollution issues and their origin across EU urban areas. Additionally, this work evidenced that similar air quality problems can have different causes, depending on emission sources and specific characteristics of each location.

The main outcomes from Chapters 3 and 5 allowed to conclude that, nowadays, the industrial activity sector, i.e., SNAP3&4, is the one that most contributes to the air pollution levels registered in the Aveiro Region. For PM, transboundary transport also plays an important role in the region, highlighting the importance of long-range transport. For NO_2, alongside with SNAP3&4, the sectors with the highest contribution are SNAP7 and SNAP8, i.e., road transport and aviation, maritime, and off-road transport, respectively, highlighting the importance of local sources for NO_2 concentrations. In the medium-term future, when compared to the recent-past, SNAP3&4 will be the only sector that will increase its contribution to the PM2.5 and PM10 concentrations. On the other hand, transboundary transport will maintain its contribution. For NO_2, the contributions of SNAP8, and SNAP3&4 will increase in the future; and, on contrary, SNAP7 will decrease. This is a result of the planned strategies for carbon neutrality, that also have impacts on air pollutant emissions, as further detailed below, in the answer to question 3.

Regarding the source regions contribution, in general, the areas surrounding the Region of Aveiro are those that most contribute to the PM2.5, PM10 and NO_2 concentrations estimated for the region. However, analysing the contribution of each Aveiro Region municipality, Aveiro, the municipality with more inhabitants and greater economic activity, is the one that most contributes to all the analysed receptor points. This analysis showed similar results for both recent-past and medium-term future.

3. What is the impact of existing climate policies on air quality and health?

Aligned with the Paris Agreement, Portugal has defined three possible carbon neutrality trajectories in its RNC2050, with distinct implications on air pollutant emissions. This thesis work considered the emission projections of the RNC2050 intermediate scenario, since it is in line with SSP2-4.5 scenario considered in WRF simulation (Chapters 4 and 5). This medium ambition scenario considers a socioeconomic evolution based on the development and application of new technologies that, however, do not significantly change either the production structures or the population's lifestyles. In this scenario, the total emissions of each pollutant will have an average reduction between 15% and 70%, in relation to the recent-past.

The methodology applied in this thesis work, with WRF-CAMx modelling system, was able to account for the impacts that the proposed RNC2050 scenario will have in future climate and air quality (Chapters 4 and 5). According to the main outcomes, it is expected that air quality will improve at the middle of the century, in Aveiro Region. Despite the climate changes that will affect the region, with an increase in dry spells due to increased temperature and decreased precipitation, the concentrations of the main air pollutants will decrease. For both PM10 and PM2.5, the differences between mid-term future climate and recent-past climate are between -1 and -4 $\mu g.m^{-3}$. Regarding NO_2, differences between mid-term future and recent-past climates range from -1 to -22 $\mu g.m^{-3}$. For O_3 concentrations (only evaluated in Chapter 4) no relevant changes are expected. The expected reduction in PM10, PM2.5 and NO_2 annual concentrations will ensure that, in the medium-term future, there will be no exceedances to the limit values of the European Union (EU) Air Quality Directive. But, in some areas of the Aveiro Region, if the new directive is approved in its current form, future PM2.5 annual concentration will be above the new limit value of 10 $\mu g.m^{-3}$.

Regarding the impact on human health, as already mentioned in the research question 1, the health impact assessment methodology was extensively described and applied in Chapters 4 and 5. In a first approach, this assessment was carried out for the pollutants with the most robust scientific evidence, i.e., PM2.5, NO_2, and O_3 (Chapter 4) and after, as O_3 pollution did not have associated premature deaths in any of the analysed periods, only for PM2.5 and NO_2 were considered (Chapter 5). For these pollutants, the mortality health outcomes due to long-term exposure were analysed as it is the most serious health impact. In both studies, when considering the recent-past climate, only PM2.5 and NO_2 long-term exposure lead to premature deaths in Aveiro Region. In the medium-term future, following the air quality trend, the number of premature deaths will also decrease, with only premature deaths related to PM2.5 pollution being expected.

In conclusion, there is potential for improving air quality, and the effects on human health, arising from measures taken to tackle climate change. However, despite the general improvement in air quality expected for the medium-term future, PM2.5 pollution will continue to occur in the Aveiro region, with exceedances to the limit value defined in the proposed revision of the EU Ambient Air Quality Directive, continuing to affect the human health and leading to premature deaths.

4. What additional policies are needed to support win-win strategies for air quality and climate mitigation and/or adaptation, at urban scale?

From the analysis carried out in Chapter 5, it was possible to state that, in the medium-term future, SNAP3&4 will be the main sector responsible for the PM2.5 concentrations, and those concentration levels will come mainly from the Aveiro Region surrounding areas. As the RNC2050 intermediate scenario foresees an increase in the use of biomass, as alternative to the fossil fuels use, this will contribute to the increase in PM emissions.

In this context, by 2050, additional emission abatement strategies should be applied to the SNAP3&4 sector and to the entire territory of mainland Portugal. New possible emission reduction scenarios were tested through the application of the previously defined integrated modelling framework, which includes the use of source apportionment brute force method (Chapter 5). Thus, it was possible to conclude that, if medium-term future PM emissions are kept equal to those considered in the recent-past, PM2.5 concentrations will not exceed the new proposed EU annual limit value. However, this will not prevent premature deaths due to PM2.5 long-term exposure, since only annual concentrations less than 5 μg.m^{-3} will not impact human health. To achieve this objective, an additional effort to establish efficient and win-win strategies in the SNAP3&4 sector must be made to benefit both air quality and human health in the future. As an example, the use of clean technologies that reduce industrial smokestack emissions may be a possible strategy to reduce SNAP3&4 PM2.5 concentrations.

To summarize, the integrated modelling framework defined in this thesis work, was able to assess the impacts of future climate and projected emissions, including the relative contribution of different source regions and sectors, on air quality, as well as the air pollution impact on human health. The developed integrated methodology can be easily replicated to other case studies, fulfilling the need for further studies that assess the future climate effects on air quality, in urban areas. This approach can represent a powerful tool to provide valuable information for the design of best win-win strategies on climate and air quality management, to support urban planning oriented for citizen welfare, and help citizens increasing awareness about both air pollution and climate change. Additionally, both modelling and epidemiologist scientific communities may benefit from the advances in the coordination between climate change, air quality, and health impact assessments in urban areas.

6.2. FUTURE CHALLENGES

The integrated modelling framework developed in this thesis work enables a deeper scientific knowledge about climate change and air pollution and support urban planning and decision makers. However, despite the scientific advances achieved, some additional research work must be done.

One of the recommendations is that longer time periods should be considered in the application of the integrated modelling framework, ideally over 30 years, in order to obtain the best possible climate signal. In this context, also other global model than MPI-ESM should be used as WRF climate forcing, as well as additional climate change scenarios. As socioeconomic evolution, as well as climate change, may have impact on future land use classification, these changes should be quantified and used as updated inputs in both WRF and CAMx models, to obtain better and more realistic results.

Concerning future emissions, specific projections for the defined case study should also be quantified and considered, since these may not be proportional to the national total. An example of this are the emission projections for the industrial sector, which significantly vary according to the type of industry considered.

For the health impact assessment, to achieve a more comprehensive assessment, mortality outcomes from short-term exposure, as well as morbidity short- and long-term exposure, resulting in the occurrence of illness and years lived with a disease or disability, could be quantified. The costs, direct and indirect, associated with mortality and morbidity, should also be estimated, using the value of statistical life approach. Direct costs may include hospital admission, outpatient appointments and medication as well as caregivers' time, among others. While indirect costs must include expenses associated with loss of productivity due to morbidity as well as loss of production due to morbidity or mortality. Additionally, to obtain more realistic and accurate results, this analysis should be based on specific data for Portugal, rather than generic literature data.

It is predicted that climate change will increase the dryness periods, also affecting dust concentration levels in the atmosphere, as well as in the occurrence of forest fires, both very large sources of PM to the global troposphere. Thus, these emission sources, which will certainly have relevant contributions to the air quality and human health impact in the forthcoming decades, must also be considered in a future modelling system.

Last but not least, no research work is fully recognized and valued if it is kept within the research community. Thus, it is very important to enhance the collaboration and the dialogue between scientists and policy-makers to take the best benefit of up-to-date scientific knowledge to support decision-making for environment and health protection against air pollution and climate change.