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Noela Kesline Vieira de Pina

Analysis of traffic related atmospheric aerosol particles in an urban environment

PhD Thesis in Doctoral Program in Transport Systems supervised by Professor Oxana Tchepel and presented to the Department of Civil Engineering of the Faculty of Sciences and Technology of the University of Coimbra.

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Faculty of Sciences and Technology
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“You never fail until you stop trying.”

Albert Einstein

Financial support

This research was funded by “Fundação para a Ciência e a Tecnologia” (FCT, Portugal) through research grant PD/BD/128048/2016 under the MIT Portugal Program and in the frame of two research projects TRAPHIC (PTDC /ECM-URB/3329/2014, POCI-01-0145-FEDER-016) and ISY-AIR (MITEXPL/ 0023 2017).

Acknowledgments (Agradecimentos)

É com uma enorme satisfação que agradeço a todos que me acompanharam nesta caminhada e contribuíram direta ou indiretamente para a conclusão deste trabalho.

Primeiramente, expresso a minha profunda gratidão à minha orientadora Professora Oxana Tchepel. Toda a paciência dedicada, apoio, criticismo e os conselhos dados ao longo destes anos, permitiram-me crescer como investigadora. Agradeço profundamente a oportunidade de trabalhar em diferentes projetos de investigação, tornando-me hoje a profissional que sou. Mas acima de tudo, agradeço a humanidade e gentileza que me permitiu chegar aqui.

Também expresso a minha gratidão ao Professor António Pais Antunes, pelo apoio deste o primeiro momento, por ter sido a primeira pessoa que me motivou a realizar este doutoramento e por ser um profissional que inspira os alunos que por si passam.

Moreover, I want to express my gratitude to all my colleagues from the University of Coimbra and MIT Portugal Program. A special thanks to my friends Daniela, Ana Beatriz, Susana, João Pedro, Marta, Ana Ibraeva, Rita, Aymen, Nathale, Anne Patricio, Kiana, Vância and Inês Cunha. Daniela muito obrigada pelo carinho e suporte na minha investigação, a quem eu gosto de chamar a minha segunda “mãezinha” deste doutoramento. Obrigada por todo o aprendizado e motivação. Ana Beatriz obrigada por seres essa “luz ambulante” e pelo carinho.

Agradeço também à família UBIWHERE, pela amizade e compreensão para executar este doutoramento nas horas “vagas” do trabalho.

Obrigada também às minhas meninas, Carmen, Júlia e Kátia, por serem as minhas “psicólogas” de plantão mesmo com oceanos a separar-nos. Há quem tenha uma, eu sou sortuda e tenho três, obrigada.

Gratidão é pouco pelo suporte dado pela minha família nesta caminhada. Aos meus pais e aos meus irmãos, obrigada. Maninha obrigada pelas longas horas de conversa e suporte.

Estarei sempre em dívida ao Gilberto e ao meu filhote Leo. Gilberto, meu amor, dedico-te esta tese por seres o meu companheiro de luta e sempre acreditares em nós. Leo, meu lindo, obrigada por seres a minha fonte de inspiração.

Abstract

Urban air pollution is one of the major concerns throughout the world. In Europe, the urban air quality (AQ) has been constantly improved. However, exceedances of particulate matter (PM), a harmful pollutant to the human health and environment, are still observed in the cities. Road traffic is identified as one of the major anthropogenic sources contributing globally about 25% of urban ambient air pollution from $PM_{2.5}$. Hence, detailed characterization of PM emission sources is seen as a priority when AQ improvement measures are on the agenda. Furthermore, in order to support short-term measures, the estimation and forecast of urban AQ is highly important, which allows the anticipation of exposure to higher levels of harmful pollutants.

The main objective of the current study was the development and implementation of a methodology to estimate and forecast aerosol particles in the urban atmosphere, by considering road traffic as one of the primary urban pollution sources. For this purpose, novel approaches were developed to access both background and local contributions to urban air pollution. Therefore, existing AQ forecasting services at a regional scale were explored and implemented to obtain the background concentrations. The traffic-related local contributions required further advances of a transport-emission-air dispersion modelling system. Thus, a traffic emission model was extended and improved to include cold start and non-exhaust emissions with high spatial and temporal resolution, which was crucial to produce better predictions of PM and black carbon (BC) concentrations. Furthermore, field measurements implemented for the study area were considered for the validation of the modelling system and to improve the knowledge on aerosol particles in the urban atmosphere.

Overall, this research work expected to enhance the comprehension of urban aerosol pollution, allowing to depict road traffic contribution. Actually, although the recent vehicle technologies advances, this thesis demonstrates that, for the case study deeply analysed, road traffic remains one of the major sources of urban air pollution. Additionally, the integrated modelling approach improvements offers an innovative opportunity for urban AQ assessment and forecast. Furthermore, the aerosol temporal and spatial variation in addition to the PM characterization obtained from this research, provides relevant data for analysis of health impacts and research in the field of atmospheric pollution and climate change, thus, supporting decision-making towards urban sustainable development.

Key Words: particulate matter; road traffic emissions; air pollution; urban areas; forecast; modelling.

Resumo

A poluição do ar em meio urbano representa uma das principais preocupações globais. Na Europa, a qualidade do ar (QA) urbano tem vindo a melhorar, contudo em diversas cidades são registadas excedências de material particulado (PM), um poluente prejudicial para a saúde humana e o meio ambiente. O tráfego rodoviário é identificado como uma das principais fontes de emissões antropogénicas nas cidades, sendo responsável por cerca de 25% do $PM_{2.5}$ presente na atmosfera urbana. Portanto, a caraterização detalhada das fontes de emissão de PM torna-se prioridade quando se trata de melhorar a QA. Além disso, a fim de apoiar medidas de mitigação a curto prazo, a estimativa e a previsão da QA urbano desempenham um papel importante, permitindo antecipar a exposição a níveis elevados de poluição.

Este estudo teve como principal objetivo o desenvolvimento e a implementação de uma metodologia para estimar e prever a concentração de aerossóis na atmosfera urbana, considerando o tráfego rodoviário como uma das principais fontes de poluição. Para tal, foram desenvolvidas novas abordagens para obter as contribuições de fundo e locais para a poluição do ar urbano. Portanto, foram explorados e implementados serviços existentes de previsão da qualidade do ar em escala regional para obter concentrações de fundo. Já as contribuições locais provenientes do tráfego exigiram novos avanços no sistema de modelação de transporte-emissão-qualidade do ar. Neste contexto, aprimorou-se um modelo de emissões de tráfego, incluindo as emissões originadas de processos de arranque a frio e de não exaustão, com uma alta resolução espacial e temporal, permitindo produzir melhores previsões das concentrações de PM e de carbono negro (BC) no ambiente urbano. Adicionalmente, foram realizadas campanhas de medição para validar o sistema de modelação e aprofundar o conhecimento sobre a presença de aerossóis na atmosfera urbana.

Em suma, este trabalho de investigação teve como intuito aprimorar a compreensão da poluição urbana por aerossóis, permitindo obter a contribuição do tráfego rodoviário. De facto, embora os recentes avanços da tecnologia automóvel, esta tese demonstra que, para o estudo de caso profundamente analisado, o tráfego rodoviário continua a ser uma das principais fontes de poluição do ar urbano. Além disso, as melhorias no sistema de modelação oferecem uma oportunidade inovadora para avaliar/prever a QA urbano. A análise temporal e espacial de aerossóis, juntamente com a caracterização do PM obtido por meio desta pesquisa, fornecem dados relevantes para a análise dos impactos na saúde e para a investigação sobre poluição do ar e mudanças climáticas, apoiando, assim, a tomada de decisões relacionadas ao desenvolvimento urbano sustentável.

Palavras-chave: material particulado; emissões de tráfego rodoviário; poluição do ar; aéreas urbanas; previsão; modelação.

Table of contents

List of figures

List of tables

List of abbreviations and acronyms

ADMS – Atmospheric Dispersion Modelling System
AP – Afternoon Period
AQ – Air Quality
AQM – Air Quality Models
BC – Black Carbon
BEV – Battery Electric Vehicles
CAMS – Copernicus Atmosphere Monitoring Service
CO – Carbon Monoxide
CO_2 – Carbon Dioxide
COPERT – Computer Program to Calculate Emissions from Road Transport
CSEE – cold start excess emissions
CTMs – Chemical-transport models
EC – Elemental Carbon
EEA – European Environment Agency
EF – Emission Factors
EMEP – European Monitoring and Evaluation Programme
EU – European Union
FAIRMODE – Forum for Air quality Modeling
GIS – Geographic Information Systems
HBEFA – Handbook of Emission Factors for Road Transport
HC – Hydrocarbons
HDV – Heavy Duty Vehicles
LDV – Light Duty Vehicles
LUT – Land Use Transport models
MP – Morning Period
NO – Nitrogen Oxide
NO_2 – Nitrogen Dioxide
NOx – Nitrogen Oxides
OC – Organic Carbon
O-D – Origin-Destination
PC – Passenger Cars
PM – Particulate Matter

PM_{10} – Particulate Matter with aerodynamic diameter less than 10 µm

$PM_{2.5}$– Particulate Matter with aerodynamic diameter less than 2.5 µm

PN – Particulate Number

PT – Portugal

QTraffic – Traffic Emission and Energy Consumption Model

VOC – Volatile Organic Compounds

WHO – World Health Organization

Chapter 1

1. Introduction

1.1. Context

Ambient air pollution is a primary global public health concern, causing 4.2 million premature deaths worldwide (WHO, 2022) and around half a million in European Region in 2019 (WHO, 2023), especially in urban centres where most population are exposed to harmful pollutants. Regarding that the world is becoming a “big urban centre” as 55% of world population lived in cities during 2018, which is expected to increase to 68% until 2050 (UN, 2018), the monitoring and management of urban air quality turns into an urgent matter.

In Europe, the urban air pollution improved considerably in the last decades due to implementation of several legislations and procedures. However, in many cities pollutants like particulate matter (aerosol particles) frequently exceed the limit values defined by the current European Air Quality (AQ) legislation (Directive 2008/50/EC) (EEA, 2023a). Additionally, this legislation is under review in order to be aligned with the guidelines recently defined by the World Health Organization (WHO, 2021), which imposes more restrictive limit values. Thus, Europe needs to continuously move towards a cleaner air and a massive reduction of the emissions that mostly contribute to the degradation of urban AQ.

Although the relevance of several urban emission sources (e.g. power generation, residential heating and industry), road transport stands out as one of the primary pollution sources in cities, being observed the highest pollutant concentrations in the vicinity of high traffic routes (Colvile et al., 2001; Karagulian et al., 2015). Road transport emits several atmospheric pollutants, with particular emphasis on particulate matter (PM) due to its harmful effects on human health (Kim et al., 2015; Li et al., 2003; Samoli et al., 2013), degradation of natural and built environment (Gao et al., 2002; Polissar et al., 2001) and contribution to climate change (Lohmann and Feichter, 2005; Ramanathan and Feng, 2009). Furthermore, black carbon (BC), a component of PM with known health implications (Janssen et al., 2012) and significant contribution to climate change (Szopa et al., 2021), is mainly emitted by road traffic in cities and is often used as a traffic emissions tracer (Hudda et al., 2020; Luoma et al., 2021; Reche et al., 2011).

Road transport demand continues to increase with millions of vehicles moving people and goods around Europe's road transport infrastructure (EEA, 2023b). Furthermore, most vehicles in the EU still rely on petrol and diesel, despite a shift towards electric vehicles in recent years, implicating a slower improvement of road transport sector to meet the goals set out in the European Green deal (EEA, 2023b). Besides the European Green deal, the policies to achieve a sustainable road transport and reduce vehicles emission are shaped by the Sustainable and Smart Mobility strategy (COM/2020/789), the EU emission standards and National Emissions reduction Commitments (NEC) Directive (2016/2284/EU). While the EU is making a big commitment to fostering sustainability in road transport, a substantial journey lies ahead.

The dream is a scenario with zero-emissions and consequently abate air pollution, however, it is unrealistic to speak about no air pollution among an area due to the impossibility of eliminating all anthropogenic sources and the difficulty of controlling natural emission sources. Therefore, the AQ programs, plans and strategies aims to reduce the levels of atmospheric pollutants to a point that the well-being is assured. The reduction of air pollution in large populated areas remains a challenge because it requires a substantial economic investment and changes in human behaviours and energy use. Though COVID-19 calamity, the policies implemented during the lockdown have shown the positive impact of reducing road traffic activity to the AQ in European cities (EEA, 2023c). However, the uncertainty in travel habits introduced by new realities resulted from the pandemic, e.g. teleworking and hybrid working, imposes new challenges in policy decision-making (ITF, 2023).

Policy makers, thus, face a challenging decision-making environment, exacerbated with the continuous increase of road traffic demand and introduction of new technologies like electric vehicles and digital transformation in mobility. New policy measures development and implementation should be supported by AQ assessment, which can be done both by monitoring and by modelling. Historically the AQ assessment has been made by monitoring. Nevertheless, AQ modelling allows to overcome the limited spatial representativeness that point measurements presents and enables AQ forecasting. AQ forecast has gained an increasing research interest. However, in most cases the AQ forecast has been made at the global or regional level, with only few studies at urban scale. Although the rarity of urban-scale air quality forecast, it is very important for population exposure, short-term mitigation and prevention of severe air pollution episodes.

Therefore, the thesis work addresses urban air quality focusing on contribution of road traffic recognized as one of the main sources of urban air pollution, especially looking on aerosol particles and its harmful black carbon component. The final goal of this work was to implement an air quality modelling system in assessment and forecast modes that required new developments and improvements of the previously available system. For this purpose, several tasks were developed, such as the improvement of the algorithm used to quantify road traffic emissions by including cold start and non-exhaust emissions. Additionally, the thesis work focused on a novel approach to obtain urban background concentrations for forecasting purposes from available regional scale services based on earth observations and atmospheric modelling. Furthermore, measurement field campaigns were implemented to increase the knowledge on aerosol particles in the urban atmosphere and to provide data to validate the air quality modelling. In order to test the usability and the performance of the modelling system, a case study was applied in a Portuguese city (Coimbra), where also the measurements were obtained.

1.2. Research gaps and objectives

In order to define the objectives of the work, an exhaustive literature review was made to first identify the research gaps taking into account air quality management priorities in urban areas.

As part of AQ management, AQ assessment can be made by monitoring and by modelling. However, both approaches reveal some advantages and limitations. In general, measurements from monitoring stations provide data only on mass concentration of PM_{10} and $PM_{2.5}$. However, in order to be able to study urban aerosol dynamics and the potential health effects of aerosol particles, there are other important metrics that should be considered such as the number and surface area of particles, aerosol size distribution. Moreover, point measurements have a limited spatial coverage and representativeness.

Given the complexity of urban air quality, modelling results involve several uncertainties which must be reduced particularly when used to support decision making and urban plans. Some uncertainties are related to simplifications considered in the modelling approach. For example, the dispersion of pollutants in the presence of obstacles can be modelled based on more simplistic approach using Street Canyon formulation, or complex approach using Computational Fluid Dynamic techniques. However, in many cases given the data available, the simplistic models may be sufficient to fulfil the objectives of the study. Furthermore, the simplistic approaches have also the advantage of a faster running time. Besides, the uncertainty can be also related to the input data and their processing. For instance, most of the inventories of traffic emissions used in air quality research do not consider non-exhaust emissions. Nowadays, given the relative reduction of exhaust emissions, methods to quantify non-exhaust emissions becomes very important. However, there is still a lack of knowledge in this field.

Another issue regarding the traffic emissions data is the cold start emissions. In many studies focused on emission modelling, only the hot emissions are usually considered. However, the majority of urban trips are short and made with the engine at the transient 'warming-up' phase. Therefore, the cold start emissions inventory is crucial for urban air quality research. The difficulty of the cold start emissions calculation lies in the question: How to estimate and implement the trip distance in cold start regime for each pollutant species? Regarding this, further developments must be done to include the cold start distances.

Additionally, AQ forecast has gained recognition in the last decades in the research works, taking into account that it provides the quantification of pollutants concentrations for a few days in advance and set alarms, which allows to work on air quality impact prevention instead of mitigation only. However, AQ modelling with high resolution is a challenge. Currently, the number of published studies focused on urban air pollution forecast are very few. Hence, the advance in simulation of urban air pollution must be rooted in methods that support forecasting.

Therefore, the main goal of the thesis was to implement an AQ modelling capable of assessment and forecasting aerosol particles in the urban atmosphere, by considering road traffic as one of the main contributors to urban air pollution. In order to improve the characterization of the urban aerosol, modelling techniques was combined with aerosol measurements. The following key objectives were defined:

a) To improve a traffic-related emission modelling (previously focused on hot exhaust emissions) in order to incorporate cold start and non-exhaust emissions. Therefore, developments in terms of the algorithms of emission models and how they integrate road traffic data were made;
b) To explore available regional scale services based on Earth observations and Atmospheric modelling and to evaluate their applicability to urban scale air pollution forecast. Moreover, a new approach was developed to incorporate the background concentrations from the regional scale service into urban air quality dispersion model;
c) To improve characterization of urban aerosol number, size distribution and composition, especially focused on PM components like Black Carbon (BC).

Several aerosol measurements were performed within the urban area to gather data and validate the integrated modelling approach.

To fulfil the previously outlined objectives, this thesis combines the implementation of modelling and aerosol measurements. The measurements conducted throughout various stages of this thesis primarily served to validate the modelling system. The modelling system developed in previous research (Dias et al., 2016, 2019a) was considered as a starting point to implement new developments addressed in the current thesis as illustrated in Figure 1.1. The modelling system was enhanced to incorporate quantification of cold start emissions non-exhaust emissions from road traffic. Furthermore, an innovative methodology was developed to incorporate background contribution from regional-scale services, allowing the prediction of air pollution at an urban scale. Additionally, the system was expanded to incorporate the emissions and dispersion modelling of Black Carbon, a component of atmospheric aerosol with impacts on human health and climate.

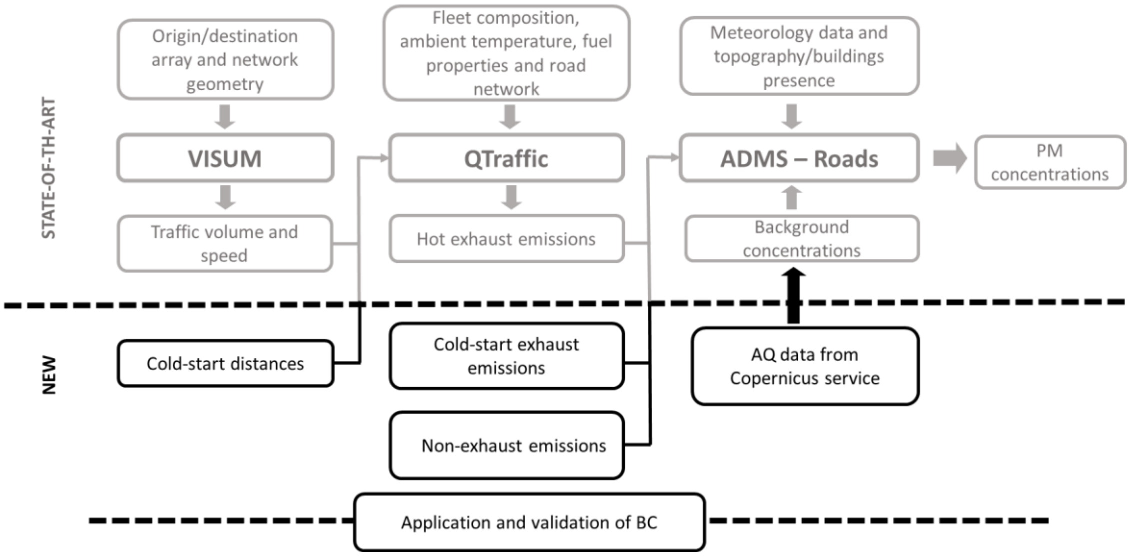


Figure 1.1 Modelling system framework.

1.3. Thesis structure

This thesis is divided into 7 chapters, where Chapters 1 and 7 are the overall introduction and the final remarks of the thesis, respectively. Chapter 2 presents a general overview of road traffic issues and air pollution in order to contextualize the research studies developed in the following chapters. The research developed in this work is structured as journal publications and presented from Chapters 3 to Chapter 6, allowing their reading to be independent. However, all these studies were focused on Coimbra and some information about the city could be repeated in different chapters.

Chapter 3 presents the new developments made to implement origin/destination information from the traffic flow model to address the cold start distances and quantify cold start emissions. This study shows the importance of handling cold start emissions at a road link level and distributed within the cold distance, which allowed to understand the spatial distribution of travels in different daily periods and seasons and at an urban context.

Chapter 4 deals with the development and integration of new module to quantify non-exhaust emissions into the modelling approach, which allowed to access the contribution of non-exhaust emissions to atmospheric aerosols. An integrated modelling approach was applied to a Portuguese mid-sized city. Moreover, the importance of fine particles to urban aerosol pollution is demonstrated in this chapter using PM measurements and taking into account particles number and size distribution.

Chapter 5 is focused on the evaluation of different regional scale models and services in order to obtain urban background concentrations and combine it with the integrated modelling approach to assess/forecast PM concentrations at the urban scale. Thus, this research aimed to compare the Portuguese national forecast service based on the CHIMERE model and the European CAMS service for which the ENSEMBLE product were accessed. Both approaches were possible to couple with the integrated modelling system, being further validated with *in-situ* observations.

Chapter 6 is focused on studying BC, a component of PM that imposes human health negative impacts and significantly contributes to the climate change. Thus, this research aimed to evaluate the contribution of road traffic to BC pollution levels at urban scale and validate the modelling approach using observations collected during 6-months field campaign. For this purpose, new developments were required in order to extend the modelling system to BC and to obtain traffic-related concentrations and emissions including cold start, hot exhaust and non-exhaust processes.

Finally, in Chapter 7 a brief summary of the main achievements, some final remarks on the developed work and future lines for the research are presented.

1.4. Publications

This thesis is based on the work contained in original journal publications. The list of publications, including the contribution of Noela Pina for each of them, is presented below.

- **Pina, N.** and Tchepel, O. (2023). A bottom-up modeling approach to quantify cold start emissions from urban road traffic. *International Journal of Sustainable Transportation, 17*(8), 942-955.

  Cold start emissions may contribute significantly to urban road traffic emissions. However, in several studies they are addressed in an aggregated manner or not included. Hence, this publication surges with the aim of implementing a bottom-up approach for emission quantification in urban context to address the distribution of pollution with fine temporal and spatial resolution and to establish local mitigation measures and plans. In this research a modelling approach to quantify cold start excess emissions using the concept of cold distance, is proposed and applied. In combination with transportation modelling, a new module was developed to estimate cold start emissions at road segment level and for a trip, while preserving information on the Origin-Destination of trips contributing to those emissions. The methodology was applied to Coimbra case study. Unlike hot exhaust emissions, the area that extends beyond the study area is crucial for calculating cold start emissions since they decrease during a trip as an engine heats up. Thus, the implementation of the methodology took into account the whole Region area of Coimbra, not only the urban area. Finally, the contribution of cold start emissions to the total exhaust emissions and their spatial distribution were accessed for the case study, finding out the importance of cold start emissions at local roads, where most people leave or work. The methodology developed and applied by the first author highlights the importance of cold start emission quantification for traffic pollution studies. The preliminary results of this study were disseminated at the 23rd International Transport and Air Pollution Conference which allowed the authors to refine the methodology. Furthermore, such methodology was further used in the last submitted publication allowing to access the cold start contribution to BC concentration levels within urban areas.

- Dias, D**, Pina, N.**, Tchepel, O. (2019). Characterization of traffic-related particulate matter at urban scale. *International Journal of Transport Development and Integration, 3* (2), 144-151.

This work aimed to provide a detailed characterization of traffic-related PM concentrations at urban scale by using an integrated modelling approach *and in-situ* aerosol measurements. For this purpose, a modelling cascade based on transportation-emission-dispersion approach was implemented for Coimbra city. Moreover, optical aerosol measurements were obtained from an experimental field monitoring campaign implemented at a city hotspot to provide relevant *in-situ* data on number, surface and mass concentrations distribution into 31 sizes. This was the first journal publication that N.Pina have done in collaboration with other authors involved in the TRAPHIC project, for which she was responsible of the *in-situ* observations: gathering, treating and analysing the data obtained. Furthermore, N.Pina prepared the traffic activity data for the whole urban area in GIS environment, which was crucial for both emission and air quality modelling, in a way of overcoming some limitations of the air quality tool used. Additionally to being a part of the preparation of the field campaign of the TRAPHIC project, fundamental to this research, N.Pina disseminated the results of this research in two international conferences (*1ª Conferência Internacional de Ambiente em Língua Portuguesa*, 25th International Conference on Urban Transport and the Environment).

- Tchepel, O., Monteiro, A., Dias, D., Gama, C., **Pina, N.**; Rodrigues, J.P.; Ferreira, M.; Miranda, A.I. (2020). Urban aerosol assessment and forecast: Coimbra case study. *Atmospheric Pollution Research, 11*(7), 1155–1164.

The primary objective of this study was to explore existing services at a regional scale and study their applicability to assess/forecast urban AQ. Two approaches were evaluated to characterize urban background pollution levels from regional operational forecasting: (i) Copernicus Atmosphere Monitoring Service (CAMS) and (ii) Chemical Transport Model CHIMERE from the national forecasting system. The local contribution of road traffic was analysed using the integrated modelling approach with 10 m grid resolution. The methodology was applied to the city of Coimbra to estimate PM concentrations for a period of one year. This research is a product of the ISY-AIR project funded by MIT, which provided N. Pina an opportunity to collaborate with different prominent air quality research groups. Within this collaborative effort, N. Pina played an important role. Her contributions encompassed conducting an exhaustive literature review integral to this study and, more significantly, the implementation of the modelling system at urban scale. This involved integrating the background concentrations obtained from regional scale services and subsequently validating the modelling outcomes with observations from QualAr. The findings of this study were disseminated through presentations at several international conferences and during the ISY-AIR workshop.

- **Pina, N.**, Almeida, S. M., Alves, C., Tchepel, O. *(Submitted).* Contribution of road traffic to black carbon aerosols at city level – validation of the modelling approach. *Atmospheric Environment.*

  Black carbon (BC) is a harmful pollutant to human health and the environment. The aim of this paper was to evaluate the contribution of road traffic to BC pollution levels at urban scale by implementing the integrated modelling approach to quantify BC traffic-related emissions and concentrations in Coimbra city, with high spatial and temporal resolution. Hence, the Traffic Emission and Energy Consumption Model (QTraffic) was extended in order to include BC emission factors considering road network as line emission sources, thus providing the necessary information for the assessment of BC pollution levels in the urban environment. The modelling approach proposed and implemented in this work was validated with observations collected during a six-month monitoring campaign at an urban traffic station using gravimetric measurements for elemental carbon (EC) and combustion-derived BC concentrations from an aethalometer. This investigation is also an ISY-AIR project result, which allowed N. Pina to participate in an exhaustive measurement campaign, being responsible for collecting data every two days and ensuring the proper functioning of the equipment throughout the 6-month campaign. N. Pina has also conceived the modelling approach and simulations, analysed the data, and drafted and wrote the paper. Preliminary results of this study were presented in the 13th International Conference on Air Quality – Science and Application.

Although not contributing directly to this thesis, the work undertaken in the framework of the thesis research and different research projects (TRAPHIC and ISY-AIR), resulted in further publications:

- Pio, C., Rienda, I. C., Nunes, T., Gonçalves, C., Tchepel, O., **Pina, N. K**., Rodrigues, J., Lucarelli, F., Alves, C. A. (2022). Impact of biomass burning and non-exhaust vehicle emissions on PM10 levels in a mid-size non-industrial western Iberian city. *Atmospheric Environment, 289*, 119293.
- Gonçalves, C., Rienda, I. C., **Pina, N**., Gama, C., Nunes, T., Tchepel, O., Alves, C. (2021). PM10-bound sugars: chemical composition, sources and seasonal variations. *Atmosphere, 12*(2), 194.

Chapter 2

2. Road traffic and air pollution: an overview

An overview of traffic-related air pollution and air quality managements is presented in this chapter. Additionally, the literature review on different modelling tools (transportation, emission and urban AQ dispersion models) is also presented to identify the research gaps. Additionally, key concepts are addressed to facilitate the reading of the following chapters.

2.1. Traffic-related air pollution

Transport is one of the major source of air pollution around the world. In Europe, the contribution of transport sector to the total emissions can vary from 9% to 55% depending on the pollutant (EC, 2022a), of which road traffic may achieve 86% (Figure 2.1). The contribution of road traffic is particularly important since it emits large amounts of harmful chemicals close to populated areas. In fact, in urban areas road traffic is the dominant anthropogenic source of air pollution (Colvile et al., 2001; Karagulian et al., 2015). The emissions induced by transport have significantly improved in EU since 1990s due to the technological measures implemented. Nevertheless, we can expect that the road transport will still remain one of the important sources of pollutions in European urban areas during next years, because the reduction of road traffic emissions is at least partially offset by traffic growth and elevated congestion levels within the urban areas, even thought that electric mobility will play a key role in reducing some negative impacts.

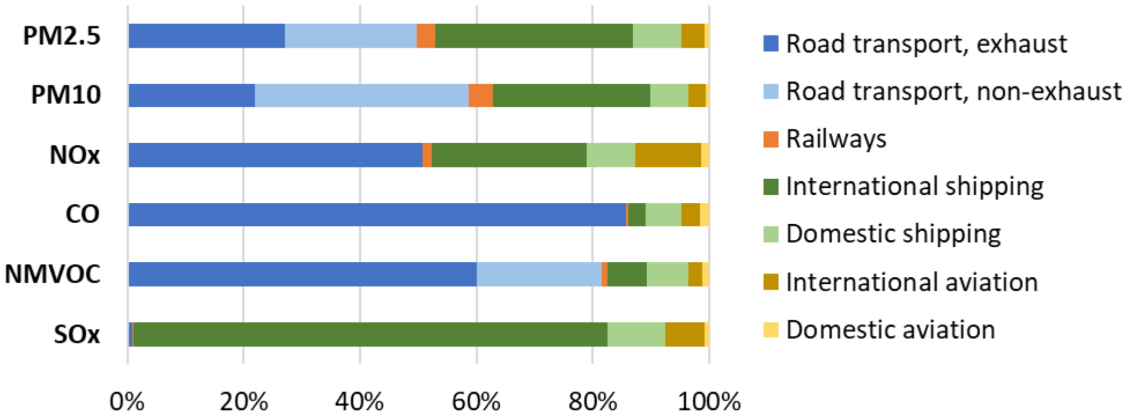


Figure 2.1 Split of transport emissions of the main air pollutants considering different modes (adapted from EEA, 2019a).

2.1.1. Road transport demand in Europe

The road transport demand can be analysed in terms of freight and passengers. Figure 2.2 presents the evolution of both demands. Overall, road freight transport grew in the EU-27 from 1995 until 2019 (57%). However, this growth has been affected by the economic recession from 2009 to 2012 and recently COVID-19 conducted to a slightly reduction of 1% in 2020 (in comparison to 2019 demand; EC, 2017, 2022a). Such reduction has been already overpassed to values higher than 2019 in 2021 (Eurostat, 2023).

In comparison to road freight transport, the demand of road passenger transport suffered a lower growth in the EU-27 from 1995 to 2019 (27%), also being affected by the economic crisis and COVID-19 pandemic (EC, 2017, 2022a). However, the COVI-19 affected strongly the demand on road passenger transport, registering a decrease of 19% when compared to 2019 values, which have already increased since then (Eurostat, 2023). Moreover, between 1995 and 2019, the passenger demand modal split was dominated by passenger cars (increasing from 85 % to 88 %), followed by buses and coaches (decreasing from 12 % to 10 %) and powered two-wheelers (decreasing from 3% to 2%) (EC, 2022a). The passenger cars dominance is related to the gradually decrease of prices for car purchasing in relation to average consumer prices, which enabled people to afford a car (EC, 2017). In 2020, the passenger cars dominance increased to 90% and bus and coaches decreased to 7%, reflecting the impact of the COVID-19 crisis on the use of transport in general, especially on public transport (EC, 2022a). Among the EU Member States, Portugal occupied the second place in the ranking of countries with highest shares of inland passenger transport for passenger cars (Eurostat, 2022).

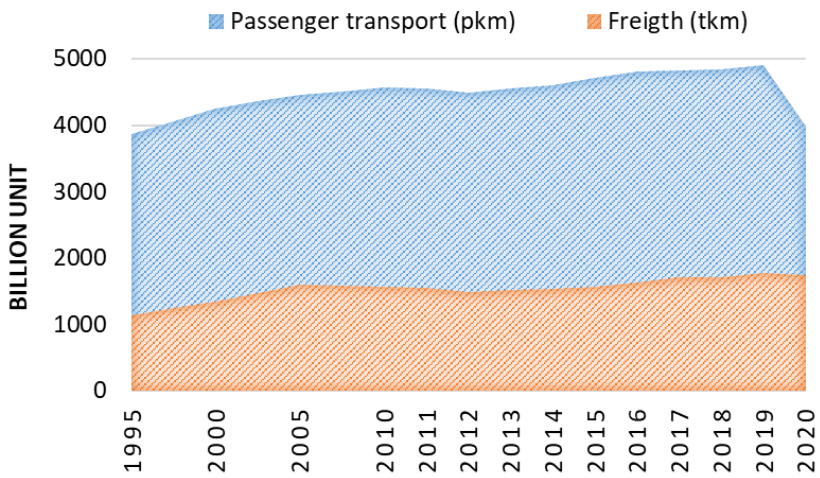


Figure 2.2 Passenger (billion pkm) and freight (billion tkm) road transport demand in EU-27 (adapted from EC, 2022a).

2.1.2. Road traffic emissions

Road vehicles emits several greenhouse gases and air pollutants, which may be divided into two groups: regulated and unregulated pollutants (EEA, 2016). The regulated pollutants under EU emission standards are:

- **Carbon dioxide (CO_2)**, which is the principal product of fuel combustion and the most important greenhouse gas influencing climate change.
- **Hydrocarbons (HC)**, which are a result of incomplete or partial combustion, being very toxic to human health. HC, specially the volatile organic compounds (VOC), contribute to the formation of photochemical smog (which irritates the eyes, damages the lungs and aggravates respiratory problems).
- **Carbon monoxide (CO)**, which is also a product of incomplete combustion, occurs when the carbon in the fuel is only partially oxidized, forming CO and not CO_2. When directly exposed to this pollutant the oxygen flow in the blood stream is reduced. This pollutant is particularly dangerous for people with heart disease and also contributes to the formation of ozone at ground level.
- **Particulate matter (PM)**, is one of the most dangerous pollutants to the human health as it penetrates into sensitive regions of the respiratory system, and may cause or aggravate cardiovascular and lung diseases and cancers. While primary PM is a product of incomplete combustion and abrasive processes, secondary PM origins in the atmosphere following the emission of precursor gases (mostly sulphur dioxide (SO_2), nitrogen oxides (NO_X), ammonia (NH_3) and some VOC).

- **Nitrogen oxides (NO_X)**, is a group of different chemicals comprised by nitric oxide (NO) and nitrogen dioxide (NO_2), which result from the reaction between nitrogen and oxygen in the combustion air and the oxidation of nitrogen in the fuel. Contrary to CO and VOC, NO_X is higher near the stoichiometric ratio in vehicle engines (where combustion is more efficient), since its formation occurs in the presence of high temperatures. Thus, measures typically used to control CO/VOC by increasing combustion efficiency increases NO_X; and the vice versa. Although causing harm to the environment by contributing to the acidification and eutrophication of waters and soils, NO_X emissions also lead to the subsequent formation of secondary PM and ground-level ozone in the atmosphere.

The unregulated pollutants induced by vehicles comprises a wide list of pollutants, including: acidifying pollutants (e.g. SO_2); carcinogenic and toxic organic pollutants (e.g. polycyclic aromatic hydrocarbons (PAHs), persistent organic pollutants (POPs), dioxins and furans); and heavy metals (e.g. lead, arsenic, cadmium, copper, chromium, mercury, nickel, selenium and zinc). Additionally, Black carbon (BC), a component of PM, is also an important unregulated pollutant for both air quality and climate change issues.

The road traffic emissions result from different processes (see Figure 2.3), which enable to classify them into:

- **Exhaust emissions** (e.g. CO; CO_2; NOx; HC; VOC; PM): the emissions that result from fuel combustion. In a perfect engine the only products of combustion would be water and CO_2. However, there is no such engine, therefore several extra pollutants result from the combustion process. The amount of the pollutants emitted depend of the fuel used, vehicle technology and age. Furthermore, when the vehicle is running in a cold regime (i.e. engines and catalysts are not (fully) warmed up) the emissions are higher.
- **Abrasion emissions** (e.g. PM): the emissions that result of mechanical abrasion and corrosion of vehicle components (tyres, brake and clutch) and road pavement abrasion. Abrasion is only important for PM emissions and some heavy metal emissions.
- **Evaporative emissions** (e.g. VOC): the emissions that result of losses of vapours through the vehicle's fuel system. They are only important for VOC, which can be emitted any time there is petrol fuel (which is a mixture of different hydrocarbons) in the tank, even when the engine is turned off.

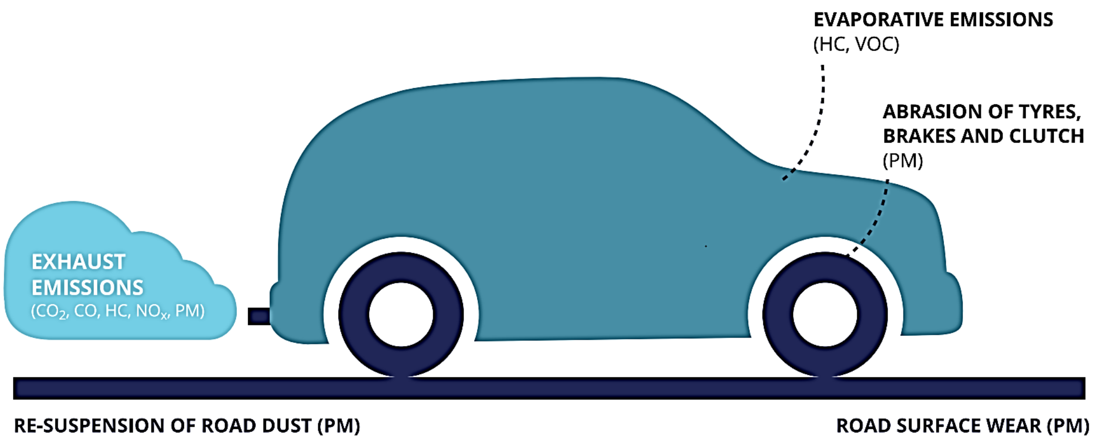


Figure 2.3 Different types of road transport emissions (adapted from EEA, 2016).

The vehicle's emissions have been regulated through the settings of emission limits since 1970's. The first European Council Directive to control and check exhaust vehicle emissions was published in 1970 and 22 years later the emission standards were introduced (Euro 1), followed by successively stricter standards (Euro 2 to Euro 6) (EEA, 2016), see Table 2.1. The date that each Euro standard was implemented depends of the fuel type and the vehicle category. Currently, the new Euro 7 standard have been proposed (EC, 2022b), which besides have stricter limits for exhaust emissions also impose a limit for brake PM emissions (applying 0.007 g/km until 31/12/2034 and 0.003 g/km after).

Table 2.1 Evolution of Euro standards for emissions from passenger cars (EEA, 2016; EC, 2022b).

Euro standard	Fuel	Date	CO (g/km)	NMHC* (g/km)	NO_X (g/km)	HC + NO_X (g/km)	PM (g/km)	PN** (#/km)
Euro 1	**Diesel**	07/1992	2.72	–	–	0.97	0.14	–
	Petrol	07/1992	2.72	–	–	0.97	–	–
Euro 2	**Diesel**	01/1996	1	–	–	0.7	0.08	–
	Petrol	01/1996	2.2	–	–	0.5	–	–
Euro 3	**Diesel**	01/2000	0.64	–	0.5	0.56	0.05	–
	Petrol	01/2000	2.3	–	0.15	–	–	–
Euro 4	**Diesel**	01/2005	0.5	–	0.25	0.3	0.025	–
	Petrol	01/2005	1	–	0.08	–	–	–
Euro 5	**Diesel**	09/2009	0.5	–	0.18	0.23	0.005	6.0×10^{11}***
	Petrol	09/2009	1	0.068	0.06	–	0.005	–
Euro 6	**Diesel**	09/2014	0.5	–	0.08	0.17	0.005	6.0×10^{11}
	Petrol	09/2014	1	0.068	0.06	–	0.005	6.0×10^{11}
Euro 7 (proposed)	**Diesel**	07/2025	0.5	0.068	0.06	–	0.0045	6.0×10^{11}
	Petrol	07/2025	0.5	0.068	0.06	–	0.0045	6.0×10^{11}

*NMHC – Non-methane hydrocarbon; **PN (Particulate Number); ***Introduced in September 2011.

The quantification of vehicles emissions is an important issue and can be made by emissions modelling or by emissions measurements. Measurement of vehicles emissions may be done through controlled situations in the laboratory or through actual driving conditions. On the other hand, the estimation of these emissions is a complex and difficult task, as they are a function of several parameters that are usually interrelated (Smit et al., 2009). First, fuel consumption and emissions varies significantly with vehicles characteristics, such as: size and weight; engine type; type of fuel consumed; presence and type of emission control technology; presence of auxiliary equipment (e.g. air conditioning); aerodynamic characteristics. Additionally, the emissions and fuel consumption of a vehicle are also affected by the behaviour of the driver, which is a result of many factors such as: personal driving style; weather conditions; level of congestion; road characteristics (e.g. width, gradient; pavement conditions; presence of intersections and curves; speed limit; etc.). At least, but not less important, the level of vehicle emissions is dependent of the driving mode (running, idling), fuel characteristics, ambient temperature (which just influence the emissions with cold engine), age, maintenance and tuning of the vehicle. Furthermore, several factors such as occupancy rate, vehicle load and number of axes are also crucial for heavy vehicles.

Regarding that the research objectives of this thesis are mostly focused on traffic-related PM, more information on this pollutant is provided. Thus, PM emissions from road vehicles comprise exhaust and non-exhaust processes (i.e. abrasion emissions). While, the exhaust emissions contribute predominantly to fine PM (Diameter of particles (Dp) < 2.5 µm), the non-exhaust emissions contribute mainly to the coarse fraction of PM (Dp > 2.5 µm) (Pant and Harrison, 2013). However, according to Narváez et al. (2008), abrasion can contribute significantly to fine particles too.

Old vehicles with diesel engines emitted a greater PM mass and a large number of ultrafine particles when compared to petrol cars (Rose et al., 2006). However, the situation in relation to exhaust emissions from traffic have improved considerably with the abatement strategies and technology development. For instance, the use of diesel particulate filters reduced substantially the mass of exhaust emissions.

The continuous reduction of PM exhaust emissions, poses new worries to PM from non-exhaust processes. Although their important contributions to total PM, including also a relevant contribution to BC particles, they are still unregulated. Therefore, information on non-exhaust emissions are sparse and highly uncertain due to different quantification approaches and many complex variables (e.g. vehicle class, environmental/meteorological parameters, driving styles, vehicle speed, acceleration/deceleration, vehicle mass, brake/tire/road material compositions) (Fussell et al., 2022).

2.1.3. Measures towards less transport emissions in Europe

Dating to the first EU emission standards introduced (see previous topic) and the adoption of the White Paper on the future development of the common transport policy (COM(1992)0494), the transport emission reduction has become one of the main points of transportation plans and strategies. The introduction of the EU emission standards resulted in vehicle technologies to control traffic emissions, such as the direct fuel injection in petrol vehicles (to reduce fuel consumption), start-stop systems (prevents waste of fuel when the vehicle is stopped) and the diesel particle filters (used with diesel engines to remove PM). Additionally to technological solutions, policy measures to reduce the traffic emissions have been applied, such as the implementation of low-emission zones or congestion charges and the incentive to use soft modes (e.g. bicycles, electric scooters). Moreover, internalization of the external costs of transport is also an important way to give users the right incentives towards a ‘cleaner’ transport sector. Hence, the 2011 Transport White Paper recommend fair and efficient price signals in the transport sector. In fact, it is interesting to notice that from 2018 to 2022 the share of electric vehicles in the new passenger cars among EU have increased significantly (Figure 2.4). This is a consequence of different initiatives, including tax benefits implemented to purchase electric vehicles. On 2023, 20 EU member states offer incentives for purchasing electric cars, while 7 countries do not provide any purchase incentives; however, most of them grant tax reductions or exemptions (ACEA, 2023a). Portugal (PT) offers incentives for private users to buy a new battery electric vehicles - BEV (M1 vehicle, €3000 with purchasing price of up to €62500 limited to one vehicle per person), acquisition car taxes exemption (100% for BEV, 75% for plug-in hybrid vehicles and 40% for hybrid electric vehicles) and exemption for BEVs ownership.

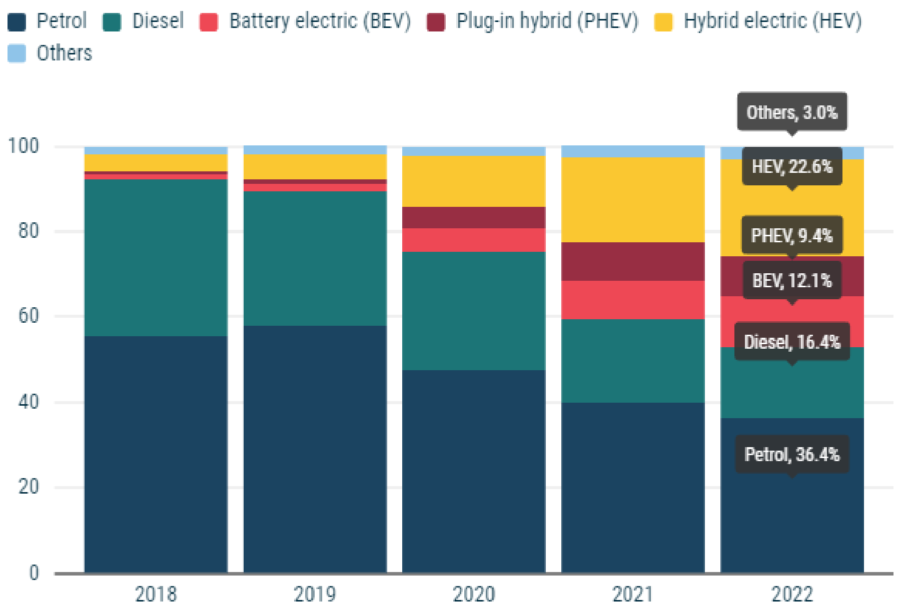


Figure 2.4 Share of new passenger cars by fuel in EU from 2018 to 2022 (ACEA, 2023b).

Despite the recent increase in sales, electrically-chargeable cars represent only 1.5% of the total EU car fleet and only three countries have a share of battery electric cars higher than 2% (ACEA, 2023c). Actually, road transport (passenger cars) continues to rely heavily on fossil fuels, especially petrol (EU – 51%; PT – 37%) and diesel (EU – 42%; PT – 59%) (ACEA, 2023c). Moreover, it is important to understand that not only the end product (e.g. electricity, biofuels) determines how environmentally sustainable a fuel is. The production process is also very important and should be taken into account. For instance, the electricity produced by wind turbines is much cleaner than electricity produced by coal. Therefore, transport's demand for energy and their related emissions can be best addressed through a comprehensive analysis and a holistic vision. This is a crucial view when the issue is decarbonisation of mobility by considering the carbon footprint of the whole life cycle of a transport mode choice. However, when the issue is local air pollution we may look to the impact of vehicle use in the specific location, understanding that the so-called “zero-emission vehicles” will always induce emissions to the atmosphere (e.g. non-exhaust PM emissions due to the abrasion of vehicle components and road surface and resuspension of deposited particles).

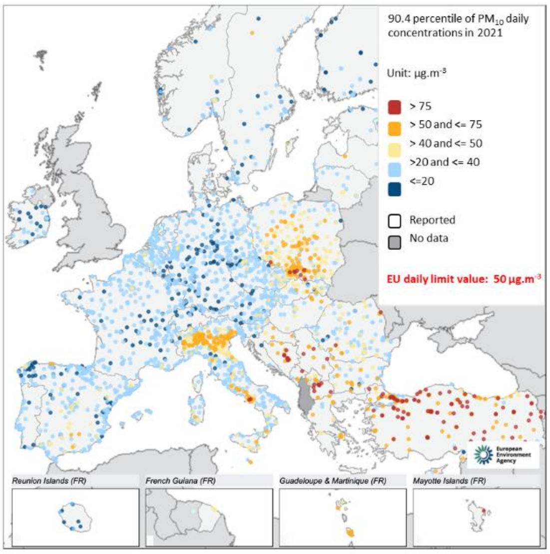
In spite of the continuous reduction of traffic-related emissions verified in Europe, the increase of mobility within cities due to urban growth and inefficient land use organization led to accessibility and congestion problems, which are in the core of the remaining traffic emission issue. All around Europe, transportation concepts such as carpooling and car sharing has been implemented to reduce road congestions and external impacts. Furthermore, parking policies have become an integral part of modern urban planning strategies due to their impact on mode choice, travel routes and traffic emissions (Pitsiava–Latinopoulou et al., 2012). For instance, less parking space exacerbates transport emissions since drivers tend to move in circles in order to find an empty space to park their vehicles. Moreover, automation (from driver assistance to full automation) in both freight transport and shared urban vehicles has the potential to reduce associated environmental impacts. It is generally expected that autonomous vehicles could operate with higher fuel efficiency compared to traditional vehicles, though uncertainties are still very high and digitalization may be the key to these results (EEA, 2022b). According to the EEA (2022b), digitalization may potentially impact a number of transportation system aspects, including the demand and supply of transport services, the effectiveness of using the available transportation infrastructure's capacity, the environmental performance of vehicles, and the quantity and sources of energy used for transportation. Additionally, digitalization can support in improving transportation policies, considerably influencing the transportation impacts on environment.

In conclusion, given the efforts that have been done, traffic pollutants emissions are expected to continue their reduction across Europe, with positive impacts on human health. Nevertheless, the proportion of city dwellers in EU exposed to levels of air pollutants considered harmful by WHO during 2021 is 97%, 76%, 90% and 94% for $PM_{2.5,}$ PM_{10}, NO_2 and O_3, respectively (EEA, 2023a). Therefore, Europe will need to further integrate policy measures and actions to reduce air pollutant emissions and create the conditions for better health and wellbeing of European citizens. Reducing traffic pollutant emissions could certainly help improve air quality, in urban areas in particular.

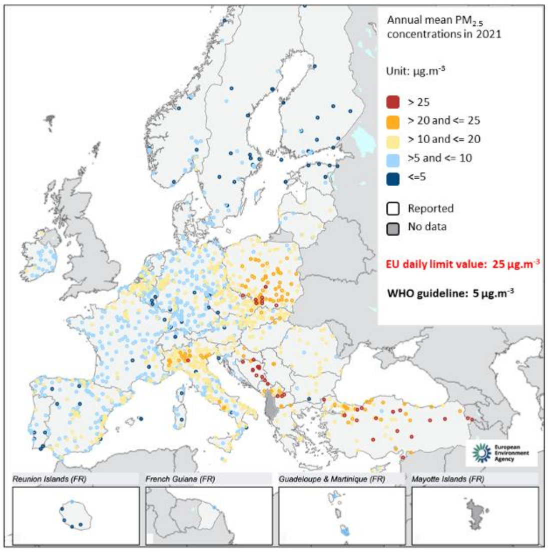
2.2. Air quality management

Since the last decades, the presence of pollutants in the atmosphere have been decreased in Europe. However, in many cities the level of concentration of pollutants continues to exceed the European AQ limits, as we can see from the monitoring station results in Figure 2.5. In order to improve the air quality, management practices must be implemented. The AQ management consists in a combination of assessment and planning (Figure 2.6). The AQ assessment is based on monitoring (by measurements) and modelling. Moreover, air quality measurements are used to validate simulations, which are used as a decision support to AQ planning. The AQ planning includes both long-term air quality plans (from now referred as AQ planning) and short-term air quality action plans in regard to exceedances of alert thresholds sustained by AQ forecasting.

a) PM10 concentrations



b) PM2.5 concentrations



c) NO2 concentrations on Europe

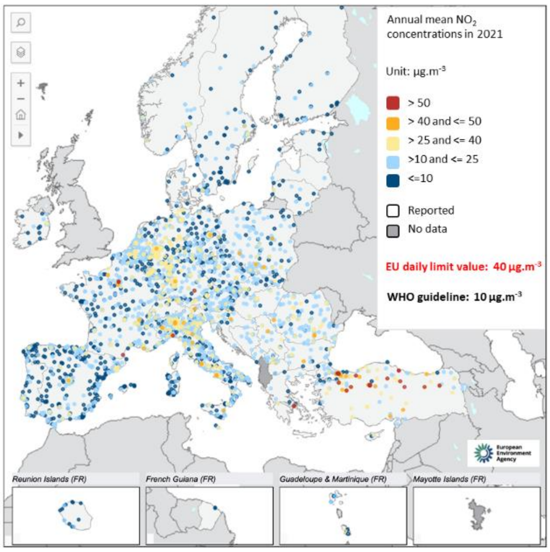


Figure 2.5 Annual mean values observed at monitoring stations in Europe in 2021 (EEA, 2023a).

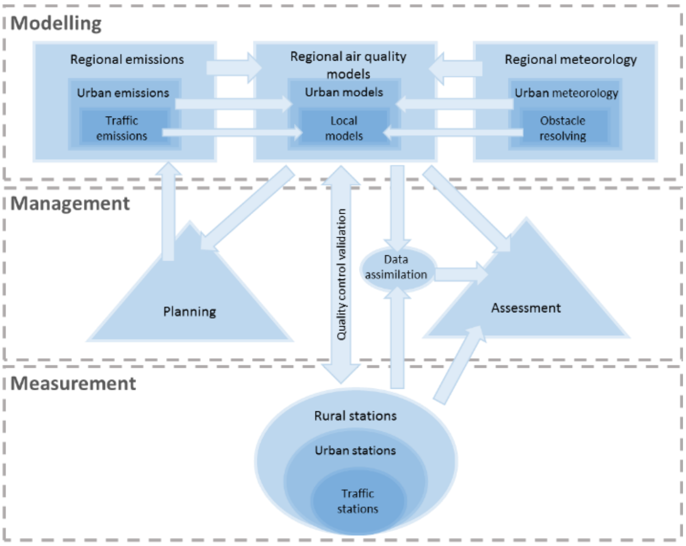


Figure 2.6 Schematic representation of air quality management and the connections with modelling and monitoring activities (Denby, 2011).

2.2.1. Air quality legislation

Actions to manage and improve air quality are largely driven by European (EU) legislation, which has been evolving since 70's (see Figure 2.7).

Figure 2.7 Evolution of the air quality legislation in Europe.

According to the current AQ directive (Directive 2008/50/EC) the main purpose of ambient air quality (AQ) control in European Union is “to reduce pollution to levels which minimize harmful effects on human health, paying particular attention to sensitive populations, and the environment as a whole”. Therefore, different air quality indicators are established for specific pollutants: particulate matter, nitrogen dioxide and oxides of nitrogen, sulphur dioxide, carbon monoxide, ozone, lead and benzene. These pollutants have been selected taking into consideration different environmental issues and challenges, including climate change; ozone layer depletion; acidification; toxic contamination; urban air quality; and traffic air pollution (Sivertsen, 2008). For air pollution induced by traffic, the most frequently selected pollutants are: nitrogen dioxide; carbon monoxide; and particles with aerodynamic diameter less than 10 µm (PM_{10}) and less than 2.5 µm (and $PM_{2.5}$). Overall, these pollutants are controlled given the limits established in the current AQ directive. Recently, the European Commission undertake a comprehensive revision of the current AQ directive, under the European Green Deal’s zero pollution pillar, with the aim of better aligning EU air quality standards with the World Health Organization (WHO) recommendations from 2021 (EP, 2023). In light of these endeavours, Table 2.2 systematically presents the limit values and targets assigned to traffic related air pollutants, considering the limit values under the current AQ directive, the WHO AQ guidelines and the proposed revision.

Table 2.2 EU AQ standards (current and revised) for the protection of human health and WHO AQ guidelines.

Pollutant	**Averaging period**	**Limit values**		
		Current standard	**Proposed standard**	**WHO guidelines**
$PM_{2.5}$	Annual	25 µg.m^{-3}	10 µg.m^{-3}	5 µg.m^{-3}
	24-hour	-	25 µg.m^{-3} **	15 µg.m^{-3} ***
PM_{10}	Annual	40 µg.m^{-3}	20 µg.m^{-3}	15 µg.m^{-3}
	24-hour	50 µg.m^{-3} *	45 µg.m^{-3} **	45 µg.m^{-3} ***
NO_2	Annual	40 µg.m^{-3}	20 µg.m^{-3}	10 µg.m^{-3}
	24-hour	-	50 µg.m^{-3} **	25 µg.m^{-3} ***
CO	24-hour	-	4 mg.m^{-3} **	4 mg.m^{-3} ***
	Max. daily 8-hour mean	10 mg.m^{-3}	10 mg.m^{-3}	-
Exceedance limit: * 35 times/year; ** 18 times/year; *** 3-4 days/year.				

2.2.2. Air quality assessment

The AQ assessment can be made by monitoring or modelling, or even by the combination of both methods. Actually, the Directive 2008/50/EC encourages combining both methods to allow the determination of pollutants’ spatial distribution and reduction of the required minimum number of fixed sampling points. Furthermore, according to the new AQ revision, in zones where pollutant levels exceed the established limits, fixed measurements will be required, while where the limits are not exceeded modelling applications, indicative measurements, objective-estimation techniques, or a combination of them will be sufficient (EP, 2023).

a) Air quality monitoring

AQ monitoring provides information regarding the status of present air quality and helps in evaluating the existing policies and their effective implementation (Gulia et al., 2015). The monitoring is usually performed using fixed stations, whose location allows to represent the population exposure and to see if the limit values imposed by the AQ directive are met.

Monitoring stations can be classified according to location and proximity to emission sources (Directive 2008/50/EC), as follows:

- Location:
  - Urban stations, which are located in urban areas;
  - Suburban stations, which are located in urban residential areas, however remote from the urban centre;
  - Rural stations, which are located far from urban centres and away from urban pollution sources.
- Proximity to emission sources:
  - Traffic stations, which with the purpose to measure the traffic-related contribution to pollutants concentration, are placed near the main highways;
  - Industrial stations, which are located near industrial areas, in a way that the industries are the main pollution source;
  - Background stations, which enable to assess the air quality farthest from pollution sources, i.e., they are not associated with any specific source of pollution but with the combination of all sources to windward of a given area.

There are several techniques and instruments used to measure pollutants, which depends on several factors, considering the type of pollutant, the property to be measured and also the type of measuring location. Ideally, the technique must be sensitive enough to detect the species of interest, fast enough to track changes in concentration and composition, and free of interferences from other species present in the same air mass (Alfarra, 2004).

According to Directive 2015/1480/EC, PM_{10} and $PM_{2.5}$ should be measured by the standard gravimetric method (reference method) described in EN12341:2014, that provide information on mass concentration. The gravimetric method is based on the determination of the PM mass concentration by a low porosity filter, which is weighed under controlled conditions of temperature and relative humidity. Moreover, the AQ directive allows a European Member State to “use any other method which it can demonstrate gives results equivalent to the (reference) method”. The method that is widely used for PM measurements on automatic monitoring stations is beta attenuation. In this method, PM is continuously collected and the attenuation of beta radiation by the collected material is recorded (WHO, 2002).

As an alternative, optical methods are used to measure particle concentration that can be based on the principles of scattering, absorption and light extinction (Amaral et al., 2015). Optical particle counters (OPCs) are well-known instruments of this class allowing the measurement of particle size and number then converted into PM mass (Giechaskiel et al., 2014). Another important example of optical instruments are Aethalometers. An Aethalometer collects PM on a filter and measures the variation in light transmission as a function of time at multiple wavelengths. This approach allows the determination of BC concentrations since it is a component of PM that strongly absorbs light (Giechaskiel et al., 2014). These alternative methods have the advantage of providing instantaneous information on different PM metrics, however, they must be calibrated with the reference method data.

b) Air quality modelling

AQ models are powerful tools for the assessment and forecast of pollutants concentrations in the atmosphere (Thunis et al., 2012a). They are used to identify the causal relationship between the emissions of pollutants, meteorology, atmospheric concentrations, depositions and other factors (Daly and Zannetti, 2007). This models usually deals with (Denby, 2011):

- Assessment and reporting of the exceedance of limit values;
- Assessment of source contributions;
- Long term air quality planning and management;
- Short term mitigation and forecasting of air quality.

Historically, the air quality management has been based on monitoring data, because it is considered the method that gives data closer to reality (Denby et al., 2010). However, while the air quality models can give the spatial and time distribution of pollutants concentrations, air pollution measurements can only describe air quality at specific locations and times, without giving clear guidance on the identification of the causes of the air quality problem (Daly and Zannetti, 2007). Moreover, modelling can provide pollutants concentration data for places that monitoring cannot access.

Additionally to the advantages related to spatial coverage, models can be used to simulate future scenarios. For instance, they can be used to access a hypothetical situation before it occurs, such as the implementation of metro transport within a city or traffic restrictions. Furthermore, air quality models play a key role in identifying the most cost-effective strategies for reducing emissions and protecting human health and welfare, thus making them essential in regulatory and research applications.

Nevertheless, the air quality modelling comprises a number of limitations. These models are interesting to access past and future ambient pollutant concentrations and test scenarios. However, their performance depends of several factors including the quality of input data. AQ models require an amount of input data (e.g. emissions and meteorology), which are not always easy to obtain and very reliable. Although the advances in computational modelling, the models remain uncertain in their predictions and it is crucial to validate the results (Denby et al., 2010; Sokhi et al., 2022).

The air quality models (AQM) can be divided, broadly speaking, into two categories: physical and mathematical models. The physical models are used to simulate the atmospheric processes that affects the pollutants distribution in a smaller scale representation (e.g. wind tunnel). They can provide interesting results, however, they cannot serve the needs of ambient air quality capable of relating the emissions to air quality under a variety of conditions over an urban area (Seinfeld, 1986). In the case of mathematical models, a set of analytical/numerical algorithms is used to describe the physical and chemical processes that affect the pollutants dispersion in the atmosphere. The mathematical models can be divided in two classes, statistical and process-oriented models.

The first class is based on statistical analysis of air quality data and several correlated parameters. An example of this class is the Land Use Regression (LUR) models which spatially link ambient observed pollutant concentrations (treated as dependent variable) with other independent variables such as distance to pollutant source, topography, building types, land use and traffic volume within GIS (Cordioli et al., 2016; de Hoogh et al., 2016; Domhnaill et al. 2023; Liu et al., 2015). Applications have demonstrated a good agreement between measured and modelled pollutants (Cordioli et al., 2016; Crouse et al., 2009), though, they require a considerable amount of pollutant measurements to define the correlations, being usually made an exclusive measurement campaign with several measuring points distributed within the study area.

The second class is based on the fundamental description of atmospheric transport and chemical processes. They include the traditional air dispersion models, which can be categorized by their type (Gaussian, Lagrangian and Eulerian models) or by their application (e.g. street canyon, urban or regional models) (Denby, 2011). In general, the dispersion models treat the primary pollutants emitted from the emission sources. Nevertheless, they may include the chemical processes that are in the core of the secondary pollutants formation, thus, being referred as the chemical-transport models (CTMs).

Additionally, several studies have integrated process-oriented models with statistical models (Fang et al., 2023; Liu et al., 2018; Michanowicz et al., 2016; Murray et al., 2023; Konovalov et al., 2009) in a way of combining the strengths of both approaches and improve the outcomes.

Finally, an important concern must be the quality of the final results. Therefore, data quality objectives must be established for both monitoring and modelling approaches. Data quality objectives (DQOs) are qualitative and quantitative statements that clarify the purpose of the study, define the most appropriate type of information to collect, determine the most appropriate conditions from which to collect that information, and specify tolerable levels of potential decision errors (USEPA, 2004). The data quality objectives for both monitoring and modelling approaches are defined in the Annex of the Directive 2015/1480/EC. Additionally, to evaluate AQ modelling performance several quantitative and qualitative indicators are recommended by FAIRMODE (Denby et al., 2010; Janssen et al., 2022).

2.2.3. Air quality planning and forecast

Since the 1960s, with the development of air pollution management and research, it has become urgent for people to understand the current impacts air pollution and the future trends of pollution (Bai et al., 2018). As previously stated, AQ models can be used for different purposes, including long-term planning and short-term AQ forecast. For AQ planning, several policies can be tested before their implementation by simulating future scenarios. For example, the effect of technological advances (affect emission factor) or structural changes (affect activity data) can be assessed. As such, policy makers may come with a range of possible strategies that will need to be quantifiably assessed using air quality models, which will consist in multiple model runs to evaluate the response in the modelled pollution levels to a range of emission scenarios. How this is carried out depends on the requirements for any particular application.

AQ forecast may be used for short-term mitigation and prevention of severe air pollution episodes. Several techniques have been developed and used in air quality forecast, which varies from statistical (e.g. artificial neural networks, regression models, non-parametric regressions) to process-oriented approaches (e.g. chemical transport models – CTMs). These approaches have been widely applied for global and regional AQ forecast, however, their application to urban scale forecast is limited. Some examples of models applied at a global scale are MOCAGE (Rouïl et al., 2009) and GOCART (Chin et al., 2003). In research for regional AQ forecast, models like CHIMERE (Borrego et al., 2011; Menut, 2023; Menut et al., 2013; Rouïl et al., 2009), CAMx (Astitha et al., 2008; Dalla Fontana et al., 2021; Liu et al., 2018), CMAQ (Hogrefe et al., 2018; Liu et al., 2018; Mathur et al., 2017) and WRF-CHEM (Kalita et al., 2023; Silveira et al., 2021; Zhang et al., 2010) are frequently used. In Portugal, the forecast service is provided by QualAr (https://qualar.apambiente.pt/), using numerical forecast modelling (developed by GEMAC) for the mainland and using statistical modelling for Madeira Archipelago (developed by DCEA/FCT/UNL). The service may be complemented, whenever necessary, by forecast data from the European Union Copernicus Atmospheric Monitoring Service (CAMS, https://atmosphere.copernicus.eu/), one of the six COPERNICUS services which provides freely and continuous data and information on atmospheric composition, being used to assess/forecast the AQ at global/regional scale.

At urban scale, AQ forecast have been performed also based on statistical approaches (Donnelly et al., 2015; Barthwal and Acharya, 2021; Guo et al., 2023), and based on deterministic modelling, including Lagrangian and Gaussian models (Finardi et al., 2008; Righi et al., 2009; Gonzalez et al., 2022). Furthermore, Computational Fluid Dynamics (CFD) models have been coupled to CTMs models (or, regional models) creating multiscale-system that allow to forecast urban air quality (Heinold et al., 2019; Sokhi et al., 2022; Zhang et al., 2012). These systems typically use the regional model to solve the urban background concentrations, which is linked to a model with a higher resolution to obtain the AQ forecast at urban/local scale. Recently, a novel approach based on the CAMS service have been used to obtain background concentrations for urban AQ forecast (Johansson et al., 2023; Maiheu et al., 2018; Stidworthy et al., 2018).

Due to both AQ planning and forecasting typically relies on AQM, they involve some uncertainties. Actually, a well-known quote by George Box states that “all models are wrong but some are useful”. The performance of AQM is in many cases affected by the uncertainties related with model simplifications and model input data. Therefore, data assimilation techniques have been applied in conjunction with AQM, such as 4-dimensional variational method (4D-Var), Kalman-filtering, and ensemble methods (Copernicus, 2020). Thus, hybrid methods using both statistical and process-oriented models have been applied to improve the accuracy of forecasting results (Johansson et al., 2023; Bai et al., 2018; Zhang et al., 2012). According to Bai et al.(2018), the hybrid methods has good robusteness with low risk and strong adaptability, however, the process of building models is very complex. In conclusion, there is no best approach to produce accurate results. The methodology must instead be adaptate to the objectives and the input data available. Moreover, the use of different approaches in the same study can be useful.

The complexity of model implementation in urban areas with non-homogeneous distribution of pollution sources, dispersion conditions and obstacles, require complex and flexible modelling systems that should be able to provide quantitative information for decision making.

2.3. Modelling tools

2.3.1. Transportation models

Since the mid-19th century, the urban and transport planners recognized the importance of the travel demand models. Firstly, the transport planners relied on aggregated approaches such as the gravity and entropy models to assess the travel demand. In the mid-20th century, basing on the aggregated approaches, a sequential process to obtain the travel demand, named the four-step model, was developed (Mcnally, 2007). A relatively disaggregate version of this model is nowadays used by several urban and metropolitan planning organizations worldwide (Mcnally, 2007; Rich, 2015; Sivakumar, 2007).

Simultaneously to the development of the four-step model, urban planners also acknowledged the interconnectivity between the transport system and the rest of the urban system, which resulted in the development and application of the land use-transport (LUT) models (Sivakumar, 2007). In the 20th century, it was developed the first LUT model, named the Lowry's Model of Metropolis (Wegener, 2004). Lowry’s model was based on theories of spatial interaction, including the gravity model that was popular in quantitative geography at the time (Iacono et al., 2008). They have evolved from simple aggregate models to complex economic and econometric models of the market processes (Iacono et al., 2008).

Meanwhile, the implementation of disaggregate choice models within the four-step approach became possible due to the advances in econometric modelling. The disaggregate models consider the influence of individual socio-demographics on travel-related choices by apply modelling methodologies such as constrained optimization and random utility maximization. However, because of data limitations, they are usually implemented in clustered zones according to socio-demographic data (Sivakumar, 2007). Moreover, these trip-based models continued to exhibit several critical limitations such as the fact that they do not consider the linkages between trips. For instance, home-work commute trips were considered independently of the return from work to home travel, being both travels classified as home-based work trips. As a result, the tour based models were developed to overcome this limitation, which divides all individual travels into tours based at home and trips not based at home. A key principle in the model is that if a car is to be used on a tour, then it must be used for the entire trip, since the car must be returned home at the end of the tour (Miller et al., 2005). This implies that if someone travels from home to work and then stops at a store on the way back home, this would be viewed as one home-based work trip and one non-home based other trip. For that reason, the tour-based models continue to neglect linkages between trips.

Due to the limitations of conventional four-step modelling, a paradigm shift occurred with the development of the activity-based approach to travel demand modelling. Activity-based models recognize that the travel demand depends of the individual desire to participate in different activities among an area. Therefore, it forecast activities and related travel choices by taking into account time and space constraints as well as individual characteristics. Activity-based models thus shift the emphasis from a flow analysis to an intricate understanding of individual decision-making, being the bridge between the macro and the microscopic travel modelling. However, they are computationally difficult as they require solving many optimization problems simultaneously. Therefore, some models employ external aggregate methods such as User Equilibrium to consider the activity participation in time-dependent route choice behaviours of travellers (Lam and Yin, 2001; Wang et al., 2017).

Furthermore, an alternative to address the activity in traffic modelling is the agent-based travel demand modelling. Agents are the basic unit of activity in the model, which are like people, who have characteristics, goals, and rules of behaviour. This modelling approach is flexible and capable to model individual decision making process, by means of the interaction between agents and environment based in their behavioural rules. Microscopic traffic simulation can be viewed as an example of agent-based models, where vehicles are agents and the road network is the environment (Zhang and Levinson, 2004). The vehicle behaves and interacts with other vehicles and the environment are defined by rules, such as free-flow driving, car-following, and lane-changing. Moreover, some researchers are integrating this models in the LUT models (Ziemke et al., 2016).

Simulation modelling is an effective and increasingly popular tool to evaluate complex dynamical problems which may be difficult to describe in analytical terms. Traffic simulation models may be categorized according to various criteria (level of detail, scale of application, scale of the independent variables, and representation of the processes). In a broader sense, they are classified as microscopic, mesoscopic or macroscopic models according to the level of detail used to represent the traffic system (Hoogendoorn and Bovy, 2001). Whereas macroscopic models focus on a lower detail and deals with traffic flow in terms of aggregate variables (i.e. the behaviour of individual drivers is not considered); microscopic models focus on a higher detail and consider the time-space behaviour of individual drivers under the influence of vehicles in their proximity. In microscopic models the individual vehicles are traced, for instance, a lane-change of a vehicle is described as a detailed chain of drivers' decisions. Mesoscopic models fall in between the previous two models, with a medium level of detail. Thus, individual vehicles are not distinguished or traced, but the behaviour of individuals are considered, for instance in probabilistic terms.

Many research studies have been carried out on driver's behaviour in different situations like a case when he meets a static or a dynamic obstacle. The knowledge achieved by these researches have been implemented in the microscopic models, which have been applied in several traffic flow studies. These models require a higher amount of input data when compared with the models with a lower level of detail (Figure 2.8). Consequently, although the scientific and computational evolution, they are usually applied for smaller domains. For instance, in urban research they are frequently applied just for parts of the urban area, not being usually an option for citywide analyses. In general, macroscopic models are chosen for urban scale studies (Gulliver and Briggs, 2005; Righi et al., 2009). Macroscopic models have evolved considerably over time, however, in practice their application usually relies on the classical approach, the four-step model (Mcnally, 2007).

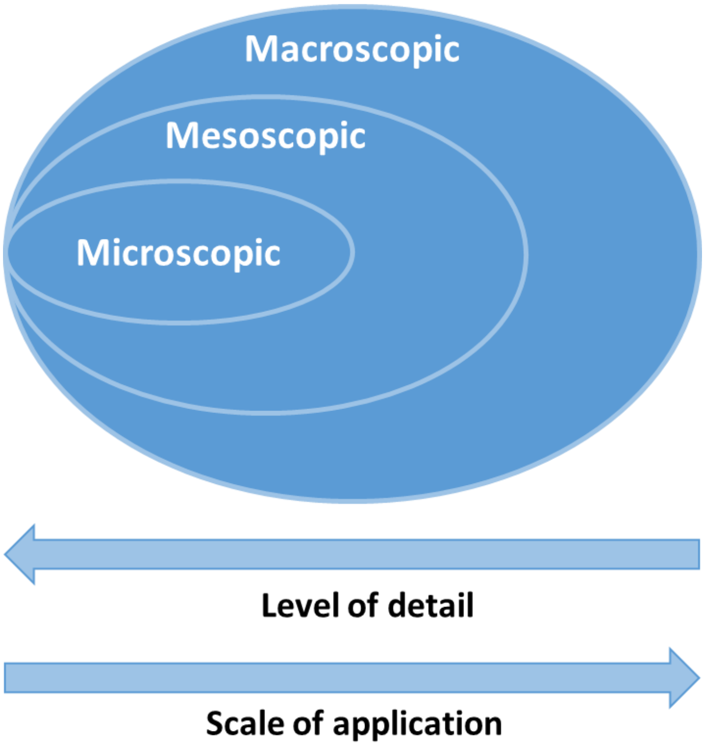


Figure 2.8. Different types of traffic simulation models.

Due to the growing environmental concern, several research studies established the integration of traffic simulation models with external emissions models. Both microscopic and macroscopic models that are linkable to emission models are presented in Table 2.3. While, the macroscopic models are typically used to calculate traffic emissions based on the average speed and kilometres travelled by each vehicle group; the individual vehicle movement simulations by microscopic models provide specific speeds and accelerations that are fed into subsequent emission calculation approach (Mądziel, 2023). Overall, PTV group have developed the most famous traffic simulation models used worldwide, VISUM and VISSIM. Whereas VISUM is widely used for macro and mesoscale simulation (Dias et al., 2019a; Hamad et al., 2022; Rodriguez-Rey et al., 2021), VISSIM was designed for microsimulation (Fernandes and Coelho, 2023, 2017; Gemma et al., 2023), being either integrated or coupled with different emission models.

Table 2.3. Application examples of traffic simulation software to vehicular emission studies.

Software	Type	Description	Reference
AIMSUN	Microscopic	An integrated traffic modelling software, which stands for focusing static and dynamic traffic assignment with mesoscopic, microscopic and hybrid simulation. It allows to include cars, trucks, trams and pedestrians.	Acuto et al. (2022); Rodriguez-Rey et al. (2021); Samaras et al., 2022
CORSIM	Microscopic	A microscopic traffic simulation model developed by the Federal Highway Administration (FHWA) in1996. It includes the analysis of freeways, urban streets, and corridors or networks.	Karekla et al. (2018); Kosman et al. (2003)
INTEGRATION	Microscopic	A trip-based microscopic simulation and traffic assignment model that was developed by the late Michel Van Aerde in 1983.	Ahn and Rakha (2013, 2008); Ahn et al. (2002)
METANET	Macroscopic	A macroscopic traffic model that simulates complex traffic flow phenomena over all traffic conditions (free flow, critical, congested). Modelling of the drivers' routing behaviour is enabled and several related options are offered to the user.	Csikós et al. (2018); Luspay et al. (2020, 2017); Zegeye et al. (2013)
PARAMICS	Microscopic	A microsimulation tool initially developed at the University of Edinburgh in the early 1990's. It allows simulations of individual vehicle movements to estimate future travel pattern behaviour due to a variation in traffic volume or geometric road layout.	Mascia et al. (2017); Misra et al. (2013); Song et al. (2020)
SUMO	Microscopic	An open-source, microscopic, inter- and multi-modal, space-continuous and time-discrete traffic flow simulation software that can manage large networks. It was developed and made available by Eclipse foundation since 2001.	Adamidis et al. (2020); Borrego et al. (2016); Gao et al. (2020); Aguiar et al. (2022)
TRANUS	Macroscopic	An aggregate LUT modelling system that has been applied to a large number of cities and regions throughout the world. Its development began in 1982, with the first practical applications since 1985.	Bandeira et al. (2011); Dias et al. (2021); Yuan et al. (2017)
VISSIM	Microscopic	A microscopic, behaviour-based multi-purpose traffic simulation software, which offers a wide diversity of urban and highway applications and integrates public and private transportation. It was developed at the University of Karlsruhe, Karlsruhe, Germany, in the early 1970s.	Abou-Senna et al. (2013); Dias et al. (2018); Fernandes and Coelho (2023, 2017); Gemma et al. (2023); Kosman et al. (2003); Liu et al. (2023)
VISUM	Macroscopic	A macroscopic model that has become a recognized standard in transport planning. It consistently models all road users and their interactions. It is used to model transport networks and travel demand, to analyse expected traffic flows, to plan public transport services and to develop advanced transport strategies and solutions.	Borrego et al. (2003); Dias et al. (2019a); Hamad et al. (2022); Rodriguez-Rey et al. (2021); Tang et al. (2017)

2.3.2. Emission models

Emission models of air pollutants provides emission data, an important input to dispersion and photochemical models, impact studies and AQ management. These models typically require detailed information on activity data (e.g. traffic volume, vehicle fleet composition, traffic situations) and emission factors (EF). The activity data may be obtained from statistical data, traffic counting or provided by the traffic simulation modelling. The EF are based on the emission measurements that traditionally were defined on the basis of chassis dynamometer tests under laboratory controlled environment (Smit et al., 2009). However, new approaches allowing the collection of emission data while driving in real traffic conditions become a valuable alternative (Franco et al., 2023).

There are different approaches for traffic related emission modelling, that generally are chosen according to the modelling scale, i.e. whether the objective is to predict emissions on a national level (i.e. macro-scale) or for a given road (micro-scale) (Mądziel, 2023; Faris et al., 2011; Smit el al., 2010), and depending on the details of available transport activity data. Smit et al. (2008) characterize the emission models by taking into account the way they incorporate the driving behaviour, which allowed to define different types as following:

a) <u>Average speed models:</u>
   These models are based upon the principle that the EF is a function of one (continuous) driving pattern variable (i.e. average speed), where the average speed is defined as the overall speed on a road section or for a whole trip. The information on average speed can be obtained from traffic models or travel time surveys. This is one of the common approaches used to model road traffic emissions (Smit et al., 2008). Over the years, the average speed models increased their complexity by including a higher number of pollutants, vehicle classes, new technologies, influencing factors and different types of emissions (Smit et al., 2009). Some models also incorporate the distinction between urban, rural and highway driving to account for variations in driving performance (Kousoulidou et al., 2013). The estimation of EF based on the average speed is relatively simple, thus, these models are generally less demanding computationally and less time consuming than other models. Therefore, they are a suitable choice for macro and mesoscale studies. However, for microscale studies, these models may not be representative for the estimation of emissions (Baškovič and Knez, 2013; Smit et al., 2009). For instance, a local traffic

intervention with large impacts on the stop-and-go pattern of vehicles but not affecting their mean travelling speed cannot be considered in this type of approach. Moreover, for microscale studies the single (mean) speed may reduce the accuracy of the results. Other parameters, such as the acceleration of road vehicles, must be addressed to understand the driving behaviour effects at local emissions.

b) <u>Traffic situation models:</u>

These type of models uses discrete emission factors for certain “traffic situations” by incorporating the speed and the driving cycle dynamic into the calculation. Different traffic situations may be related to different conditions that cause specific emission rates, which may not be justified with the average speed of the vehicles. Therefore, these models tend to be more suitable for local emission studies, although they can be also applied to national and regional studies (Boulter et al., 2007). The traffic situation models require the travel distance of each vehicle and information on what particular traffic situation can be applied to each road link (e.g. freeway, heavy traffic, stop-and-go, etc.). The concept of service level of a road introduced by this models is established by means of emission factors derived from on-board measurements during real driving cycles (Borge et al., 2012). These emission factors when compared with those obtained directly from laboratory testing, tend to be more realistic. Therefore, the traffic situation models would present more accurate values for some studies then the average speed models. However, according to Smit et al. (2010), both models tend to overestimate some pollutant emissions.

c) <u>Modal models:</u>

In these models the emission factors are a function of specific engine or vehicle operating modes. Different types of these models are referred. In the ‘simple modal models’, the vehicle operation is defined with a few number of parameters such as idle, acceleration, deceleration and cruise (Boulter et al., 2007). A number of more detailed modal models with the purpose of providing a more precise characterization of the vehicle emission behaviour correlate the emissions rate to vehicle operation at a high resolution (usually one second). These models are referred as ‘instantaneous models’ which are usually applied for microscale studies (Acuto et al., 2022; Boulter et al., 2007; Fernandes et al., 2016; Fernandes and Coelho, 2023, 2017; Gastaldi et al., 2014; Smit et al., 2009). They typically require very detailed information on vehicle flow (e.g. instantaneous speed, acceleration and road gradient) that can only be obtained by microscopic traffic models or by Global Positioning System (GPS) equipment placed on the vehicles (Fernandes and Coelho, 2023, 2017; Kousoulidou et al., 2013). In theory, these models allow to calculate the emissions for any vehicle operation profile and they take into account the dynamics of driving cycles (Boulter et al., 2007). Therefore, they allow to explain the fluctuations in the vehicle emissions that were difficult to be justified by the average speed parameter. Moreover, they enable to understand the spatial variation of emissions, thus, potentially improving the prediction of air pollution. However, they require a substantial amount of input data which collection is expensive and resource-intensive (Fernandes and Coelho, 2023, 2017; Gastaldi et al., 2014; Smit et al., 2009).

Overall, highly detailed vehicle data serve as input of instantaneous emission models which allow quantification of emissions induced by an individual vehicle under a specific set of conditions (e.g. during acceleration or idling, Boulter et al., 2007). However, conceptually simpler methodologies are frequently used to simulate emissions induced by national fleets and during longer time periods by means of drive-cycle or average-speed emission factors and traffic data. These simpler approaches are often used to calculate local-scale emissions, though not designed for this purpose (AQEG-Defra, 2021). Such result of the inherent complexity in calculating road traffic emissions with a high level of accuracy, since it is a function of vehicle operating conditions (e.g. temperature in the engine, after treatment systems, driving style), vehicle technologies, age and driving distance. Thus, considering all these complexities may not be appropriate given the aim of the emission model or inventory, since uncertainties will always remain (AQEG-Defra, 2021).

The EMEP/EEA Emissions Inventory Guidebook (EMEP/EEA, 2019a, 2019b) provides a methodology to quantify traffic emissions for national inventory reporting. The Guidebook provides three alternative methodologies (Tier 1, 2 and 3), which increase the level of detail and input data required from Tier 1 to 3. Tier 3 approach uses EF (in g/km) as a function of average speed, varying over vehicle classes, technology (i.e. Euro standards) and fuel types. Moreover, EMEP/EEA Emissions Inventory Guidebook includes EF from different processes (i.e., exhaust and non-exhaust processes). These approaches are implemented in the EEA's software "Computer Program to Calculate Emissions from Road Transport" (COPERT) which allows European countries to develop their own national inventories. Although most European countries use the Guidebook and COPERT model (AQEG-Defra, 2021), there are other models used at a national level, as described in Table 2.4. In Portugal, besides the widely use of COPERT model for national inventories, also TREM (from the University of Aveiro - Tchepel, 2003) and later QTraffic (from the University of Coimbra, as an update of TREM model for open-source QGIS platform - Dias et al., 2019a) average speed models are used. QTraffic model is specifically designed for emissions estimation at road segment level being roads considered as line emission sources. This model also considers the EF defined in the EMEP/EEA Guidebook.

Table 2.4. Description of emission models used from a national level (macroscale) to an urban (in this case defined as mesoscale) and microscale (defined as local-scale).

Model name	**Country***	**Type of model**	**Scale**	**Description**
COmputer Programme to calculate Emissions from Road Transport (COPERT)	Thought developed in Europe is used worldwide	Average speed model	Macro to Mesoscale	It is coordinated by the EEA and gives both exhaust and non-exhaust emissions, serving as a European emission model reference.
Handbook of Emission Factors for Road Transport (HBEFA)	Worldwide, especially in Europe	Traffic situation model	Macro to Mesoscale	The emissions estimation and EF are based on the simulation of traffic situations, road type and vehicle categories. This model have already included non-exhaust emissions like COPERT, which are in the same order of magnitude, yet higher than COPERT emissions due to inclusion of resuspension emission factors as well.
Transport Emission Model (TREMOD)	Germany	Traffic situation model	Macro to Mesoscale	A model that predicts all transport modes and is closely linked to HBEFA, since it uses the same methodology and EF.
Network Emission Model (NEMO)	Belgium	Traffic situation model	Macro to Mesoscale	NEMO combines detailed simulation of vehicle fleet composition and EF, by considering data from European in-use measurements and full set of vehicle specifications available on PHEM and HBEFA.
Defra's Emissions Factors Toolkit (EFT)	UK	Average speed model	Macro to Mesoscale	It employs the assumptions from the UK National Atmospheric Emissions Inventory (NAEI) to provide average-speed-specific emissions from different vehicle types and is recommended for local emission modelling.
Motor Vehicle Emission Simulator (MOVES)	Worldwide, especially in USA	Instantaneous emission model	Macro to Microscale	The United States Environmental Protection Agency (EPA) emission model combines traffic activity data with emission measurements from various driving modes to calculate emissions rates as a direct function of vehicle specific power (VSP) and speed.
*The utilization in different countries was accessed by searching studies in the WEB of Science (on 29 of August, 2023) were these models are used, combining with the place of development and AQEG-Defra (2021) literature review.				

Table 2.4. (Continued).

Model name	**Country***	**Type of model**	**Scale**	**Description**
VERSIT+	Worldwide, especially in Europe	Traffic situation model	Macro to Microscale	An emission model developed by TNO that relates emissions to average speed, acceleration and/or VSP. While the heavy-duty module is based on PHEM, the light-duty module uses statistical relationships between situation type and emission measurement data.
Versit+^{micro}	Worldwide, especially in Europe	Instantaneous emission model	Microscale	A VERSIT+ simplified version that uses average vehicle fleets to link to microsimulation traffic models.
Enviver	Worldwide	Instantaneous emission model	Microscale	A PTV add-on to the VISSIM traffic model to allow emission quantification by integrating VERSIT+^{micro} EF.
Analysis of Instantaneous Road Emissions (AIRE)	Scotland	Instantaneous emission model	Microscale	An emission model developed by Transport Scotland that comprises PHEM look-up tables to predict emissions at a micro-scale. Although, AIRE can use traffic activity data from different sources, it was developed to be integrate with S-Paramics micro-simulation traffic model.
*The utilization in different countries was accessed by searching studies in the WEB of Science (on 29 of August, 2023) were these models are used, combining with the place of development and AQEG-Defra (2021) literature review.				

At a finer scale, typically microscale emission models are used to calculate emissions at an individual road link. These models require detailed traffic movement data so are often linked to traffic simulation models. The additional demands placed on traffic data, when compared with macro to mesoscale models, may add further sources of uncertainty.

In sum, the national scale models (which are usually based on average speed or traffic situation approaches) are relatively simpler, less demanding computationally and less time consuming than microscale models. However, for individual roads microscale emission models are more suitable since they consider complex driving behaviour effects and local traffic conditions.

Given the specific objectives of this thesis, it is important to emphasize that PM traffic emissions are classified as exhaust and non-exhaust emissions for modelling purpose. While the exhaust emissions result from fuels combustion process, the non-exhaust emissions result from the abrasion of vehicles components and road surface and the resuspension of road dust. Moreover, the exhaust emissions are a sum of the emissions that occur during the ‘hot’ stabilized phase (i.e. hot emissions) and the transient ‘warming-up’ phase (i.e. cold start emissions). Hot emissions are influenced by several factors such as the vehicle travel distance, speed (or road type), acceleration, age, engine size and vehicle load. However, in many situations robust data for each factor are not available, therefore simplistic methodologies are frequently used. All the models previously indicated (Table 2.4) incorporate the calculation of hot exhaust emissions.

Cold start emissions are calculated as an extra emission over the hot emissions that would be expected if all vehicles were only operated with hot engines and warmed-up catalysts. They are obtained by means of a ratio of cold over hot emissions which is applied to the fraction of kilometres driven with a cold engine (i.e. cold start distance). According to Reiter and Kockelman (2016), the cold start distance is a function of the pollutant studied, varying considerably from study to study. André and Joumard (2005) estimated a broad distribution (by pollutant) of cold start distance with an average of 5.2 km at 20ºC. Several studies do not take into account such emissions, or quantify them with a macroscale approaches. Given that such emissions may be the double or even the triple of hot exhaust emissions combined with the average cold distance value, it is undoubtable that the determination of such emissions is very important in urban areas, since many automobile trips are shorter than the cold distance and a significant share of urban driving occurs while the engine is cold. Thus, methodologies that may be adapted to urban scale studies with a higher level of detail are desired.

Several studies have assessed non-exhaust emissions using methods such as on-road measurements, road simulators and near-road research (Amato et al., 2009; Etyemezian et al., 2003; Gustafsson et al., 2008; Kuhns et al., 2003; Pirjola et al., 2009; Querol et al., 2004), revealing that resuspension of surface-accumulated PM is the primary contributor to non-exhaust emissions. An exhaustive analysis of road dust resuspension was recently carried out, concluding that the comparability of emission results is jeopardized by mismatch methodologies and the lack of a common framework for establishing a stable and reliable sampling protocol (Rienda and Alves, 2021). The EMEP/EEA Emissions Inventory Guidebook (EMEP/EEA, 2019b) addresses tire, brake, and road wear emissions, however neglects the resuspension parcel of PM concentrations.

Table 2.5 presents examples of emission models and their application in PM traffic-related emission modelling.

Table 2.5 Application examples of emission models used to estimate traffic-related PM emission.

Reference	Scale	Case study	Emission Model	PM emissions processes
Biswal et al. (2023)	Macroscale	Delhi, India	COPERT	Non-exhaust processes not included
Sarica et al. (2023)	Macroscale	Paris, France	COPERT	Resuspension not included
Andre et al. (2020)	Macroscale	Ile-de-France region	COPERT	Resuspension not included
Baptista et al. (2010)	Macroscale	Lisbon, Portugal	COPERT	Non-exhaust processes not included
Dias et al. (2018)	Mesoscale	Aveiro, Portugal	TREM	Only exhaust emissions
Dias et al. (2019a)	Mesoscale	Coimbra, Portugal	QTRAFFIC	Only hot exhaust emission
Borrego et al. (2016)	Microscale	Aveiro, Portugal	VSP/EMEP	Only exhaust emissions
Ehrnsperger and Klemm (2022)	Microscale	Münster, Germany	HBEFA	Includes both exhaust and non-exhaust emissions
Posada-Henao et al. (2022)	Mesoscale	Medellín, Colombia	HBEFA	Only exhaust emissions
Mascia et al. (2017)	Microscale	Glasgow, Scotland	AIRE	Only exhaust emissions
Gastaldi et al. (2014)	Microscale	Mirano, Italy	AIRE	Only exhaust emissions
Abou-Senna et al. (2013)	Macro to microscale	Orlando, US	MOVES	Resuspension not included

Typically, the quantification of PM by means of emission models is frequently underestimated because non-exhaust processes are often not considered, which poses an increasing concern with the introduction of Euro standards that continuously allow the reduction of exhaust emissions combined with the emergent electric vehicles with zero exhaust emissions. Additionally, when cold start emission modelling is used, it is often done in an aggregated manner, which does not allow to access local contribution of such emissions. Particulate matter is usually quantified in terms of mass, however, recently its quantification in terms of particle number (i.e. PN) have risen interest due to health relevance of the number of finer particles that may be inhaled, being already integrated in models like HBEFA (Boveroux et al., 2021; Ehrnsperger and Klemm, 2022). Furthermore, PM components like BC is not yet regulated, which is in the source of BC lack of data and high uncertainties on its emission quantification. However, BC is related to negative impacts on health and environment, culminating in the recommendation of further research on its risks and mitigation approaches by WHO (WHO, 2021). Consequently, quantifying aerosol particle emissions through models requires further developments to reduce associated uncertainties, which may reduce the credibility of pollutant estimation by the air quality dispersion models.

2.3.3. Urban air quality dispersion models

Ambient air pollutant concentrations are distributed non–uniformly in urban areas, with hotspots usually located in central business district, traffic intersections and signalized roadways (Johansson et al., 2007). The complex spatial and temporal variations of pollutant concentrations is a result of several factors, such as city morphology, topography and meteorological conditions. For instance, near busy traffic areas the urban canopy and microclimate may contribute to the degradation of the air quality given the poor air dispersion conditions in the area. High pollution levels have been observed in street canyons, which is defined as the urban streets flanked by buildings on both sides. Within these streets, people are likely to be exposed to pollutant concentrations that currently exceed the AQ standards and WHO guidelines.

The spatial and temporal variation of pollutants within urban areas can be simulated by air dispersion models, which is a hard task. Besides the influence of micro-scale parameters, as we can see in Figure 2.9, mesoscale phenomena (e.g. orography effects, sea breeze circulations) play also an important role in the dispersion of pollutants in urban areas. On the other hand, these mesoscale phenomena are influenced by macroscale processes (e.g. global air circulation). Therefore, the urban scale modelling should consider variations of local scale effects together with the processes that may occur at a wider scale, including the transport of pollutants from long distances which may also affect the urban air quality. Given the complexity of the phenomenon of pollution in cities, urban scale modelling is a challenge for atmospheric scientists (Srivastava and Rao, 2011; Vardoulakis et al., 2003).

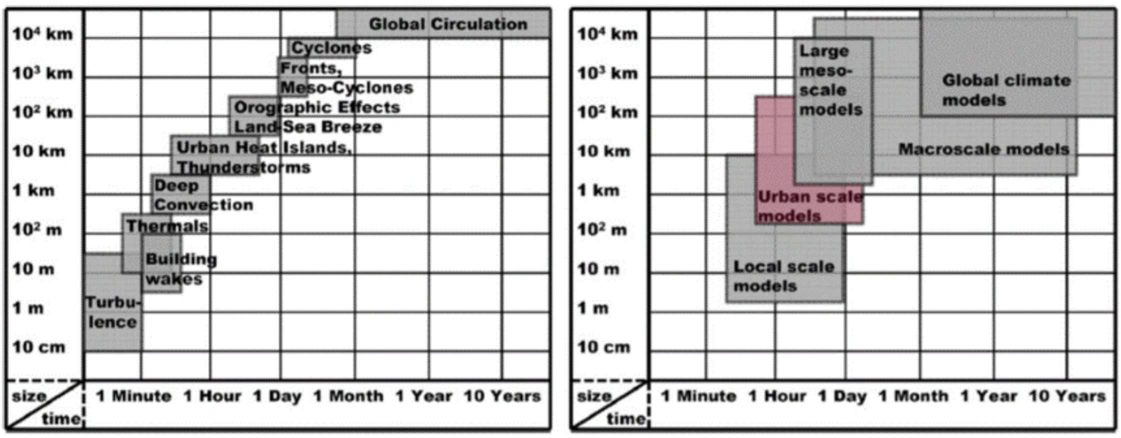


Figure 2.9. AQM spatial scales versus timescales. Left panel: common atmospheric phenomena that affect AQM. Right panel: atmospheric models covering different spatial and temporal scales (Isaksen et al., 2009).

The urban scale dispersion models may be used for assess or forecast the distribution of pollutants among a city. Application examples of these models used to simulate urban air quality are presented in Table 2.6. In general, they have a time resolution of 1 hour and the spatial resolution depends on the type of the model applied. The models applied with the resolution about 1-3 km (i.e. regional/urban scale) are able to resolve the variability in the urban background concentrations (Denby, 2011). However, at this resolution nuances that occur at a street level within a city could not be addressed. For this purpose, models with a higher spatial resolution are required, which are usually a function of street geometry, meteorology, local emissions and background concentrations. Examples of models with higher resolution (i.e. local/urban scale) are Gaussian and Lagrangian models, which allows to describe the impact of multiple emission sources present in an urban area, at a lower computational cost than the Eulerian models.

Table 2.6. Application examples of AQM used at urban scale in Europe.

Models	Description	Reference
ADMS-Urban	Advanced three dimensional quasi-Gaussian model nested within a trajectory model.	Hood et al. (2018); Barnes et al. (2014); Dėdelė and Miškinytė (2013); Duong and Murana (2023); Di Nicola et al. (2023); Righi et al. (2009)
AURORA	Eulerian chemical transport model	Beckx et al. (2009); Lefebvre et al. (2011); Mensink et al. (2003)
AUSTAL2000	Lagrangian model designed for long-term sources and continuous buoyant plumes	Dias et al. (2016); Paas and Schneider (2016); Pepe et al. (2016)
CAMX	Eulerian chemical transport model	Meroni et al. (2017); Pepe et al. (2016)
CHIMERE	Eulerian chemical transport model	Jiang et al. (2022)
CMAQ	Eulerian chemical transport model	Martins (2012); Benavides et al. (2019)
IFDM	Gaussian model designed to simulate non-reactive pollutant dispersion at a local scale.	Lefebvre et al. (2013, 2011)
UDM-FMI	Multiple source Gaussian model for hourly means developed by the Finnish Meteorological Institute	Karppinen et al. (2000)
URBIS	Multiple source Gaussian model for long term means developed by TNO, The Netherlands	Amato et al. (2016); Beelen et al. (2010)
URBAIR	A second-generation Gaussian modelling tool developed by the University of Aveiro to allow air quality management in urban areas.	Borrego et al. (2016); Fernandes et al. (2021)
R-LINE	A near-road Gaussian modelling tool that incorporates Gaussian dispersion curves to simulate dispersion of road source emissions.	Benavides et al. (2019)
WRF-CHEM	Nested and coupled meteorological and Eulerian chemical transport	Fallmann et al. (2016); Spiridonov et al., 2019

The Gaussian approach is based on a set of equations that describe the three-dimensional concentration field assuming that concentrations are proportional to the emission rate, inversely proportional to the wind speed, and that the dispersion of a plume can be described with a normal distribution ("bell-shaped curve") characterized by dispersion coefficients in horizontal (σ_y) and vertical directions (σ_z) (Zannetti, 1990). Given their relative simplicity, these models are very popular being widely used in engineering and scientific applications. They can consider a large ensemble of emission sources, dispersion situations and a receptor grid network, which is sufficiently dense spatially (of the order of tens of meters) (Karppinen et al., 2000; Srivastava and Rao, 2011). Furthermore, they may also include complex terrain, street canyon and intersection modules (Vardoulakis et al., 2003). The Gaussian models are not recommended for large scale modelling (e.g. regional scale modelling) due to the assumption of a homogeneous wind field (Holmes and Morawska, 2006).

The Lagrangian models can be applied for large scale modelling, however, they are also widely used in urban scale studies. These models were developed after 1970, when scientists began to realize that air pollution was not only a local phenomenon (Daly and Zannetti, 2007). The Lagrangian approach is based on the concept that the fluid elements follow an instantaneous flow (Zanetti, 1990), thus, it adopts a particle-based approach wherein individual particles or "parcels" are traced as they follow the wind trajectory. They are more physically realistic than the Gaussian models (Karppinen et al., 2000). However, they are numerically more complicated and computationally expensive than Gaussian models (Seinfeld and Pandis, 2006).

Since 2010s, the use of local/urban scale models to characterise concentration gradients of primary pollutants in city context has increased significantly (Singh et al., 2014; Soulhac et al., 2012). Nevertheless, both the absence of a link between the influence of local emissions and the regional transport of pollutants and the lack of a relevant representation of atmospheric reactivity, limit the scope of this type of model (Sokhi et al., 2022). Thus, over the last decade innovative approaches either coupling or nesting regional scale to urban scale models have emerged in the literature (Hamer et al., 2020; Hood et al., 2018; Khan et al., 2021; Veratti et al., 2020; Kim et al., 2018; Pepe et al. 2016), allowing to surpass these limitations and explore the advantages of both methodologies. The main focus of these studies has been the representation of road traffic and its contribution to urban AQ, showing greater comparability with *in-situ* observations than regional scale models alone. According to Sokhi et al. (2022), after statistical evaluations of these new approaches, most of the residual biases obtained could be attributed to a lack of realism in the emissions which includes the presence of poorly characterized local sources and insufficient temporal refinement of road traffic profiles. Thus, further improvements are expected to this multi-scale models. A more comprehensive literature review on current challenges of AQ models may be accessed in Sokhi et al. (2022).

Overall, this chapter highlights some research gaps in the literature regarding the quantification and forecast of aerosol particles from road traffic sources in an urban context, for which air quality modelling systems including AQ dispersion models, emission models and transportation models are crucial. Thus, the following chapters proceeds to specific developments in a way of improving such system.

Chapter 3

3. A bottom-up modelling approach to quantify cold start emissions from urban road traffic

This chapter is based on the publishing paper: "Pina, N., Tchepel, O. (2023). A bottom-up modeling approach to quantify cold start emissions from urban road traffic. International Journal of Sustainable Transportation, 17(8), 942-55".

3.1. Introduction

Road transport remains one of the most important sources of air pollution despite the effort in emissions reductions over the last decades (EEA, 2019a, 2018a). In urban areas, where population is directly exposed to traffic related pollution, emissions released during the first minutes of driving after the vehicle engine is started (i.e., cold start period) may contribute significantly to the total emissions from road traffic. Therefore, cold start emissions in urban areas should be quantified with fine spatial and temporal resolution to identify hotspots and thus establish mitigation measures and plans. This goal cannot be achieved by typical top-down downscaling of highly aggregated inventories (usually developed at national or regional scale) using spatial proxies and implementation of bottom-up emission quantification approaches starting from data collected and processed at the local level is required. The results of bottom-up approach application are highly affected by the amount and quality of data available (Cifuentes et al., 2021). However, it provides emissions with a greater spatial and temporal variations and relate emission levels directly with road transport activity (FAIRMODE, 2010), including the ones induced during the cold period. Furthermore, bottom-up approaches enable the characterization of emissions for line sources (i.e. road traffic sources) which is a crucial input for air quality modelling at a fine spatial resolution (i.e. local/urban scale).

The cold start emissions, addressed in this study, are defined as a difference between the total amount of vehicle emissions during the warm-up period and the amount of pollutant which would be emitted by the vehicle at its normal operational temperature (water temperature over 70 ºC) during the same time period (EMEP/EEA, 2019a). Experimental data revealed that during the cold start period a significant fraction of vehicle emissions occur, achieving almost 80% of absolute emissions for some pollutant species (Reiter and Kockelman, 2016). During this period, the emissions of carbon monoxide (CO) and hydrocarbons (HC) caused by petrol vehicles can increase by a factor of 11 to 15 (Clairotte et al., 2013; Dardiotis et al., 2013; Weilenmann et al., 2009).

Data from on-road emission tests of vehicles have shown that energy consumption and emission are significantly higher during the warming-up phase than under hot conditions (Du et al., 2020; Faria et al., 2018; Varella et al., 2017). In the laboratory measurements (i.e. chassis dynamometer and bench tests), road vehicle emissions are measured under controlled conditions while a driver operates a vehicle according to a predetermined test cycle that represents different real world driving conditions (e.g. urban driving). There are several studies that deals with the cold start emissions by means of laboratory measurements. Usually, the aim of these studies is to analyse the influence of different parameters on cold start emissions magnitude, such as: (i) ambient temperature (Dardiotis et al., 2013; Laurikko, 1995, 2008; Ludykar et al., 1999; Weilenmann et al., 2005, 2009); (ii) technology taking into account emission standards (Dardiotis et al., 2013; Weilenmann et al., 2009); (iii) the drive cycle (Jaworski et al., 2018); (iv) engine stop time (or parking time) (Favez et al., 2009).

Currently, chassis and engine dynamometer are the most established technologies to determine emission factors (i.e., EF - the average emission rate of a specific pollutant for a given source, per unit of activity). However, the emissions obtained by this technique may be not representative for real-world road transport emissions, due to the test cycles configuration and the limited number of vehicles of each technology tested. Also, it has been demonstrated that emission factors derived from laboratory tests underestimate real-world fuel consumption by up to 40% (Mock et al., 2014) and oxides of nitrogen (NO_X) emissions by seven times in Euro 6 vehicles (Franco et al., 2014). Therefore, several recent studies have been devoted to quantifying vehicle emissions by measuring under real-world conditions (Bishop et al., 2016; Faria et al., 2018; He et al., 2019; Outapa et al., 2018; Zheng et al., 2020). The data obtained from these measurements can play an important role in validating the EF gained from laboratory tests (Franco el al., 2013).

Both laboratory and real-world measurements are very important approaches that allow to define EF. However, they use technologies that are time consuming, expensive and usually are focused on a single pollutant species or a small sample of vehicles. Consequently, to study a variety of pollutants (Cifuentes et al., 2021; Zhang et al., 2013) or to understand the distribution of the emissions within a specific area (Borge et al., 2012; Faria et al., 2018; Tchepel et al., 2012), emission modelling is frequently used. Nowadays, several researches apply microscopic emission modelling by using Vehicle Specific Power (VSP) (Faria et al., 2018; Fernandes et al., 2016; Ferreira et al., 2022; Outapa et al., 2018). VSP models are able to provide instantaneous emissions, but require detailed inputs that are not always available, such as instantaneous data on vehicle speed, acceleration/deceleration and road gradient.

The requirements for spatial and temporal resolution should be seen in terms of the “fitness for purpose” to guarantee that the quality of modelling results is compatible with the quality objectives defined for a particular model application (EEA, 2011). Thus, in the case of traffic related air pollution assessment the data should be representative for a 100-m street segment (Annex III.B.1.b of the Air Quality Directive 2008/50/EC). In this context, emission modelling following an average speed approach (e.g. COPERT model) are commonly applied (Borge et al., 2012; Hood et al., 2018; Tchepel et al., 2020). The temporal resolution of the emission data should be also compatible with their final use. In the case of air quality assessment, the temporal resolution of emission inputs is usually one hour to guarantee that final results are comparable with the limit values defined by air quality legislation.

Additionally to hot fraction, emission modelling based on average speed approach allows quantification of cold start emissions. The ratio between cold/hot emissions in combination with the average trip length are usually considered for the quantification of cold start emissions at national scale (EMEP/EEA, 2019a). However, urban areas are characterised by short trips that might happen entirely with a cold engine. Also, parking time is very important factors that will influence cold start emissions. Therefore, a detailed methodology should be applied in urban areas to reduce uncertainties of cold emissions estimate.

In this paper, we propose a novel approach for bottom-up modelling of cold start emissions at urban scale with fine spatial resolution. A new algorithm is developed and linked to a transportation model that enables quantification of cold start excess emissions for each road segment while preserving information on Origin-Destination (O-D) of the trips contributing to those emissions. This modelling is based on widely used emission factors recommended for European fleet in combination with a novel approach developed to process the traffic activity data allowing calculation of cold start emissions with fine spatial resolution and preserving the O-D information. An application example of the developed approach to the Coimbra urban area (Portugal) is presented and discussed.

3.2. Methodology

The methodology developed for quantification of cold start emissions is presented in this topic. Additionally, a brief description of Traffic Emission and Energy Consumption Model (QTraffic) model used for hot emissions is also included.

3.2.1. QTraffic: Traffic Emission and Energy Consumption Model

The Traffic Emission and Energy Consumption Model (QTraffic) is a mesoscopic model that estimates traffic-related emissions and road vehicle fuel/energy consumption presented in previous studies (Dias et al., 2019a, 2019b). This model was developed by University of Coimbra and the current version is based on the updated European guidelines for emission factors (EMEP/EEA, 2019a). The hot emissions ($E_{hot\ seg,i}$ [g]) for each pollutant are quantified following an average speed approach (see Equation 3.1).

$$E_{hot\ seg,i} = \sum_{k} \left(EF_{hot,i,k}(v) \cdot N_k \cdot L \right) \tag{3.1}$$

where: $EF_{hot,i,k}$ is the emission factor [g·km^{-1}·vehicle^{-1}] for pollutant i and vehicle technology k defined as a function of average speed v [km·h^{-1}] for each road segment; N_k is the number of vehicles of technology k (classification based on European emission standards – Pre-euro to Euro 6); L is the road segment length.

For the model application detailed traffic data, such as number of vehicles and average speed for a line source and fleet composition (fuel type; vehicle technology and class), road parameters (e.g., slope and length) are required. Several examples of the model application could be found in previous publications (Dias et al., 2019a, 2019b; Tchepel et al., 2020).

Nevertheless, the cold start emissions were not included in the previous version of QTraffic, being this the focus of the current work.

3.2.2. Cold start excess emissions (CSEE)

The new module implemented in QTraffic considers, as the first premise, that cold start emissions are an excess to hot running emissions and released during the cold distance, see Figure 3.1. The excess amount of emissions released under cold engines operation is distributed over the cold distance until the engine achieves normal operation temperature. Any trips shorter than the cold distance are expressed as a fraction of cold distance length.

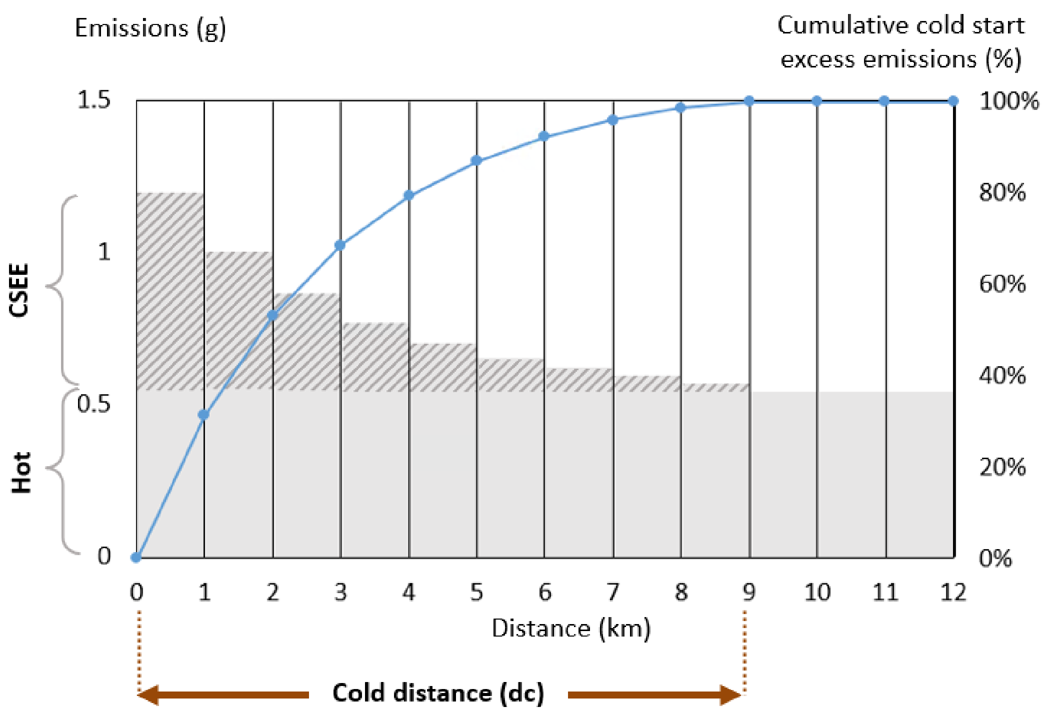


Figure 3.1. A schematic representation of cold distance and cold start excess emissions - for a selected type of passenger cars at reference conditions (ambient temperature = 20ºC; average speed = 20 km/h) for CO. Cumulative CSEE during the trip are presented by blue line.

The methodology defined in André and Journard (2005) was used to calculate cold distance (Equation 3.2 and Table 3.1). Thus, cold distance (dc) varies for each pollutant i and technology k according to: presence of catalytic converter (CC); fuel type (gasoline or diesel); ambient temperature (T) and vehicle speed (v).

$$dc_{ik} = f(T, v, CC, Fuel) \tag{3.2}$$

Table 3.1 Parameters considered for cold distance (dc) estimation.

Euro standard technologies	Diesel				Gasoline			
	CO	CO_2	HC	NOx	CO	CO_2	HC	NOx
PRE-EURO	T, v	v	v	v	T, v	v	T, v	T, v
EURO 1	v	v	v	v	v	v	v	v
EURO 2	T, v	T, v	T, v	T, v	T, v	v	T, v	v
EURO 3	v	v	v	v	T, v	T, v	T, v	T, v
EURO 4 -EURO 6	v	v	v	v	T	T	T	constant

T - ambient temperature; v - vehicle speed

A two-step approach is implemented in CSEE module (Figure 3.3) starting from the quantification of emissions per trip for each Origin-Destination (O-D) pair (step 1), and then attribution of the emissions to the road segments (step 2), as described below.

Step 1 – Trip-based cold emissions:

Trip-based emissions are calculated considering O-D of trips and average speed for each road segment, provided by transportation model. For this purpose, VISUM traffic planning software (PTV, 2013) was used considering: trip generation, trip distribution, modal split and traffic assignment. The traffic data obtained by modelling provides several advantages for the emissions quantification purpose in the comparison with real traffic point measurements due to their spatial coverage and possibility to preserve O-D information.

Assuming a possibility of different paths resulting from the traffic assignment, the trip-based CSEE for each O-D pair and pollutant i ($CSEE_{OD,i}$ [g]) are calculated as following (Equation 3.3):

$$CSEE_{OD,i} = \sum_{k} \sum_{path} w_{i,k}(T,v) \cdot h(\delta_{path,i,k}) \cdot g_{i,k}(t) \cdot N_{path,k} \tag{3.3}$$

where: w – excess emissions [g.vehicle^{-1}] corrected for ambient temperature T [ºC] and average vehicle speed v [km.h^{-1}] ; h($\delta_{path,i,k}=d_{path}/dc_{i,k}$) is the dimensionless distance correction factor (defined in Equation 3.4); $g(t)$ is the dimensionless correction factor for the parking time (t); and N is number of vehicles taking the same path.

For a given travelled distance d, a dimensionless correction factor h(δ) is defined with respect to the cold distance:

$$h(\delta_{i,k}) = \begin{cases} \dfrac{1 - e^{a_{i,k} \cdot \delta_{i,k}}}{1 - e^{a_{ik}}}, & \delta \leq 1 \\ 1, & \delta > 1 \end{cases} \tag{3.4}$$

where $\delta=d/dc$ and a < 0 is a coefficient (tabulated values can be found in André and Joumard (2005)).

It is important to highlight that the emission factors from André and Joumard (2005) considered in this work are based on measurements only for passenger cars covering average speeds between 18.7 km/h to 41.5 km/h and ambient temperatures between -20ºC to 28ºC (André and Joumard, 2005). Therefore, the other types of vehicles, such as heavy-duty vehicles, are not considered in our study but it is expected that their contribution to cold start emissions will be limited in the urban context.

Step 2 - Cold emissions per road segment:

The CSEE calculated in the previous step are attributed to the road segments considering the path between an O-D pair, the road segments sequence and length. For this purpose, the road segments are defined regarding the intersections in the road network. For each segment n of the path ($d_{path} = \sum d_n$) the emissions are attributed in accordance with the Equation 3.5 and the distance correction factor h(δ) is presented in Figure 3.2.

$$CSEE_{seg(n),i} = \sum_k \left(CSEE_{path,i,k} \cdot \frac{h(\delta_{n,i,k})}{h(\delta_{path,i,k})} - CSEE_{path,i,k} \cdot \frac{h(\delta_{n-1,i,k})}{h(\delta_{path,i,k})} \right) \quad (3.5)$$

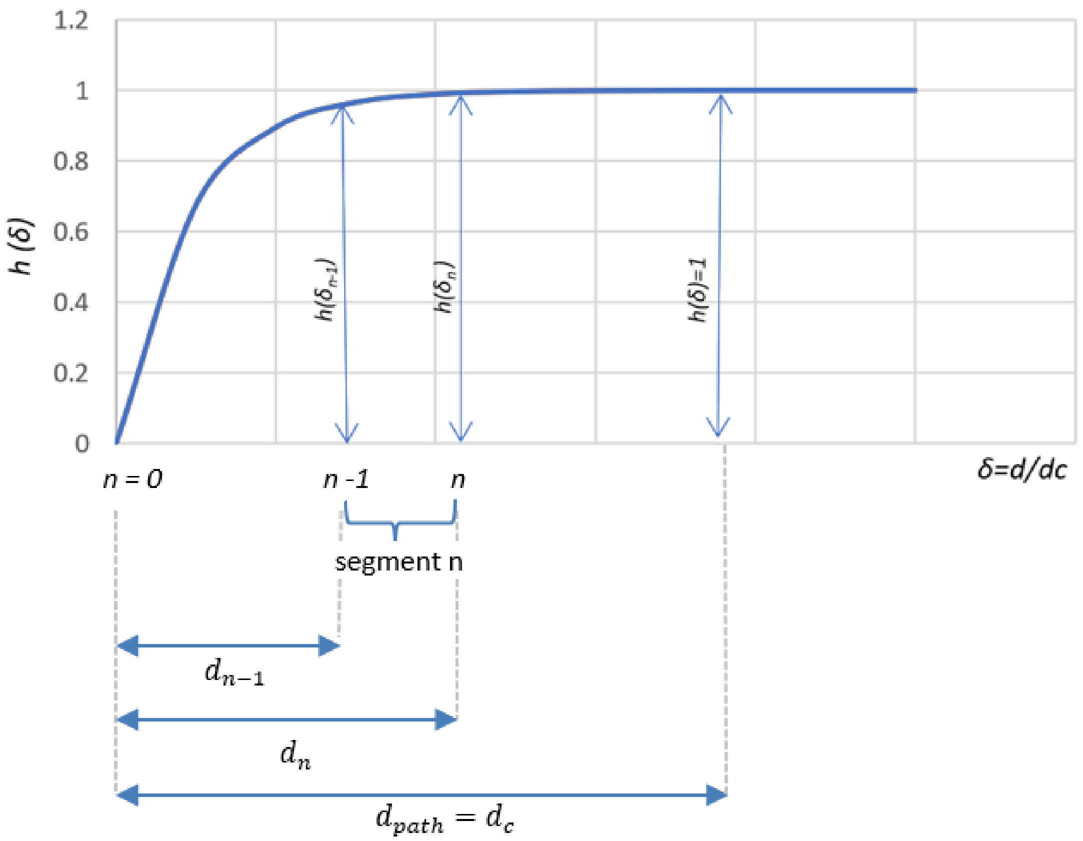


Figure 3.2 The dimensionless correction factor h(δ) as a function of travelled and cold distances, where n represents a segment of the path.

Regarding the presented methodology, information on the O-D pair of each trip made within the study area is crucial to calculate trip-based cold start emissions and attribute them to the road network. For this purpose, data from VISUM are extracted (see Figure 3.3) considering: (i) road segments (segment ID, number of private cars, segment length, vehicle speed, etc.); (ii) paths (origin, destination, path ID, volume); and (iii) the road segments belonging to each path. These data are exported as .txt files and used as inputs for the CSEE module developed in Python Environment.

Figure 3.3 schematically presents the methodology developed in this study for cold start emission quantification at road segment level.

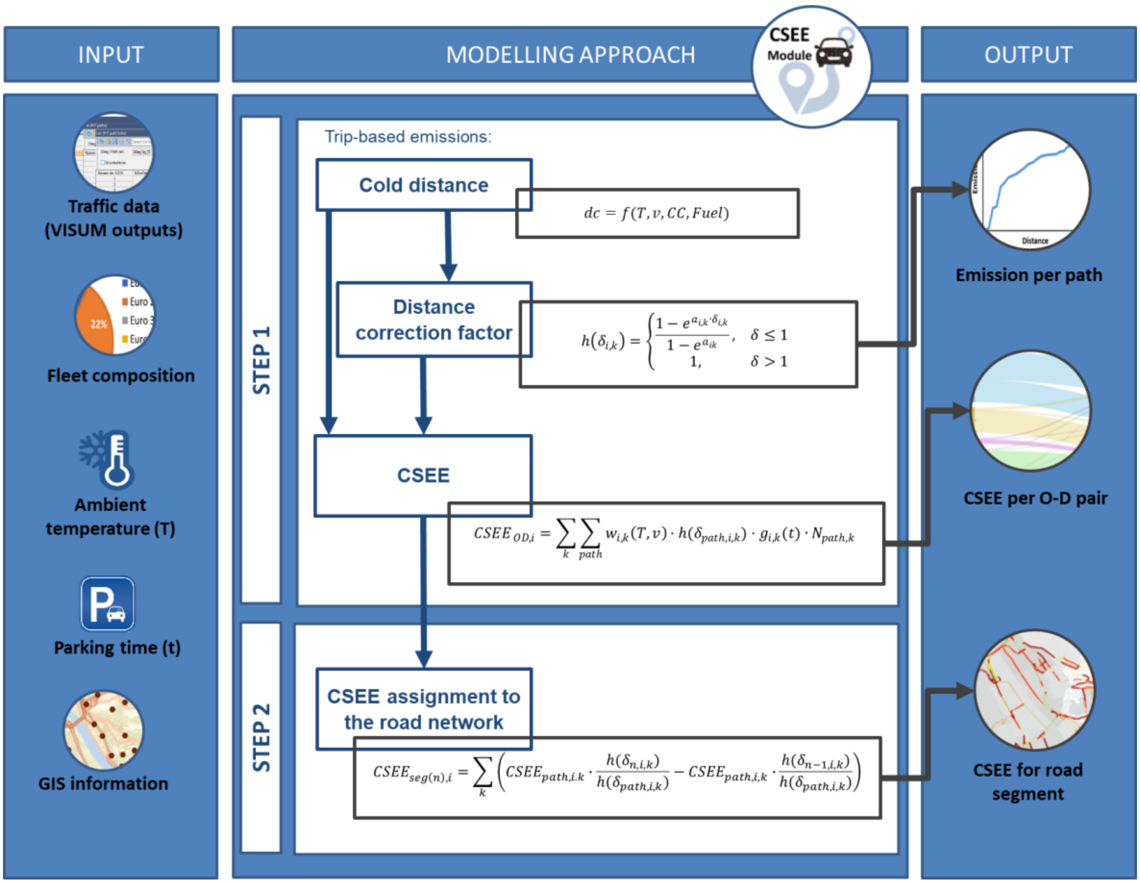


Figure 3.3 Conceptual framework of the CSEE module implemented in QTraffic.

3.3. Case study

The CSEE module developed in this research was applied to the city of Coimbra to quantify cold emissions of carbon monoxide (CO), carbon dioxide (CO_2), oxides of nitrogen (NO_x) and hydrocarbon (HC) by considering 2019 as a reference year. Coimbra urban area (see Figure 3.4) is a part of a Municipality and a Region with the same name. According to PORDATA (2021), the average population density for the Municipality and

Region of Coimbra were around 441 hab.km^{-2} and 101 hab.km^{-2}, respectively. It is third most important urban centre of Portugal, after Lisbon and Porto, given its location and importance to the Centro Region and historical significance. Combining its strategic location and its attractiveness to study, work, health activities and leisure, important commuting movements takes place in Coimbra urban area. Therefore, several travels have their origin or destination in this urban centre, which is a crucial reason for quantifying the cold start emissions in this area. Although air quality has improved somewhat in recent years, some exceedances of the limit values defined by the European Air Quality legislation (Directive 2008/50/EC) have been recorded in the study area (Dias et al., 2016; Tchepel et al., 2020). Moreover, new Guidelines recently proposed by World Health Organisation (WHO, 2021) require more ambitious objectives for air pollution reduction. Being the road transport one of the most significant sources of air pollutants in the urban context (APA, 2017), it is crucial to improve the quantification of the pollutant emissions and including the cold starts is crucial to guarantee the completeness of the emission estimates.

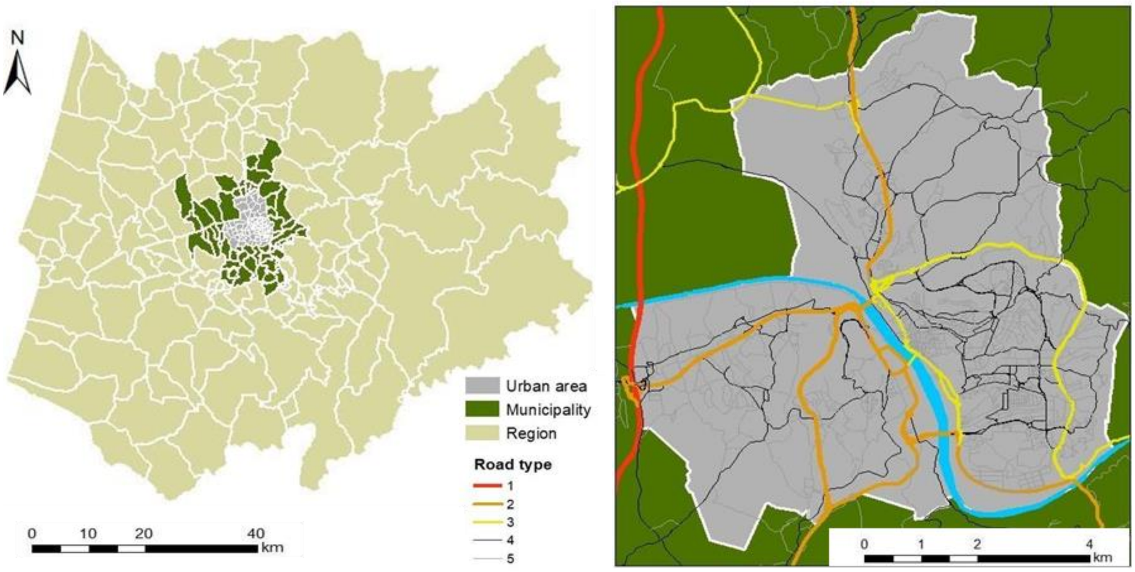
Regarding the previously described methodology, several input data are required (see Figure 3.3) to apply the CSEE module. The compilation of these inputs is described below:

3.3.1. Traffic data (VISUM outputs)

The information for each trip (number, average speed, origin and destination), requested for the CSEE step 1, were obtained from the VISUM traffic model applied for Coimbra. To characterise transport supply and demand in the study area, information from previous studies (TIS, 2011) was adapted considering: population and employment data by the zones; mobility indicators (travel generation rates, etc); private and public transport networks; transport costs per unit (fuel, parking and tariffs). According to Wardrop's principle (Ortúzar and Willumsen, 2011), the traffic flow and average driving speed for each road segment of the transportation network was determined taking into account congestion effects. For the calibration and validation of the model the following information was used: travel distances; modal split; traffic for private transport and passenger counts of public transports; observations on trips duration.

The scope of this study is to analyse cold start emissions within an urban area. However, the calculation of cold start emissions depends on the trip origin, which may be located outside of the study area. Therefore, the cold start emissions were obtained for a larger area. The Region selected (see Figure 3.4) has around 5835 km^2 and its transportation road network consists of 283 traffic generation zones (136 inside the urban centre).

About 18000 road segments were considered in this study with an average length of 130 m in urban area and about 600 m for the entire region. Traffic demand required for the transportation simulations was characterized based on daily O-D matrix considering different purposes, and transport modes (TIS, 2011). Figure 3.5 presents the modal share for passenger transport in the study Region considering active modes, private vehicles, public transport ant their combinations. Around 74% of the daily trips are made by private vehicles, which were selected for the application of the module. Based on previous studies (TIS, 2011), most of the daily trips occur in two periods: MP - morning period (from 8 AM to 10 AM; 25% of daily trips for the Region) and AP - afternoon period (from 4:30 PM to 7:30 PM; 30% of daily trips for the Region). Traffic counts recently performed within the study area confirmed these assumptions. The selection of both periods regards not only their representativeness in the traffic daily profile, but also the fact that cold emissions are higher with longer parking time. Although some travels occur in other periods of the day, they occur with a lower parking time and processed in the study as hot emissions only. We must highlight that in this study we only analyse working days.



(a) (b)

Figure 3.4 Traffic zones (a) and urban road network (b) considered in the study.

Figure 3.4b presents the urban road network by considering 5 road types, which will be crucial for the result analysis. The following hierarchical structure is considered for the road classification taking into account functional classes used in electronic navigable maps (NAVTEQ, 2011):

- Type 1 is attributed to roads that are allowed for high volume and maximum driving speed. These roads may have very rare or no speed variations.
- Road type 2 is used to direct traffic on road type 1 for travel between and through cities by the fastest way.
- Road type 3 links type 2 together and offers high traffic volumes with a lower level mobility than type 2.
- Type 4 connects with higher levels to collect and distribute road traffic between neighbourhoods at a moderate speed.
- Type 5 all other roads, including pedestrian streets.

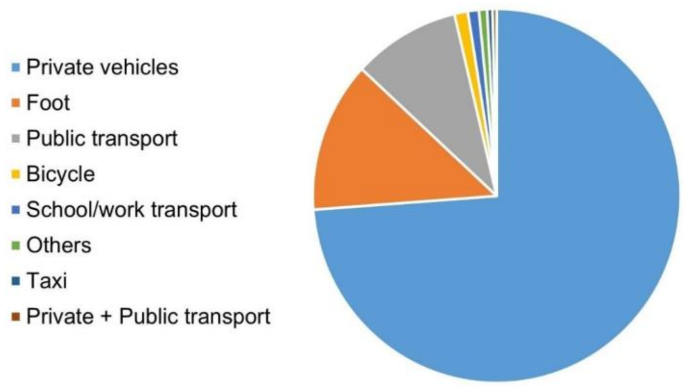


Figure 3.5 Classification of daily trips by mode for the study Region. (Notes: “Private vehicles”: trips done by motorcycles or passenger cars; “School/work transport”: trips done by school or company vehicles (usually buses or minivans); “Others”: ambulances, tractors and others; “Private + Public transport”: trips done with a combination of private vehicles and public transport).

Due to importance of parking time, the cold start emissions were calculated separately for morning and afternoon periods. Hence, the daily cold start emissions are a result of a sum of both periods. It should be stressed that the number of starts at afternoon period within the urban area (UA) is significantly higher than at morning period due to trips “return to home”. The average trip distance and average cold distance are represented in Figure 3.6 for different clusters distinguishing O-D inside and outside the urbans area. Thus, the cold distance represents about 1/5 of the total trip with origin and destination outside the urban area, while for the trips that totally occurs inside the urban area it represents almost the total trip (around 96%).

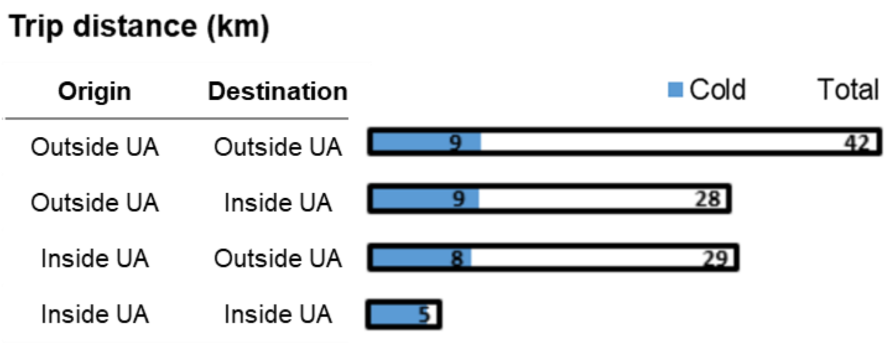


Figure 3.6 Distance of total trip and under cold engine (blue bar), considering ambient temperature for winter season (explained below) and both afternoon and morning periods.

3.3.2. Ambient temperature

In order to calculate the cold distance for each pollutant and the cold start emission factors, seasonal variations of ambient temperature were analysed (Table 3.2) using observations available for 2019 from the national meteorological station located in a proximity to the study area. To characterise the temperature for morning and afternoon periods for each season, median values were selected in order to reduce the impact of outliers.

Table 3.2 Local ambient temperature in 2019 considered for CSEE simulations.

Period	**Season**	**Ambient temperature**			
		Mean	Minimum	Maximum	Median
Morning Period	Winter	8.7	1.6	16.5	8.2
	Spring	14.2	4.5	27.5	12.0
	Summer	18.2	12.5	28.7	17.5
	Autumn	13.1	4.5	20.2	14.0
Afternoon Period	Winter	13.3	7.1	22.7	12.0
	Spring	18.5	7.7	33.7	17.5
	Summer	23.5	14.7	34.2	22.7
	Autumn	16.1	8.2	27.2	18.5

3.3.3. Parking time

The parking time is required to calculate cold start emission factors. As a simplification, it was assumed that all starts at the morning period are initiated after the parking time equal or higher than 12h, which means that the trips occur with a fully cooled engine (André and Joumard, 2005), while the parking time for the AP is influenced by working hours (in Portugal, 8h working hours per day). Therefore, for cold emission the $g(t)$ (i.e. is the correction factor for the parking time t) in Equation 3.2 will be 1 for the MP, while for AP it will take into account the 8h parking time, resulting in correction factor $g(t)$ less than 1 and thus lower emissions during the warm-up period.

3.3.4. Fleet composition

The information on vehicle technologies (i.e. Euro classes) and fuel properties is important to define the cold distance (see Equation 3.2 and Table 3.1) and the emission factors for each trip. The car fleet composition is presented in Figure 3.7. While the information on fuel properties was obtained based on statistical information for 2019 (INE, 2020), the distribution by Euro classes was obtained from local measurements performed in selected crowded road of the studied transportation network (Martins, 2020).

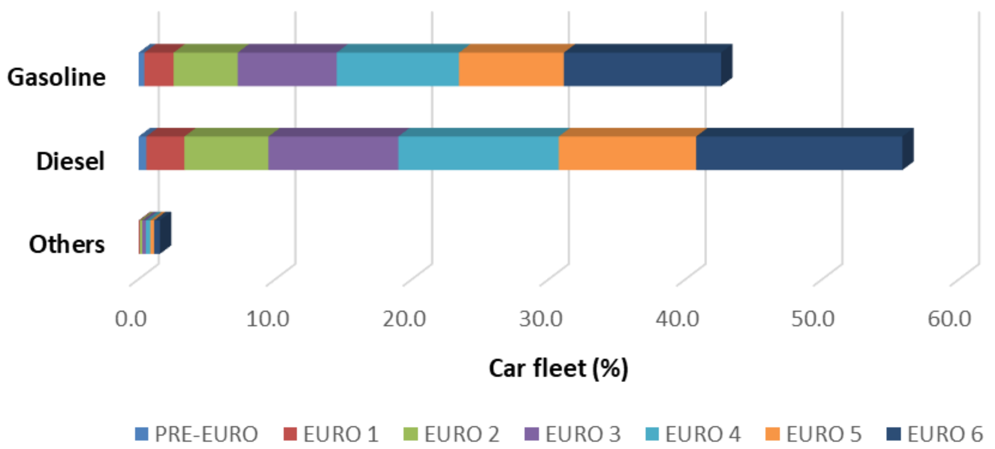


Figure 3.7 Distribution of Coimbra's passenger car fleet by fuel properties and Euro emission standards for 2019.

3.4. Results and discussion

Considering the research objectives, this section is divided in three subtopics. Firstly, a sensitivity analysis of the results provided by CSEE module is performed. Secondly, the daily exhaust emissions obtained for the case study are analysed focusing on cold and hot contributions. Finally, the cold start emissions at road segments are evaluated.

3.4.1. Sensitivity analysis

In order to support the analysis of the results obtained in this study, the cold start emission factors are investigated and an example for Euro 4 gasoline and diesel passenger cars, that correspond to most of the modelled fleet, is presented. Figures 3.8 – 3.10 present the emissions as a function of the ambient temperature, parking time and average speed, respectively. This analysis is focused on the amount of pollutants emitted during the entire cold distance and the results are represented in terms of g/start (and not g/km).

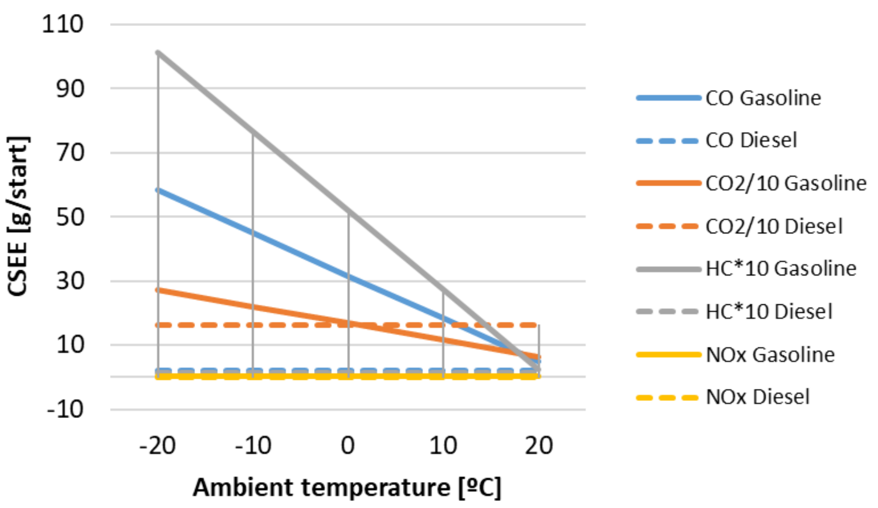


Figure 3.8 Sensitivity of cold start excess emissions (g/start) to ambient temperature for Euro 4 gasoline and diesel passenger cars (average speed = 20 km/h; parking time = 12h).

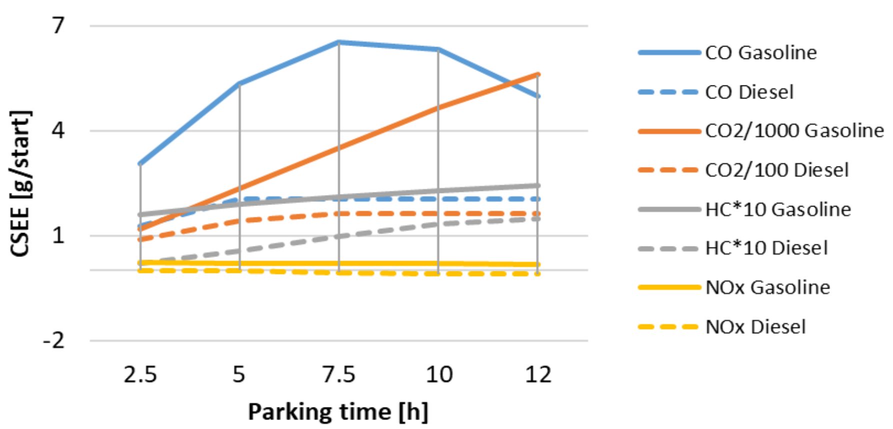


Figure 3.9 Sensitivity of cold start excess emissions (g/start) to parking time for Euro 4 gasoline and diesel passenger cars (average speed = 20 km/h; ambient temperature = 20ºC).

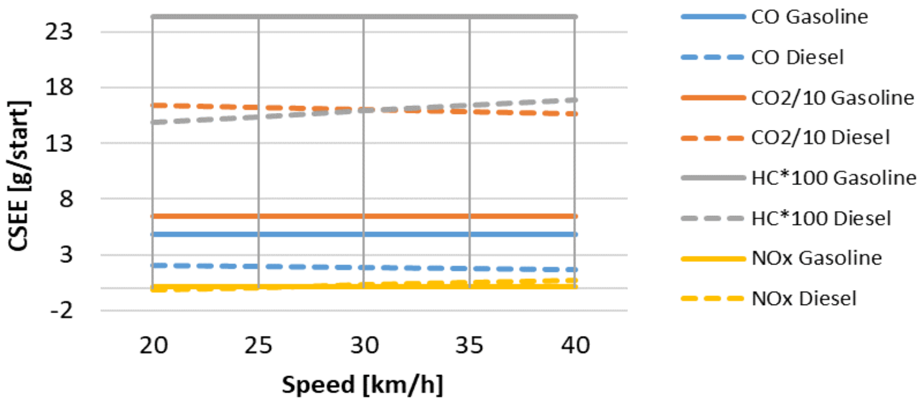


Figure 3.10 Sensitivity of cold start excess emissions (g/start) to average speed for Euro 4 gasoline and diesel passenger cars (parking time = 12h; ambient temperature = 20ºC).

Compared to CO_2 and HC, CO emissions from gasoline cars grow steeply at lower temperatures, while NO_x emissions do not change. For diesel cars, the effect of ambient temperature is explicitly considered for technologies older than Euro 3 only (Table 3.1) and, therefore, the results presented in Figure 3.8 for diesel cars are affected by this assumption. With respect to parking time, the emissions of HC and CO_2 increase till 12h of parking time, while CO emissions demonstrate non-linear behaviour and NO_x tend to decrease with the parking time. Although the average vehicle speed is very important for the cold distance calculation, the cumulative emissions along this distance expressed in $g.start^{-1}$ will be little affected by speed.

3.4.2. Contribution of cold starts to the daily exhaust emissions

The total daily emissions (in g/day) for Coimbra urban area are presented in Table 3.3 for different seasons. All the pollutants analysed in this study (CO, CO_2, HC and NO_x) have significant contribution of cold start emissions as presented in Figure 3.11. As expected, the contribution of cold emissions for the NO_x daily emissions is much lower than for the other pollutants. For winter season the contribution of cold emissions to CO and HC is higher than 50%, because of the influence of ambient temperature (see Table 3.2 and Figure 3.11). It is important to highlight that these emissions are estimated using the median temperature for the winter season (8.2ºC for MP and 12ºC for AP), while the minimum observed is 1.6ºC and 7.1ºC (respectively) that will result in even higher emissions (around 9% to 25% increase, depending on the pollutant considered). Table 3.3 also presents the daily emissions per inhabitant and per vehicle kilometres travelled (VKT) in parenthesis, which may be relevant for comparison with other studies.

Table 3.3 Total daily emissions (hot+cold) in the Coimbra urban area considering different seasons.

Season	CO			CO_2			HC			NO_x		
	1	2	3	1	2	3	1	2	3	1	2	3
Winter	1.62	17.93	0.33	688.95	7641.94	140.99	0.19	2.05	0.04	1.26	14.01	0.26
Spring	1.34	14.88	0.27	639.71	7095.75	130.91	0.15	1.63	0.03	1.26	13.98	0.26
Summer	1.05	11.66	0.22	583.34	6470.48	119.38	0.10	1.16	0.02	1.26	13.95	0.26
Autumn	1.27	14.10	0.26	624.72	6929.40	127.84	0.14	1.51	0.03	1.26	13.97	0.26

1-Total daily emissions for the domain ($t.day^{-1}$)
2- Daily emissions per inhabitant ($g.day^{-1}.inhab^{-1}$)
3- Daily emissions per VKT ($g.day^{-1}.VKT^{-1}$)

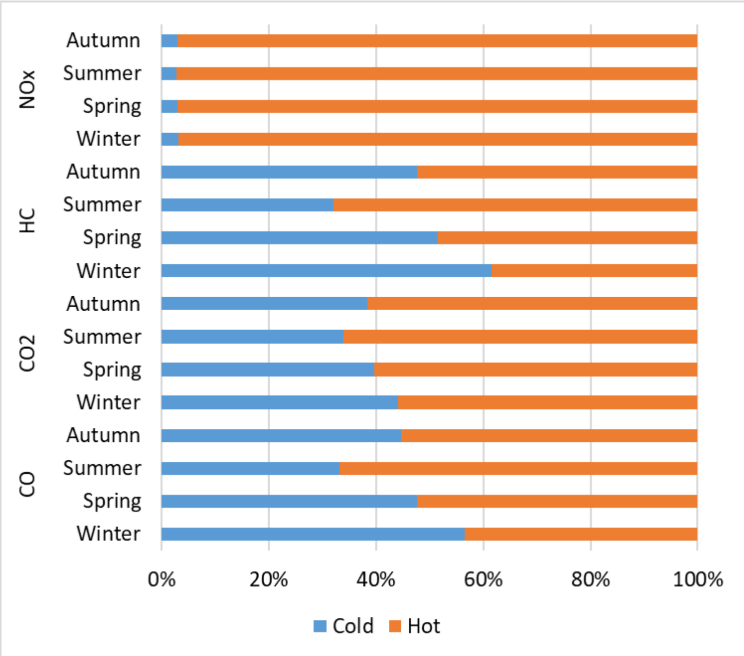


Figure 3.11 Contribution of cold and hot emissions to the total daily exhaust emissions in Coimbra urban area.

Figure 3.12 presents how the daily CSEE can be related to the origin and destination of a trip. First, it is important to stress that a large part of the trips under cold engine is related with trips that start or finish outside the urban area. In Figure 3.12 (right) only the CSEE that results from travels with O-D within the urban area are emphasised and aggregated by administrative units (parishes). Overall, the parishes that most generate trips and consequently emissions are also the most attractive parishes, except for parishes number 8 (“Eiras e São Paulo de Frades”) and number 13 (“São Martinho do Bispo e Ribeira de Frades”). This is because in the urban area, especial in the centre of the city, residential areas may be located near working/studying areas. Therefore, the traffic induced CSEE at the beginning of a trip (residence or work), which will contribute to the local air pollution, will have its impacts in the population that are driving for their work (in the morning) or their residence (in the evening). However, is also important to point out, that the analysis at the parishes level may be too much aggregated to make these conclusions, therefore, further research must be done.

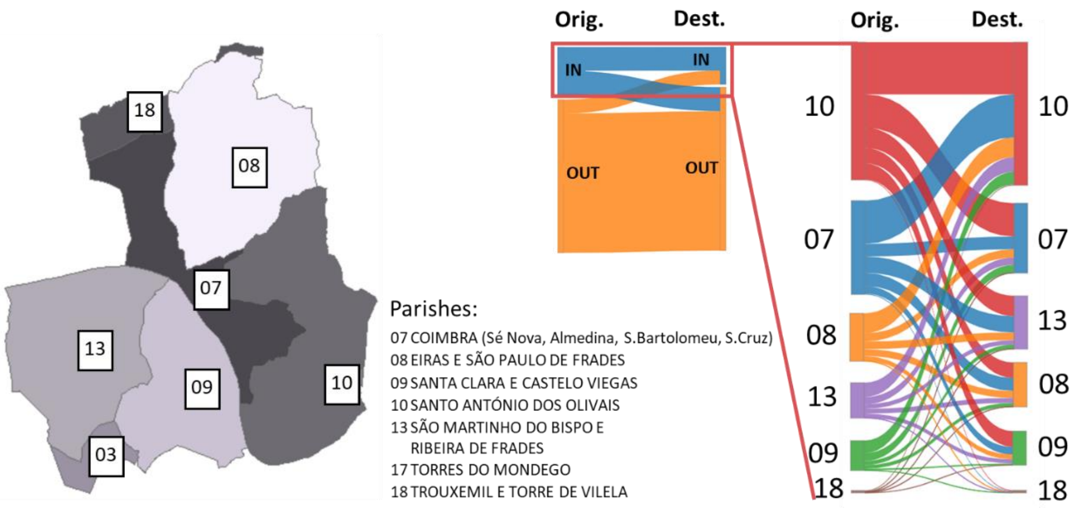


Figure 3.12 Distribution of daily cold start emissions, considering Origin and Destination of trips, for different administrative units in Coimbra urban area.

Information presented in Figure 3.13 highlights different contribution of cold emissions during the year. In all cases more than 50% of the CSEE occurs during the Morning Period due to lower ambient temperature and longer parking time. Furthermore, cold distance tends to decrease with the increase of ambient temperature (for instance for CO_2 it will reduce on average half a km from winter to summer). Therefore, due to lower ambient temperature during the morning period the cold distance will be higher, increasing the cold start emissions per vehicle during this period. AP and MP contributions will be different for different locations (Figure 3.14). Thus, AP contribution is very important in the locations with high proportion of commuting trips coming from outside the city. If during the morning period vehicles are entering to the urban area mainly with hot engine, the “return to home” trips are starting in the city and thus contribute to urban cold start excess emissions. Figure 3.14 presents the influence of a trip’s origin to the CSEE. One can see that for the AP most of the CSEE are related to the working/studying attraction poles, while for the MP the CSEE contribution is higher in residential areas (reinforcing the conclusions from Figure 3.12).

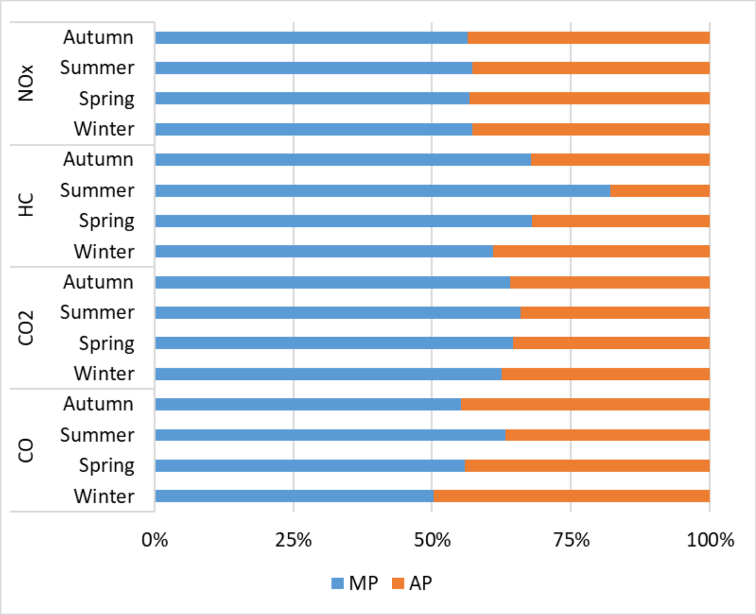


Figure 3.13 Contribution of each period (MP – Morning Period and AP – Afternoon Period) considered for the calculation of daily CSEE (in g/start).

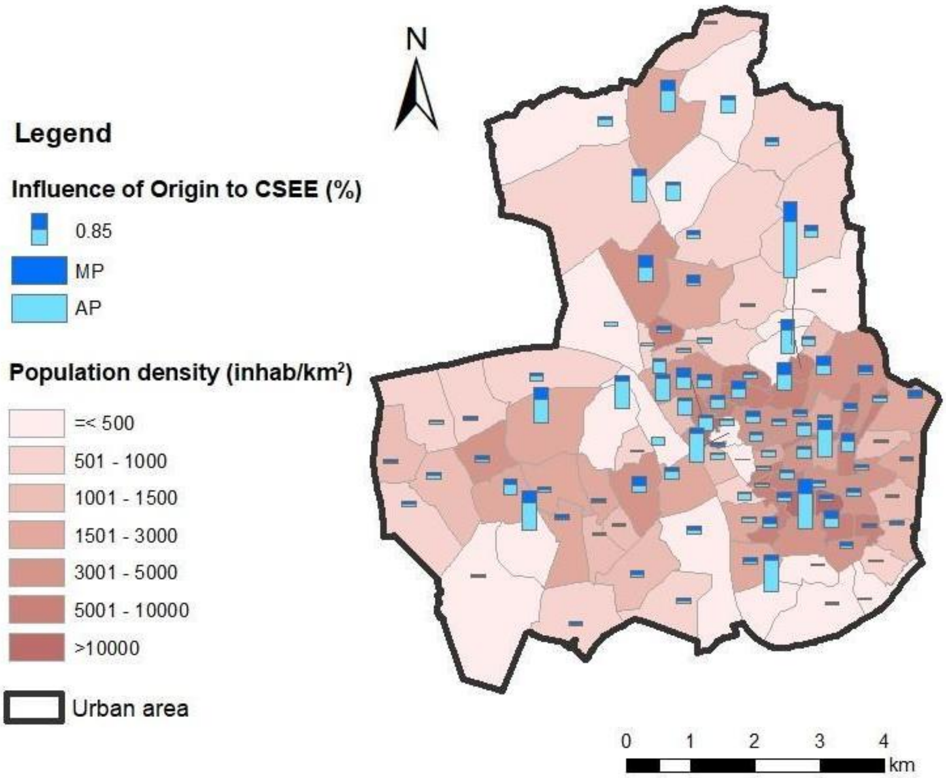


Figure 3.14 Influence of a trips' origin to cold start excess emissions at Morning Period (MP) and Afternoon Period (AP).

3.4.3. Cold start excess contribution for line sources

In this subsection the CSEE at the road segment level for the worst situation (winter season) is analysed. As an example, line sources are presented in Figure 3.15 showing CSEE contribution to the total daily emissions of CO. Overall, it is demonstrated that the greatest contributions occur on local roads (Type 5). This finding is consistent with other research focused on cold start emissions at city level, such as Faria et al. (2018). Local roads mainly offer land access, providing the lowest mobility level.

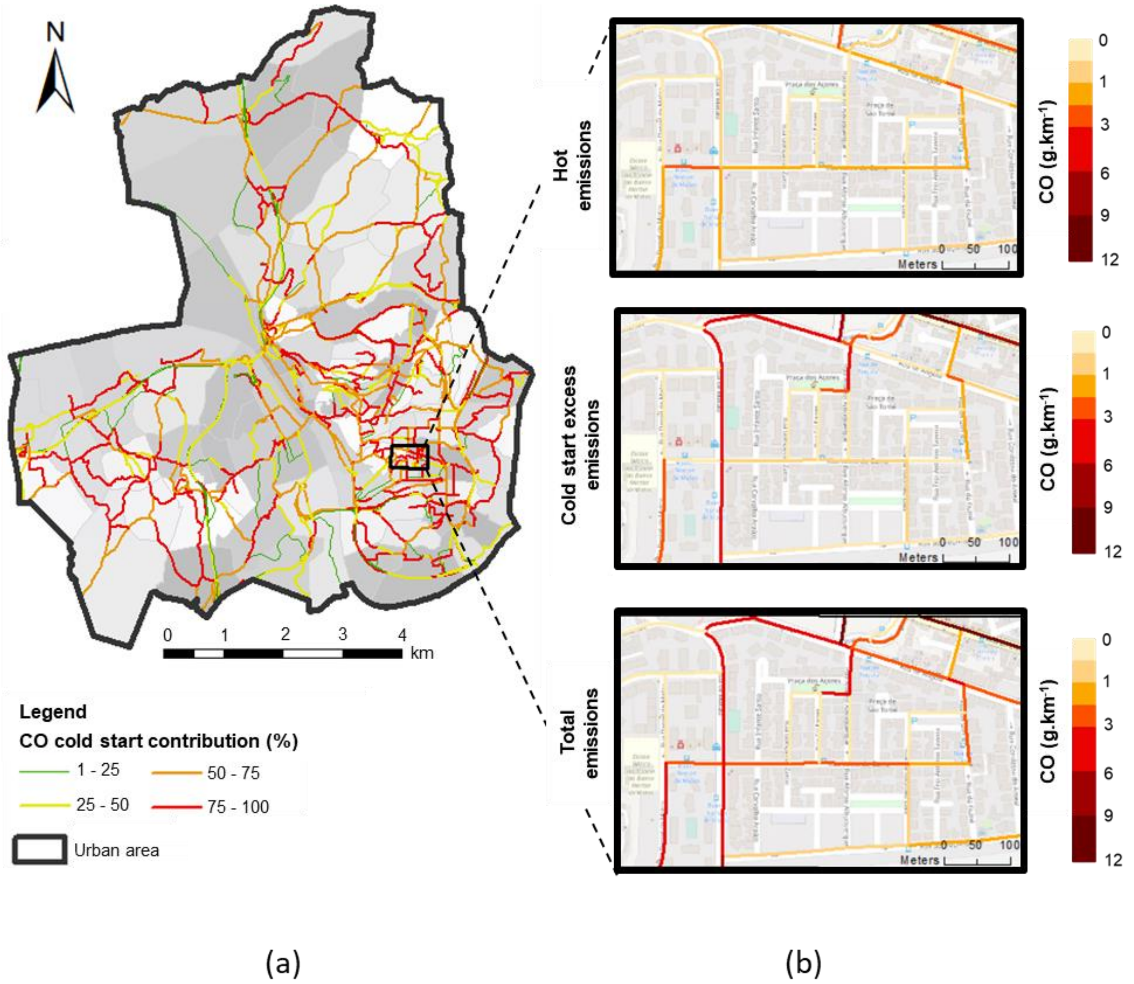


Figure 3.15 a) Contribution of cold start CO emissions to the daily total (hot+cold) emissions for line sources (considering winter season); b) and an example of the hot, cold and total emissions for a selected residential area in Coimbra.

The frequency distribution of the data is depicted in Figure 3.16. A fraction of CSEE in the total (hot+cold) emissions is presented on vertical axis and the histogram analysis reveal how often different values occur on the road segments within the study area. It could be observed that for NO_x the values are concentrated in a narrow range, mainly below 0.1 (i.e. CSEE contribution <10%). Also, the road segments classified as Type 1 reveal very low contribution of CSEE for all the pollutants that also confirmed by the statistics summarised in Table 3.4. However, it should be noted that only a small sample of the road segments classified as Type 1 is presented in the study domain, including a part of motorway close to the road entrance. Therefore, the results should not be extrapolated outside the urban area where, in general, no cold emissions could be expected for the Type 1. A left-skewed distribution is observed for other situations with an increasing importance of CSEE on the roads of Type 4 and Type 5. Thus, a significant number of values for CO and HC at local roads (Type 5) is above 0.9, i.e. CSEE contribution >90%. Also, local roads are characterised by higher variability in the comparison with other type of roads that confirmed by standard deviation and interquartile range presented in Table 3.4. Quantification of emissions with fine spatial resolution reveals inhomogeneities of CSEE contribution within the urban area and highlights a significant difference between the total values in urban inventory in a comparison with the results obtained for road segments. For example, cold start emissions contribute only 3% for daily NOx emissions at urban scale, but at individual road segment may achieve 43%. This contribution is different for the pollutants analysed achieving 97%, 92 % and 98% for CO, CO_2 and HC respectively. An example for CO is presented in Figure 3.15 showing inhomogeneities of cold start contribution at road segment level and the importance of emission quantification with fine spatial resolution. Thus, the example presented for a selected area (Figure 3.15b) reveal that cold emissions might duplicate or even quadruplicate in comparison to hot emissions.

According to Reiter and Kockelman (2016), the cold start distance varies considerably from study to study. André and Joumard (2005) assessed a broad distribution (by pollutant) of cold start distance with an average of 5.2 km at 20ºC. In Figure 3.6, one may see that the parcel of a trip made with a cold engine will vary according to the region analysed (outside the urban area: 21%; inside: 96%). Therefore, in densely developed cities a significant part of urban trips take place with a cold engine. The methodology applied in the current study demonstrates that the impact of cold starts increases from arterial to local streets. This founding is in agreement with previous studies (e.g. Faria et al., 2018) highlighting that higher emissions occur where most population live or work. Furthermore, from Figure 3.14 it is conclusive that most of the starts within the urban area that impact the daily emissions happen during the afternoon period (“return to home”), therefore, future mobility strategies should take this into consideration. With the analysis provided in this study it is possible to determine CSEE hotspots and trace their origin, which may play a key role in achieving the sustainable development goals regarding air quality and climate change issues. Therefore, future mobility and air quality policies should consider cold start impacts.

Thus, the advantages of different parking/transportation strategies could be evaluated considering additional benefits of the avoided emissions, in this case due to cold start emissions reduction. For example, environmental benefits of flexible-time communing or multimodal solutions, like Park&Ride, would be more evident if cold start emissions reduction could be considered additionally to hot emissions. However, such evaluation is only possible based on methodologies with fine spatial resolution and preserving O-D information to establish cause-effect relationships. Therefore, the modelling approach developed in this work could provide crucial input for the evaluation of environmental benefits and comparison of different transportation policies considering emissions reduction (hot+cold) at urban scale.

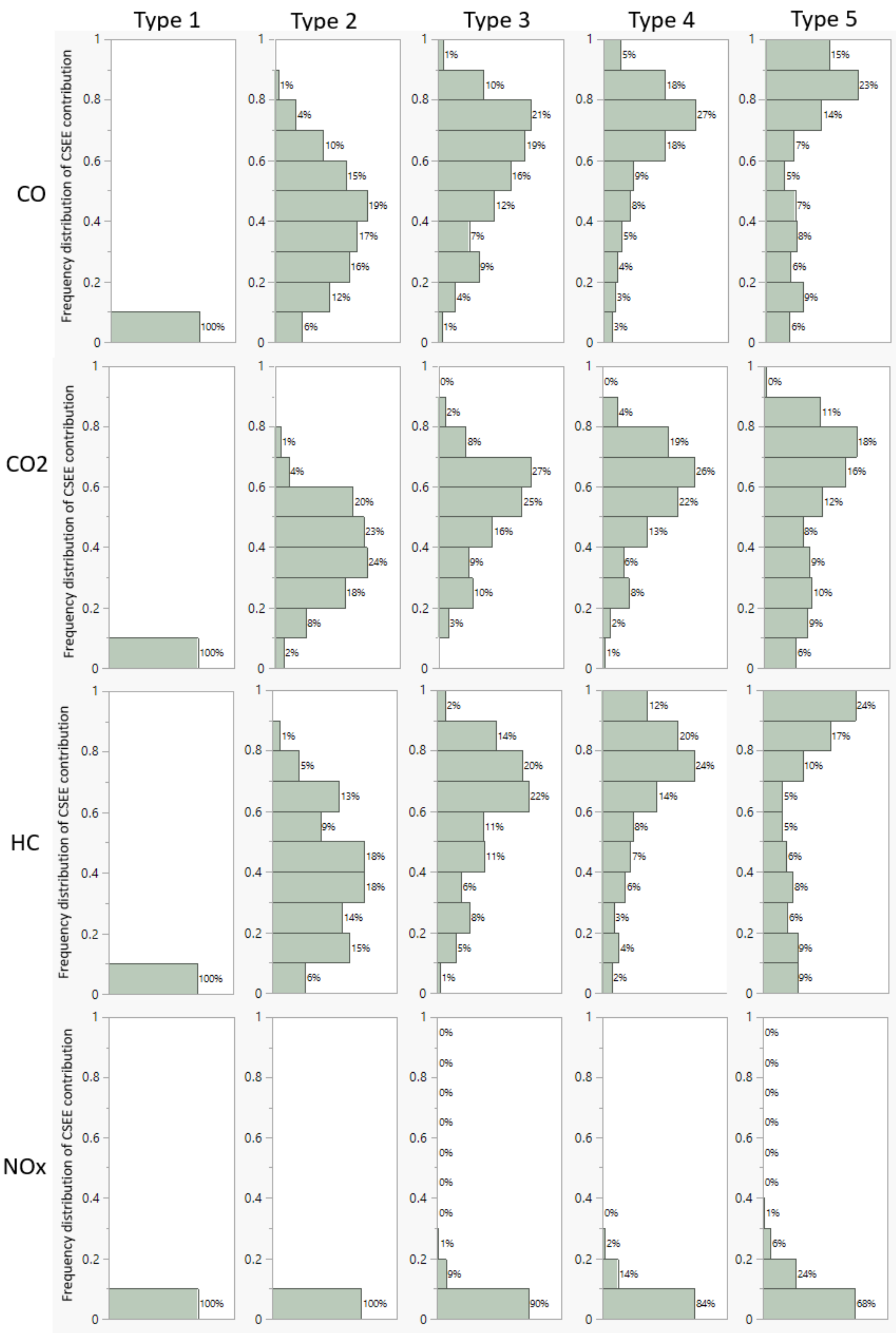


Figure 3.16 Frequency distribution of CSEE contribution to daily emissions (hot+cold) for different type of roads during winter season.

Table 3.4 Summary statistics of CSEE contribution (%) to daily emissions (hot+cold) for different type of roads considering winter season.

Pollutant	Type	Mean	Std Dev	75th Percentile	Median	25th Percentile
CO	1	0.7%	1.4%	1.2%	0.0%	0.0%
	2	39.3%	18.6%	53.8%	39.8%	23.9%
	3	57.3%	20.0%	73.3%	60.8%	45.3%
	4	63.8%	21.7%	79.3%	70.1%	51.6%
	5	60.8%	29.7%	87.4%	72.2%	33.3%
CO_2	1	0.9%	1.5%	1.7%	0.1%	0.0%
	2	39.5%	14.4%	51.1%	38.4%	29.1%
	3	52.6%	16.1%	64.5%	55.4%	41.5%
	4	57.0%	16.9%	68.9%	59.7%	46.7%
	5	52.0%	25.1%	73.4%	58.6%	30.5%
HC	1	0.8%	1.6%	1.4%	0.0%	0.0%
	2	38.4%	19.9%	52.1%	37.4%	22.8%
	3	59.4%	20.9%	75.5%	63.5%	44.2%
	4	65.9%	23.0%	82.6%	72.8%	52.7%
	5	60.4%	32.0%	89.9%	72.1%	31.5%
NOx	1	0.1%	0.1%	0.1%	0.0%	0.0%
	2	3.9%	2.5%	5.6%	3.4%	1.9%
	3	5.9%	6.7%	7.5%	4.8%	2.8%
	4	6.0%	4.9%	7.6%	4.6%	2.7%
	5	8.0%	10.3%	12.5%	4.8%	1.2%

3.5. Conclusions

In this paper, a new modelling approach to quantify cold start emissions at road segment was proposed allowing spatial analysis of the cold start emissions for line sources. The CSEE module was developed and applied to the Coimbra urban area (Portugal). Both cold and hot emissions for line sources were calculated to obtain the total daily emissions and the cold start contribution.

The frequency distribution and summary statistics of the cold start contribution at a road segment level was analysed for different types of roads. For winter season, a high contribution of cold start emissions is demonstrated with median values above 70% for CO and HC at road segments classified as Type 5.

The total daily emissions (hot+cold) of CO, CO_2, HC and NOx for the urban area were analysed considering their seasonal variations. For winter season the contribution of cold emissions for CO and HC is higher than 50%, but for NO_x is only 3%.

The cold start excess emissions during morning and afternoon periods were compared stressing the differences not only in absolute values but also in their spatial distribution. Thus, commuting trips of non-urban residents have no contribution to urban CSEE during the morning period if the trip is longer than “cold distance”, but their return trips originated near working/study locations in the city will be important for cold start emissions at afternoon period.

The modelling tool developed and applied in this study demonstrates the importance of quantifying cold start emissions with fine spatial resolution (i.e. at road segment level). There is large variability of CSEE contribution at urban roads that should not be neglected. Particularly, the data for local roads reveal higher variability that may contribute to the uncertainty of emission quantification in the locations where most people live or work. These variabilities were possible to obtain because the model approach preserves the trip O-D pair information, being possible to attribute the cold emissions to each road segment of the path. Furthermore, the O-D information of a trip enables to access the contribution of different regions to the cold start emissions. In this context, the bottom-up modelling of cold start emissions and the preservation of O-D information would provide valuable information for traffic pollution studies and quantification of environmental benefits due to emissions (hot+cold) reduction.

Chapter 4

4. Characterization of traffic-related particulate matter at urban scale

This chapter is based on the publishing paper: "Dias, D., Pina, N., Tchepel, O. (2019). Characterization of traffic-related particulate matter at urban scale. International Journal of Transport Development and Integration 3(2), 144 – 151".

4.1. Introduction

Road transport remains an important source of some of the most harmful air pollutants. In particular, road transport is responsible for significant contributions to emissions of particulate matter (PM), especially in urban environments and major cities. Despite slow improvements, PM concentrations continue to exceed European Union and World Health Organization limits and guidelines in large parts of Europe (EEA, 2018b). Potential impacts of air pollution on human health are investigated in several studies (Analitis et al., 2006; Lelieveld et al., 2015; Orru et al., 2011; Tchepel and Dias, 2011). It is demonstrated that in European countries the exposure to $PM_{2.5}$ concentrations reduce the average life expectancy by 8.6 months (Orru et al., 2011). Additionally, Analitis et al. (2006) found out from an investigation in 29 European countries, that mortality due to respiratory problems increased by 0.58% (0.21 to 0.95%) for every 10 $\mu g.m^{-3}$ increase of PM_{10}. Moreover, it is expected that the air pollution contribution to global premature mortality could double by 2050 (Lelieveld et al., 2015). Consequently, the air pollution remains the single largest environmental health risk in Europe (WHO, 2017), particularly in urban areas (Dias and Tchepel, 2018).

In addition to exhaust traffic-related PM emissions, the non-exhaust part of emissions such as those from brake wear, road wear, tyre wear and road dust resuspension have a large contribution to the atmospheric PM concentrations in cities. This contribution may be even larger than those from the exhaust sources, while due to continuous reduction of exhaust emissions, it is expected that the relative contribution of non-exhaust sources will increase in the forthcoming years (Amato et al., 2014). Despite the importance of non-exhaust traffic-related emissions, scientific knowledge on this source of PM is scarce, thus limiting the correct implementation of mitigation strategies to control the high atmospheric PM levels observed in cities.

Given the need to better inform policy-makers in the implementation and evaluation of cost-effective transport policies, it is essential to develop and implement methodologies to provide reliable and spatially resolved PM estimates within cities. However, estimating the temporal and spatial variabilities of traffic-related PM emissions and their concentrations at urban scale is challenging.

The aim of this study is to provide a comprehensive characterization of traffic-related PM spatial and temporal variation at urban scale, by using an integrated modelling approach and *in-situ* aerosol measurements. For this purpose, a modelling cascade based on transportation-emission-dispersion modelling was implemented for the urban area of Coimbra. The emission model has been extended to include a new module for quantifying emissions of different metrics of PM released to atmosphere from non-exhaust traffic-related sources, namely brake, tyre and road surface wear with detailed spatial discretization. These emission data will be one of the most important inputs to the urban dispersion modelling. Moreover, optical aerosol measurements, obtained during an experimental field monitoring campaign implemented at a city hotspot, are considered for the model validation.

4.2. Methodology

The Portuguese city of Coimbra, was selected in this study to estimate and measure traffic-related air pollution (Figure 4.1). This medium-size city, with approximately 150000 inhabitants, is the largest urban area in central Portugal. *In-situ* measurements campaign design and the integrated modelling system applied in this study are described in the following sections.

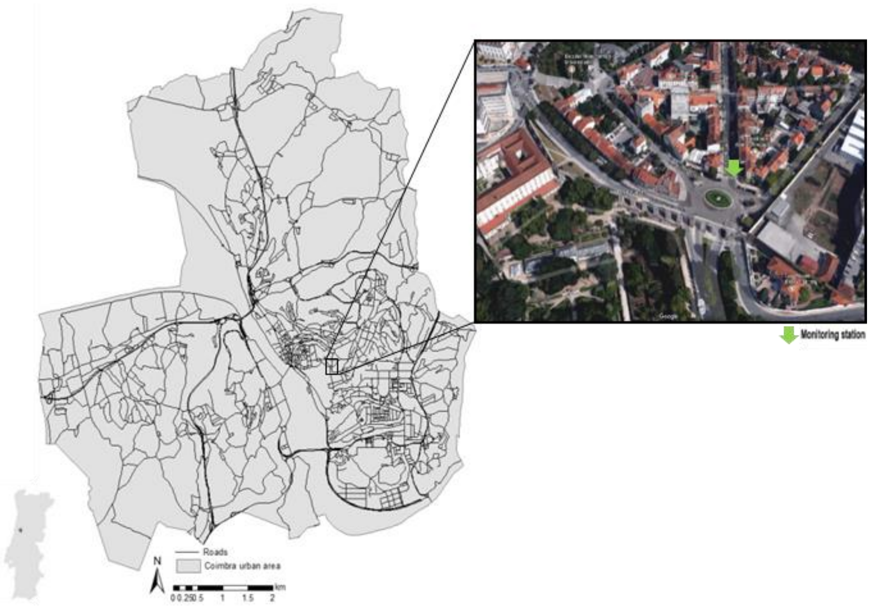


Figure 4.1. Study domain including road network and *in-situ* measurements campaign location.

4.2.1. In-situ measurements

An experimental field monitoring campaign took place in the central area of Coimbra, close to historic centre, during 5 days in June (20-24 June 2017). During this period, several parameters were collected. For characterizing PM levels, the distribution of PM in 31 size range from 0.25 to 32 μm was acquired by the GRIMM portable aerosol spectrometer (1 min resolution) (GRIMM, 2015). Meteorological conditions including wind speed and direction, temperature, precipitation and relative humidity, were measured by a mobile meteorological station. For the traffic volume characterization, eight counting point were used, providing information on temporal variation of traffic distinguished between four vehicle categories, including passenger cars, light duty vehicles, buses, and motorcycles.

4.2.2. Integrated modelling approach

The proposed modelling approach to evaluate traffic-related PM concentrations consists of three interconnected models: a transportation model, an emissions model and an air quality model (Figure 4.2).

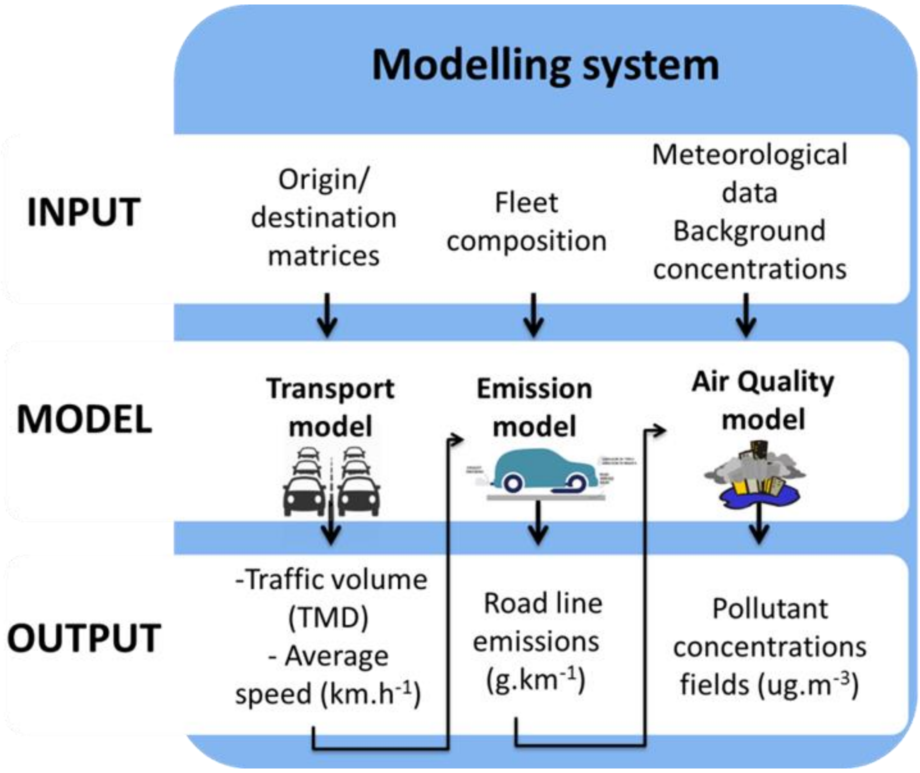


Figure 4.2. Integrated modelling system implemented for Coimbra urban area.

Firstly, a macroscopic transportation model VISUM (PTV, 2013) was applied to quantify transport activity data. Thus, based on the “four-steps” traffic modelling approach, data related with traffic volume and vehicle speed for each road segment were estimated by VISUM. Information on origin, destination, trip duration, route taken, average speed, road length and width of the city network was fully characterized. The outputs of this model were used as inputs by the emission model to quantify the emission amounts with high temporal and spatial resolution.

The Traffic Emission and Energy Consumption Model (QTraffic) was applied to Coimbra urban area in order to quantify PM emissions induced by road traffic from exhaust and non-exhaust sources. This model is based on the updated European guidelines, following an average-speed approach, and is particularly designed for line sources, estimating emissions individually for each road segment based on emission factors determined according to average speed and vehicle class (based on engine age, type, and capacity, vehicle weight, fuel type, and emission reduction technology). Thus, the input data required by the emissions model have been compiled on the basis of statistical information on fleet composition, spatial data on road network, and traffic volume and vehicle speed from the VISUM model. The detailed emission data are considered as important inputs to simulate air pollution dispersion at urban scale.

In this work, a version of the Atmospheric Dispersion Modelling System adapted for line sources (ADMS-Roads) is used. This model is based on the Gaussian approach enabling establishment of relationship between emissions and air quality at urban scale (CERC, 2017). Additionally to the emission data, a continuous time series of meteorological parameters (including wind direction, wind speed and cloudiness) are required for the model application. The results from ADMS-Road are analysed in terms of spatial distribution of pollutant concentrations with 1h resolution.

4.3. Characterization of traffic-related PM at urban scale

The characterization of traffic-related PM at urban scale by using an integrated modelling approach and *in-situ* aerosol measurements is presented and discussed in this section.

4.3.1. Local measurements results

The measured data were analysed for 5 days in June (20-24 June 2017). It should be noted that there have been forest fires during this period (until 22nd June) in the surrounding municipalities, as indicated in the Figure 4.3.

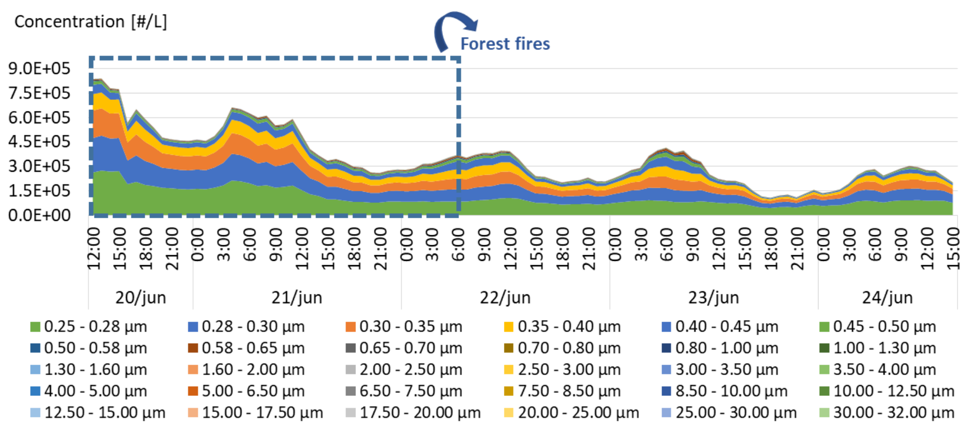


Figure 4.3. Temporal variation of particle number concentrations obtained from GRIMM.

The temporal variation of data acquired by the GRIMM portable aerosol spectrometer (1 min resolution), distributed in 31 size range from 0.25 to 32 µm, is evidenced in Figure 4.3. The influence of forest fires at the beginning of the campaign is reflected, mainly to the finest PM fraction (0.25 - 0.28 µm). After June 22, the temporal variation of the number of particles tends to follow the daily profile of road traffic sources.

Analysing the PM size distribution in terms of mass (Figure 4.4a) and surface area (Figure 4.4b) is possible to verify that the contribution of finest fraction (<0.5 µm) is more evident in terms of PM surface area in comparison to PM mass size distribution, mainly in the presence of forest fires (June 21). After the forest fires (June 23) the contribution of fine particles continues to be relevant, revealing a strong contribution of the anthropogenic sources to atmospheric PM levels measured in the study area.

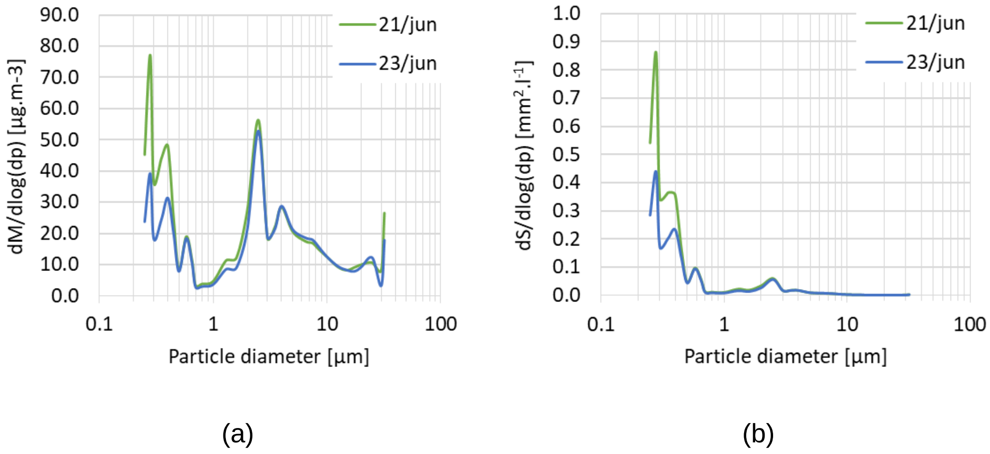
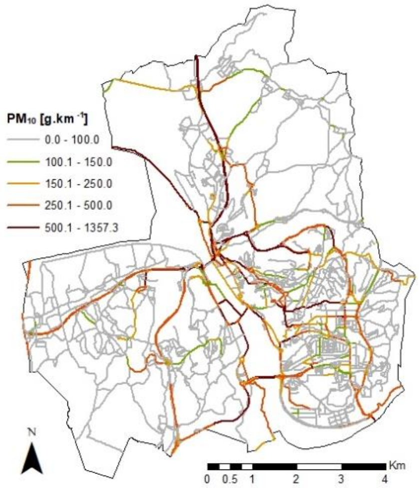


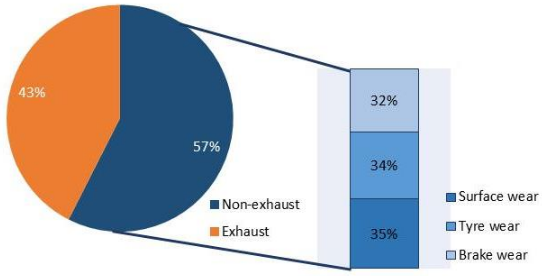
Figure 4.4. a) Distribution of hourly measurements of: a) PM mass; b) PM surface area obtained by GRIMM at 8AM of 21st June and 23rd June.

4.3.2. Integrated modelling results

The results presented in the following figures are obtained by the QTraffic model (PM emissions) and the ADMS model (PM concentrations).

Figure 4.5a illustrates, as an example, the daily PM_{10} emissions at each road in a typical working day. As is seen in Figure 4.5a, higher pollutant amount emitted by road transport to the atmosphere is observed for main city entrances as well as urban roads. Moreover, the emission modelling results are analysed in terms of individual contribution of non-exhaust sources to the total PM emissions estimated (Figure 4.5b). The outputs of the QTraffic indicate that in Coimbra urban area the non-exhaust emissions are more than half to the total traffic PM_{10} emissions, being practically equally attributed to brake, tyre and road surface wear.





(a) (b)

Figure 4.5. a) Spatial distribution of total daily PM_{10} emissions within the study domain; b) Source contribution to PM_{10} emissions.

Hourly and daily PM concentrations were estimated based on two traffic emission setups: (1) exhaust emissions only and (2) total (exhaust + non-exhaust) emissions. The PM hourly concentrations were compared with the PM observations from 22nd to 24th June. Figure 4.6 presents the scatter plot for the hourly concentrations resulting from total traffic emissions, where is seen that the correlation between the model outputs and observations is 0.74. The good performance of the ADMS model is evidenced by another statistical parameters such as the fraction of predictions within a factor of two of observations (FAC2 = 0.9), the mean normalised bias (MNB = -0.35) and the normalised mean square error (NMSE = 0.3). The comparison of two modelling runs shows that non-exhaust emissions are very important resulting in a maximum difference of 4.8 $\mu g.m^{-3}$ PM hourly concentration at the observation point on 23rd June at 9 AM. Overall, the model is underestimating PM concentrations and the most significant differences are obtained for nocturnal period that is not affected by traffic emissions.

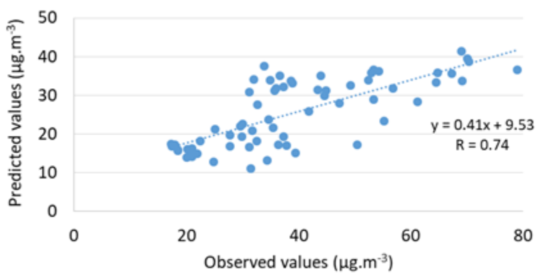
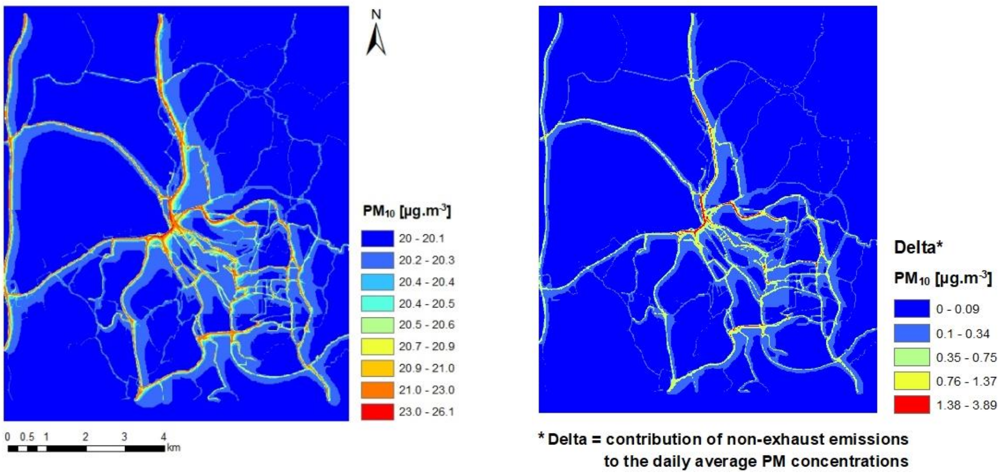


Figure 4.6. Scatter plot of PM_{10} hourly concentrations from 22nd to 24th June based on total emissions (exhaust and non-exhaust).

Figure 4.7 illustrates the spatial distribution of the PM_{10} daily mean concentrations for a selected day (23rd June, day without occurrence of forest fires). In the map is seen higher concentrations in the centre of the study domain (Figure 4.7a), where several activities occurs during the day and, therefore, people may be more exposed to harmful PM_{10} concentrations. Figure 4.7b presents a difference between the two model runs thus highlighting the contribution of non-exhaust traffic emissions that achieve 4 $\mu g.m^{-3}$ in terms of daily average concentration, and about 9 $\mu g.m^{-3}$ in terms of hourly concentration at 9AM. Therefore, the inclusion of these traffic emissions is very important as it enable to improve the model performance and identify the hotspots within the study domain.



(a) (b)

Figure 4.7. Spatial distribution of PM_{10} daily average concentrations within the study domain for 23rd of June: a) based on total traffic emissions (exhaust and non-exhaust); b) contribution of non-exhaust emissions.

4.4. Conclusions

In this study, a detailed characterization of traffic-related PM pollution at urban scale was performed by using an integrated modelling approach and *in-situ* aerosol measurements.

The *in-situ* measurements indicates a higher presence of fine particles in the first days of the field campaign, which is not justified by road traffic only and influenced by other sources such as forest fires. Furthermore, the study presents relevant data of PM distribution in 31 size range, which may be important for source apportionment and population exposure studies.

In terms of integrated modelling approach, the inclusion of non-exhaust emissions increases the PM_{10} hourly concentrations by 9 $\mu g.m^{-3}$, improving the modelling performance. Moreover, the application of the methodology enabled to identify some hotspots in the urban area by analysing the spatial distribution of the PM concentrations.

Chapter 5

5. Urban aerosol assessment and forecast: Coimbra case study

This chapter is based on the publishing paper: "Tchepel, O., Monteiro, A., Dias, D., Gama, C., Pina, N., Rodrigues, J. P., Ferreira, M., Miranda, A. I. (2020). Urban aerosol assessment and forecast: Coimbra case study. Atmospheric Pollution Research, 11(7), 1155-1164".

5.1. Introduction

Air pollution is a concern in many parts of the world with environmental and health impacts, especially in cities where population may be exposed to high levels of atmospheric pollutants. The air pollution was estimated to cause 3 million premature deaths in 2012 (WHO, 2016). Additionally, a report from the Organization for Economic Co-operation and Development (OECD) indicated that outdoor air pollution may cost the world $2.6 trillion a year by 2060 and the welfare costs associated with premature death could rise to as much as $25 trillion by 2060 (Hunt et al., 2016).

Human health is affected by different air pollutants, but some of them have more severe consequences, such as particulate matter (PM). According to health investigation, PM causes different degrees of damage to human respiratory, cardiovascular, immune systems, central nervous and to genes (Chahine et al., 2007; Lubinski et al., 2005). Scientific studies provide sufficient evidence of an association of exposure to air pollution with all-cause daily mortality, which is estimated to increase by 0.2–0.6% per 10 $\mu g.m^{-3}$ of PM_{10} (Samoli et al., 2008). On average, road transport and domestic fuel burning are the biggest anthropogenic sources of urban air pollution responsible for 25% and 15%, respectively, of particulate matter in the air (Karagulian et al., 2015).

Despite slow improvements, PM pollution levels continue to exceed European Union and World Health Organization limits and guidelines in large parts of Europe, especially in urban areas (EEA, 2018b). Therefore, improved understanding of these PM health effects requires additional information about urban air quality. Moreover, to prevent human exposure to air pollution episodes, it has become urgent to anticipate them and understand the trends of pollution. These concerns are in the core of the air quality forecast research.

The air quality forecast enables to establish air pollution alarm systems, which may effectively reduce the costs associated to air pollution. Despite the necessary control of air quality by monitoring stations and the air quality assessment by modelling, the urgency to forecast the air quality is also highlighted in the literature (Bai et al., 2018; Kukkonen et al., 2012; Zhang et al., 2012).

Forecasting pollution can be made at different scales (global/regional to urban/local) and using different methods (statistical and numerical models). Nowadays, the progress in global and regional air pollution forecast is notorious, with several online platforms (e.g. World Air Quality Forecast, https://waqi.info/forecast/#/; NOAA's National Weather Service, https://airquality.weather.gov/; Copernicus Atmosphere Monitoring Service (CAMS) service, https://atmosphere.copernicus.eu/; AirNow, https://airnow.gov/). However, the number of studies focused on urban scale forecast is very few (Urban Air®, https://www.numtech.fr/actualites.php?id=172; AIRPARIF, https://www.airparif.asso.fr/; London Air, https://www.londonair.org.uk/LondonAir/nowcast.aspx; CAMS, 2018).

At urban scale, the complex spatial and temporal variations of pollutant concentrations is a result of several factors such as emissions, meteorological conditions and urban morphology. Moreover, urban background pollution levels are influenced by the transport of pollutants from long distances. Therefore, the urban scale modelling should consider local scale effects together with the transport and chemical transformation of pollutants that occur at a wider scale. There is no simple solution how to determine the background pollution concentrations and several methodologies have been proposed (e.g. Han et al., 2015; Tchepel et al., 2010). Although background concentrations could be extracted from observations and used for modelling past air quality, application of such methods for operational forecast is limited. For this purpose, regional operational services may provide information on background pollution levels to be used at urban scale forecast. Nevertheless, a double counting of emissions considering the gridded and explicit emission data may be an issue in such applications (Hood et al., 2018; Stocker et al., 2014). Also, quality control of the modelling results focused on fulfilling of quality objectives should be an inherent part of the analysis.

The main objective of this work is to explore currently available global/regional scale services and to evaluate their applicability to urban scale air pollution assessment and forecast.

To address the defined objective, an integrated approach for urban scale air quality and forecast was implemented based on the ADMS-Roads model and using two alternative approaches to characterize background pollution levels from regional scale modelling. For this purpose, the operational Copernicus Atmosphere Monitoring Service (CAMS) was explored. Additionally, the regional chemical transport model CHIMERE, that makes part of the CAMS service and also of the operational air quality forecasting system for Portugal (http://previsao-qar.web.ua.pt), was explored with different modelling set-up. Thus, different spatial resolutions and the specific parameters appropriated for high-resolution scale were investigated in order to obtain detailed input information for the urban scale modelling. The approach developed in this research was applied to estimate PM_{10} concentrations only.

The overall methodology applied in this study is described in Section 4.2 for both regional and urban scale modelling. Section 4.3 gives the results of both scales and the models evaluation against measured concentrations while Section 4.4 presents the conclusions.

5.2. Methodology

5.2.1. Regional scale modelling

The regional scale modelling approach is based on two forecasting methodologies: the European Copernicus Atmosphere Monitoring Service (CAMS) and the Portuguese Chemical Transport Model (CHIMERE). These approaches enabled to obtain background concentrations, which are a crucial data for the urban scale modelling. The methodology is schematically represented in Figure 5.1.

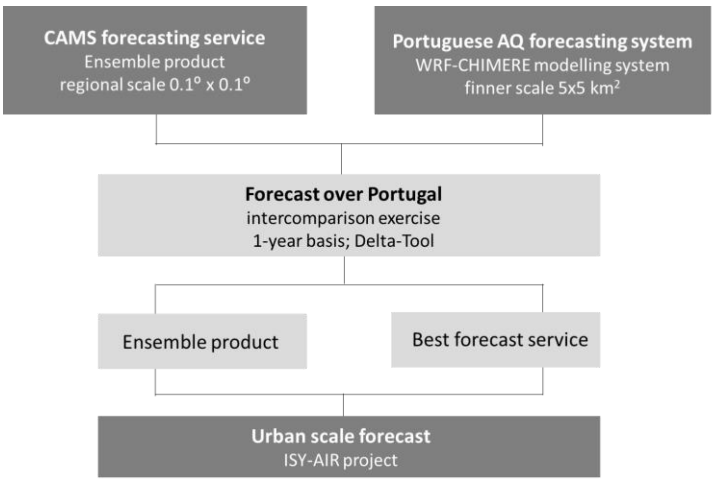


Figure 5.1. Modelling approach applied in the study.

The Copernicus Atmosphere Monitoring Service (CAMS) is one of the six services of the COPERNICUS Programme and provides freely available data on air pollution forecast from a global and regional models (http://atmosphere.copernicus.eu). At European scale, the median ensemble is calculated allowing hourly and daily forecast for four days in advance using outputs from seven atmospheric chemistry models (CHIMERE, EMEP, EURAD-IM, LOTOS-EUROS, MATCH, MOCAGE and SILAM). The ensemble forecast has a spatial grid resolution of 0.1ºx0.1º (high-resolution) and is calculated for different vertical layers, from surface (on the level of monitoring station) to 5000 meters (CAMS, 2016). Meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) are used to drive the chemical transport models. The meteorological data had the same spatial and temporal resolution, which is used by CAMS. To provide background concentrations for urban scale modelling, CAMS PM_{10} data for Continental Portugal were extracted from the European domain for the surface vertical layer and for the first forecast day (a subset of 0h-24h from 0h-96h forecast).

With the purpose of delivering daily air quality forecast for Portugal, a numerical modelling system was developed for Portugal domain (http://previsao-qar.web.ua.pt/), based on the CHIMERE model (Menut et al., 2013) and running daily at the University of Aveiro. Meteorological inputs for these simulations are driven by the Weather Research and Forecasting (WRF) model. Three nested domains covering part of the North Africa and Europe are used, with horizontal resolutions of 125x125 km^2, 25x25 km^2 and 5x5 km^2 for the innermost domain covering Portugal (see Figure 5.2). 24 vertical levels are considered up to the 200 hPa pressure level. At the boundaries of the outermost domain, climatologies from global model simulations are used: outputs from the GOCART model are used for mineral dust, and LMDz-INCA for all gaseous and other aerosol species. Boundary conditions for the nested domains are updated every hour using the previously done coarse CHIMERE simulation. The initial conditions are defined by the 24h forecast from the previous-day model run.

Figure 5.2 Three nested domains with horizontal resolutions of 125x125 km^2 (C125), 25x25 km^2 (IP25) and 5x5 km^2 (PT05) used by CHIMERE model in the Portuguese air quality forecasting system.

The main anthropogenic emissions which are given to the modelling system are derived based on data from the annual UNECE/EMEP emission database (http://www.ceip.at), following a top-down approach for spatial downscaling and standards time profiles for temporal disaggregation (Silveira et al., 2018). Biogenic emissions are computed online as well as sea salt and dust emission fluxes.

CHIMERE outputs are used to characterize PM_{10} background concentrations for urban scale modelling following the same methodology as described for CAMS, e.g., concentrations within surface vertical layer, for the first forecast day.

5.2.2. Urban scale modelling

5.2.3. Integrated modelling approach

An integrated modelling chain has been setup to assess the air quality at the urban scale, including both local and background contributions. The model chain is shown in Figure 5.3, which includes three components: macroscopic transportation model (VISUM); emission model (QTraffic); and air quality model (ADMS-Roads).

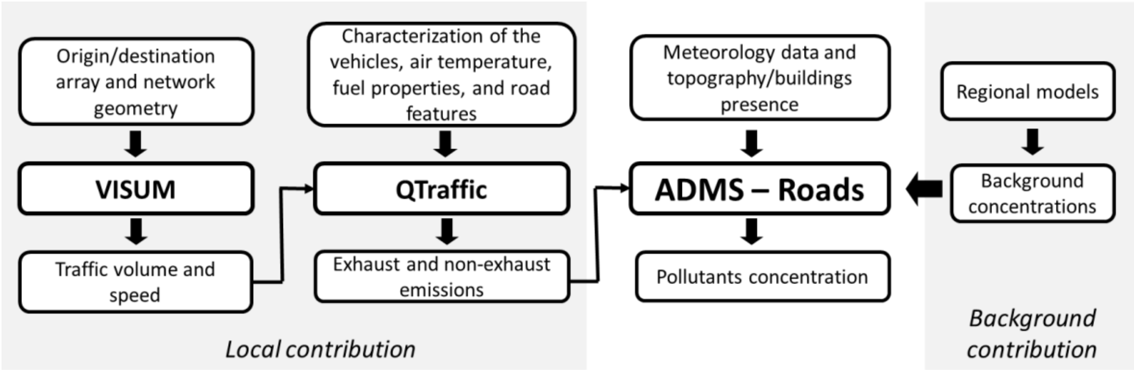


Figure 5.3. Integrated modelling system for urban scale forecast.

VISUM is a macroscopic transportation model based on the four-step approach (PTV 2013), which was chosen given its popularity and simplicity to citywide analysis. VISUM is a traffic modelling software developed by the German company of Transport Planning and Analysis of Operation, that is designed for multimodal analysis, integrating all relevant modes of transportation into one reliable network model. The software enables to simulate traffic demand of different transport modes taking into account a defined road network structure by means of estimated “Origin-Destination” (O-D) matrix. The traffic assignment is carried out considering the congestion effects according to Wardrop's principle (Ortúzar and Willumsen, 2011). Through this model, it is possible to estimate the number of vehicles and average speed for each road segment, which is the key information necessary for the emissions model.

The Traffic Emission and Energy Consumption Model (QTraffic) is a mesoscopic emission model that is based on the updated European guidelines for emission factors, following an average speed approach (EMEP/EEA, 2016). The model treats roads as line sources and being able to estimate traffic-related emissions by considering emission factors determined according to average speed and vehicle class. These estimations require information on: road network of the study area (type, length, and gradient of each road); vehicle fleet composition (emission reduction technology, engine capacity, engine age, and fuel type); and transport activity data for each road (traffic volume and average vehicle speed). The spatial information on transport activity data is requested in GIS (Geographic Information Systems) format, for that a link between VISUM and QTraffic was developed (Dias et al., 2019a). The outputs on emission data obtained at this step are important inputs to the air pollution dispersion model, as shown in Figure 5.3.

The Atmospheric Dispersion Modelling System (ADMS) (Carruthers et al., 1994) is an advanced Gaussian plume air dispersion model based on up-to-date physics using parametrization of the boundary layer structure and the boundary layer height. ADMS-Roads is a local/urban scale dispersion model, which resolves concentrations gradients that occur in the vicinity of line sources (roads), however, it also enables the dispersion estimation of pollutants emitted by point, area and volume sources. The pollutants dispersion is driven by hourly meteorological parameters (including wind direction, wind speed, precipitation, cloudiness and others), that are characterised using the Monin-Obukhov length similarity theory (CERC, 2017). Besides the meteorology, the dispersion of pollutants is highly influenced by the urban canopy. ADMS-Roads has an advanced street canyon module, and a simplified definition of street canyon parameters (height and width) near each road source.

Both local and background contributions are important in the ADMS-Roads pollutants estimations. Although the local contribution considered by the traffic-related emission modelling, the background contribution is obtained from regional modelling based on CHIMERE and CAMS as described before. Furthermore, this model allows to obtain the pollutants concentration from hourly to annual temporal resolution and in three dimensional terms, taking into account the user definition.

5.2.4. Application

The integrated modelling approach has been used to perform simulations for the Coimbra urban area, considering recent data for one year (2018). Coimbra represents one of the largest urban centre in Portugal, situated in the Coimbra´s municipality with a total of 319 km^2 and 133724 inhabitants (PORDATA, 2019). It is inserted in the Coimbra Region (NUTSIII). In this municipality, the sources with the greatest contribution to PM_{10} concentrations are the residential sector, industry and road transport (APA, 2017). In this study we will focus on particulate matter over the urban area (Coimbra).

Figure 5.4 represent the study domain of Coimbra urban area with the two monitoring stations located within the city.

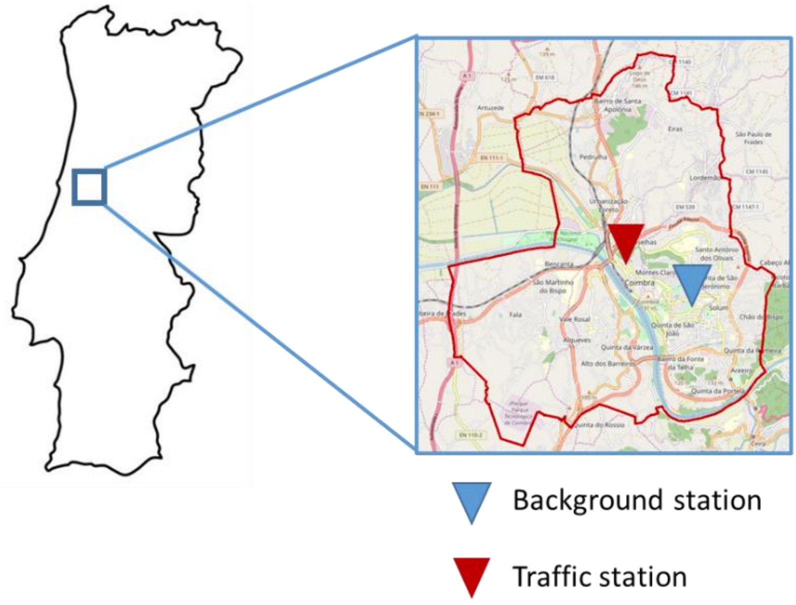


Figure 5.4. Study domain and air quality monitoring stations located within the Coimbra urban area.

For the traffic station, data available from the national database (https://qualar.apambiente.pt/) were analysed from 2010 to 2018. The limits imposed by Air Quality Directive (2008/50/EC) and transposed to the national legislation (DL 102/2010) are taken into account for daily and annual values of PM_{10}. The daily average concentrations should not exceed 50 µg.m^{-3}, which may not be exceeded more than 35 days per year. The value established for annual averages is 40 µg.m^{-3}. Statistical parameters for PM_{10} measurements at urban traffic station are presented in Table 5.1. The PM data observed from 2010 to 2015 shows an improvement, however, in the last 3 years (2016 - 2018) the data available do not allow to draw any conclusions. Moreover, these pollution levels are above World Health Organization (WHO, 2006) guidelines (PM_{10} annual average = 20 µg.m^{-3}) defined for protection of public health. Although the data collection efficiency at traffic station for 2018 was low on the annual basis, a continuous time series from April to December with the collection efficiency of 98% was selected for the analysis.

Table 5.1. Statistical parameters for PM_{10} measurements at "Avenida Fernão de Magalhães" (AFM) station.

PM_{10}	AFM			
	Annual Average Concentration (µg.m^{-3})	**Daily Maximum Concentration (µg.m^{-3})**	**Number of days with exceedances**	**Data collection efficiency**
2010	33.0	84.3	43	99.5%
2011	32.1	82.6	40	98.9%
2012	26.9	66.5	18	99.7%
2013	23.6	85.3	8	99.7%
2014	24.6	61.6	11	99.7%
2015	23.9	89.2	10	83.3%
2016	**	**	**	8.5%
2017	**	**	**	**
2018	25.4	84.9	7	68.8%
** No data available				

In order to apply the methodology described above, several input data were compiled. Traffic demand required for the transportation modelling is characterized based on daily O-D matrix (TIS, 2011) adapted to the current study. Additionally, local traffic counts were considered to define hourly profiles for working days, Saturday and Sunday used for temporal disaggregation of the VISUM outputs. Characterization of vehicles fleet, air temperature, fuel properties and road feature are necessary to apply the emission model (QTraffic). This information was obtained from national statistical data for 2018. The emission model provides exhaust and non-exhaust emissions of PM_{10} that serves as input to ADMS software. To setup the air quality dispersion model, meteorological data, topography and presence of buildings are taken into account. Meteorological data from Geophysical Institute, co-located with the urban background air quality station, are considered to provide input data required by ADMS. The street canyon parameters were acquired based on a simplistic method that is supported by local statistical information, maximum building height and type of roads. Besides the traffic contribution, the background pollution levels were considered from regional operational forecast described before. Finally, particulate matter concentrations for urban area were obtained with spatial resolution of 10 meters in order to characterize the high concentration gradients close to roads.

5.3. Results

5.3.1. Region scale modelling

The regional PM_{10} background concentrations obtained by both forecasting systems (CAMS at European scale and CHIMERE for Portugal domain) were compared using the air quality monitoring sites available for Central Portugal. Figure 5.5 shows the modelled and observed PM_{10} mean concentrations spatial distribution, for the year of 2018. The higher resolution in CHIMERE model simulations than in CAMS ensemble product allows better spatial details, particularly over the Centre and North coast (such as over the industrial areas in Estarreja and Figueira da Foz) as well as in Lisbon urban area. According to Figure 5.5, both model products show good agreement with observations regarding annual mean concentrations (where annual observed values are represented in bullets). However, this assessment is based on observations from four monitoring stations only, which were selected according to their type (background), location (Central Portugal) and efficiency of data collection (>90%) in 2018. Based on these criteria, one urban background station (IGE, data collection efficiency 98%) and three rural background stations (CHA, FUN, LNH with 98%, 93% and 99% of data collection efficiency, respectively) are considered in the analysis.

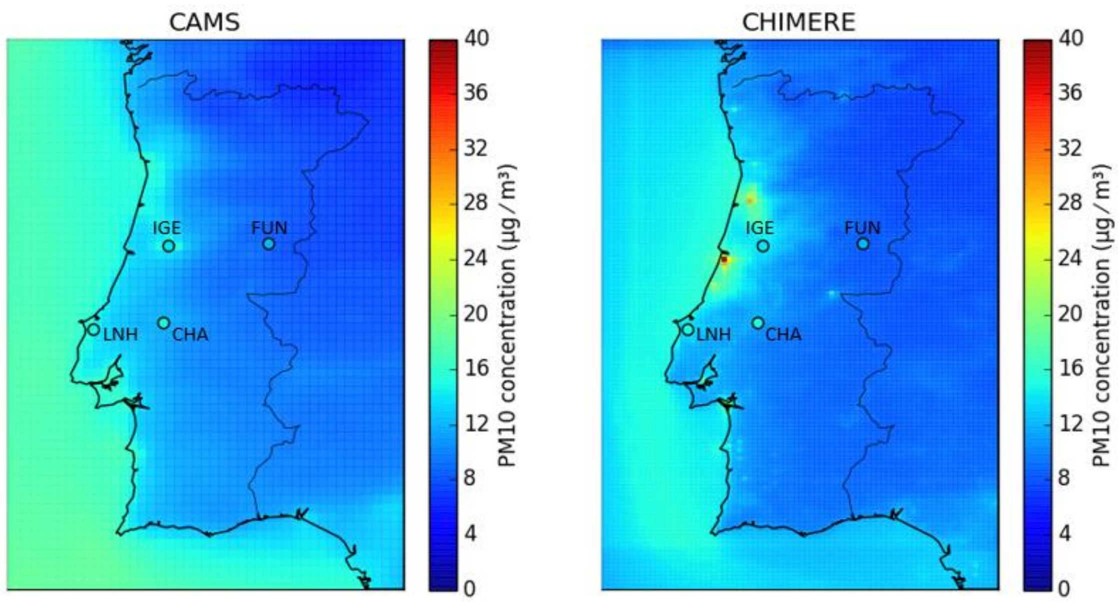


Figure 5.5. Spatial distribution of annual mean PM_{10} concentrations, modelled by CAMS (left) and CHIMERE (right), and observed in background monitoring sites located in Central Portugal (in circles), for 2018.

To assess models' agreement with observations in a daily basis, the FAIRMODE Delta Tool was applied and the assessment target plots are presented in Figure 5.6. This analysis is based on the Model Quality Indicator (MQI) calculation, including the measurement uncertainty. MQI is a statistical indicator which provides the divergence between observations and models, defined as the ratio between the RMSE between measured and modelled values, and a value representative of the maximum allowed uncertainty (βRMSU). The Model Quality Objective (MQO) is a performance criterion, that is verified when MQI is less or equal to one (MQI ≤ 1) (Thunis et al., 2012b). The MQI associated to the 90^{th} percentile worst station is calculated and indicated in the upper left corner (MQI_HD, relative to model performance considering daily mean data). Below this main indicator, also the MQI when using yearly average data is provided (MQI-YR). The green area on the Target plot identifies the area of fulfilment of the MQO, i.e. MQI less than or equal to 1.

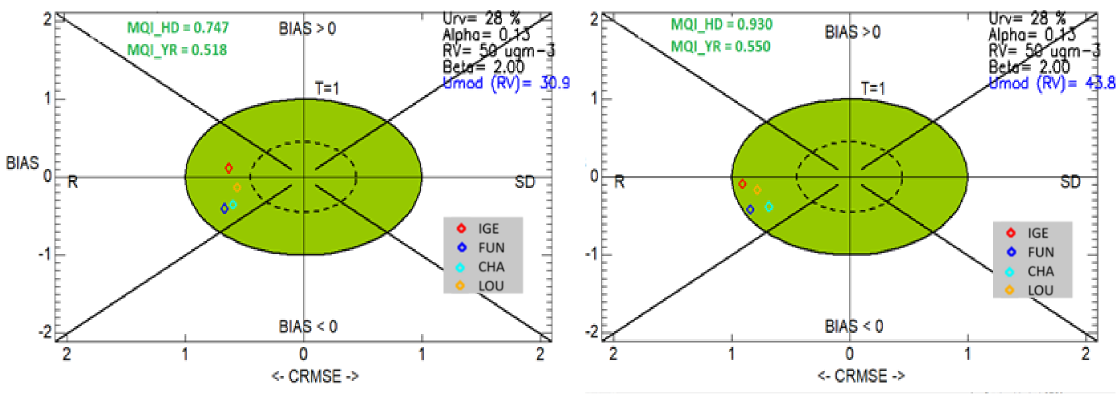


Figure 5.6 Assessment target plot for PM_{10} concentrations modelled by CAMS (left) and CHIMERE (right), for 2018.

The measurement uncertainty parameters (α, β, URV and RV) used to produce the diagram are listed on the top right-hand side. In blue colour, the resulting model uncertainty is calculated according to Thunis et al. (2013) and is provided as output information. β is set equal to 2, allowing thus deviation between modelled and measured concentrations as twice the measurement uncertainty. The BIAS and CRMSE presented in the X and Y axis are normalized by the measurement uncertainty, RMSU. For each point representing one station on the diagram the ordinate is then BIAS/βRMSU, the abscissa is CRMSE/βRMSU and the radius is proportional to RMSE. In this way, target diagrams can show whether errors are dominated by bias (either negative or positive), by correlation or standard deviation, according to the position of the stations in the diagram.

For 2018 and considering the four background monitoring stations, both CAMS ensemble and CHIMERE fulfil quality targets. According to Figure 5.6, modelling errors in forecasting regional PM_{10} concentrations in Central Portugal are dominated by correlation, for both model products, since station points are depicted on the left side of the diagrams. Nevertheless, CAMS ensemble MQI_HD (0.747) is lower than for CHIMERE (0.930) for 2018, which reveals CAMS better performance. This may be related with the assimilation of observations in the model simulations which compose the CAMS ensemble, which is not performed in CHIMERE modelling from the Portuguese operational air quality forecasting system.

5.3.2. Urban scale modelling

The ADMS-Roads results for annual mean PM_{10} concentrations (2018) are presented in Figure 5.7 using CAMS background values as an example. The maximum value of PM_{10} for the urban area (38 $\mu g.m^{-3}$) considering background pollution levels and local contribution is much higher than the concentrations obtained from measurements (25.4 $\mu g.m^{-3}$ and 15.2 $\mu g.m^{-3}$ for urban traffic and background stations, respectively) and close to the annual limit value of 40 $\mu g.m^{-3}$ defined by the legislation. The difference in the values shows that observation data may not represent the hotspots within the urban area thus highlighting the importance of the urban AQ modelling. The local highest contribution from road traffic to PM_{10} annual mean concentrations obtained from the modelling within the urban domain achieves 22 $\mu g.m^{-3}$. The ADMS-Roads results using CHIMERE background concentrations present a similar spatial pattern but with lower grid cell values.

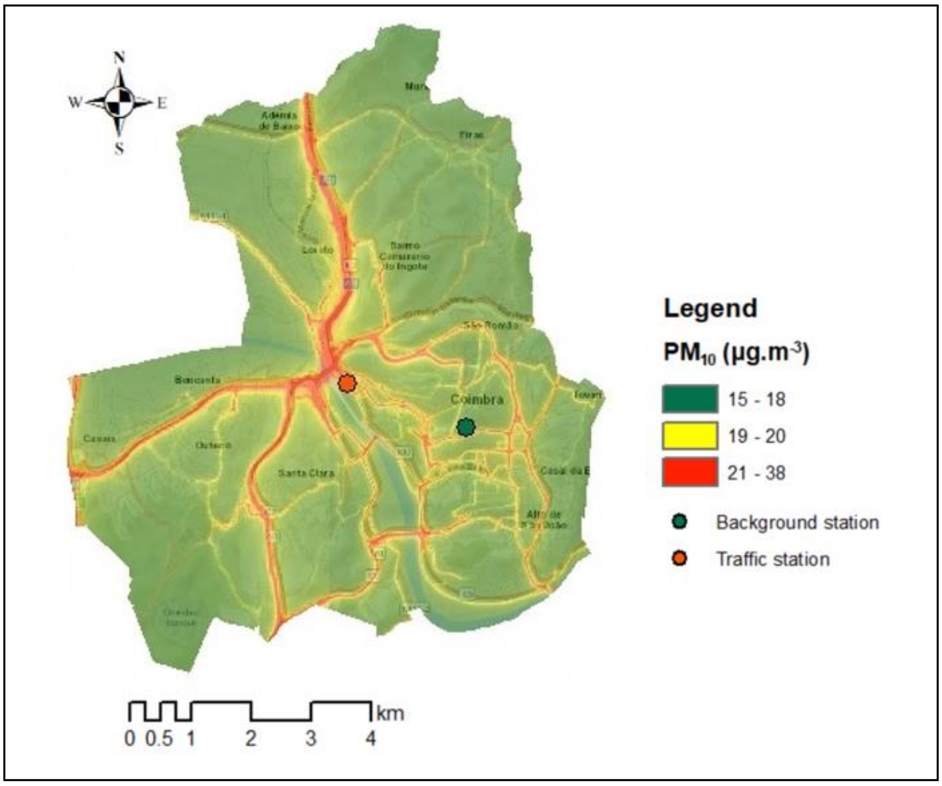


Figure 5.7 Spatial distribution of annual mean PM$_{10}$ concentrations, modelled by ADMS with CAMS background values, and observed values from monitoring sites located in Coimbra city (in circles), for 2018.

Figure 5.8 shows time series of the daily concentrations from the ADMS model (local contribution) and the measurements from the traffic station AFM located within Coimbra urban area. It should be noted that the local contribution is represented by road traffic only and this is one of the possible reasons why the results of the model are lower than observations. Moreover, only primary component of PM$_{10}$ is simulated by the Gaussian model.

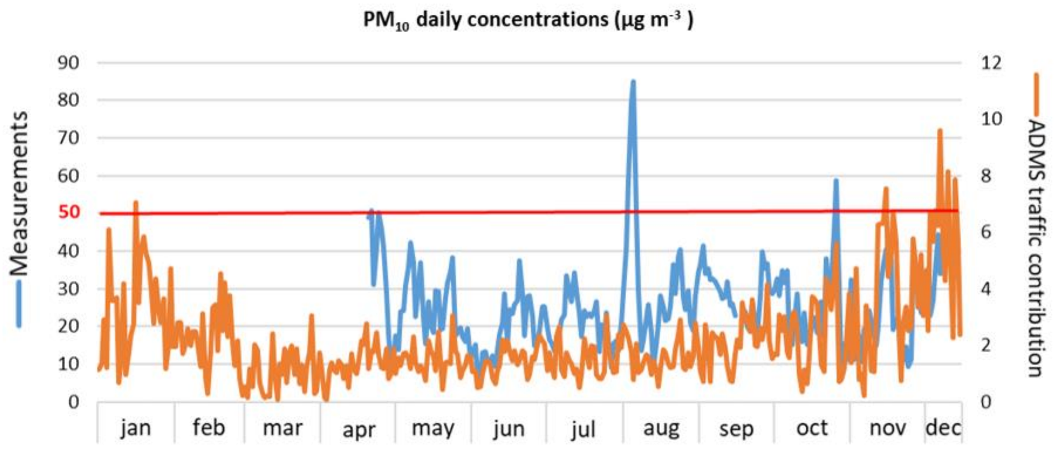


Figure 5.8 Traffic local contribution (no background) obtained from ADMS and AQ measurements at AFM urban traffic station for 2018.

During 2018, in Portugal occurred seven episodes resulting in PM_{10} exceedances at traffic station. For these days, ADMS modelling results with different background levels provided by CHIMERE and CAMS are compared with the observations (Figure 5.9). Generally, the ADMS+CAMS has a better performance when compared to the observations. However, in the presence of natural events related with sand and dust storms in North Africa as observed from 3rd to 6th August (APA, 2018) the ADMS+CHIMERE shows better performance. Also, the results highlight the necessity to improve characterization of local pollution sources, including non-traffic emissions, in order to guarantee the completeness of the emission data.

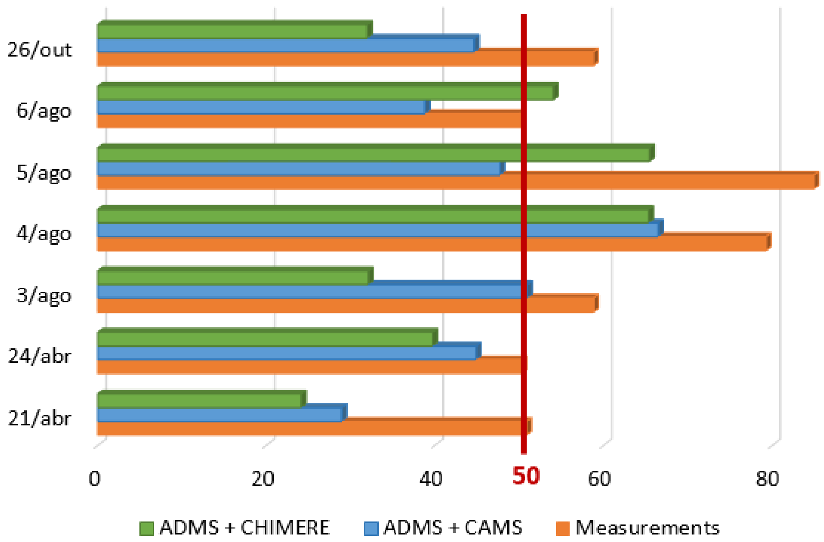
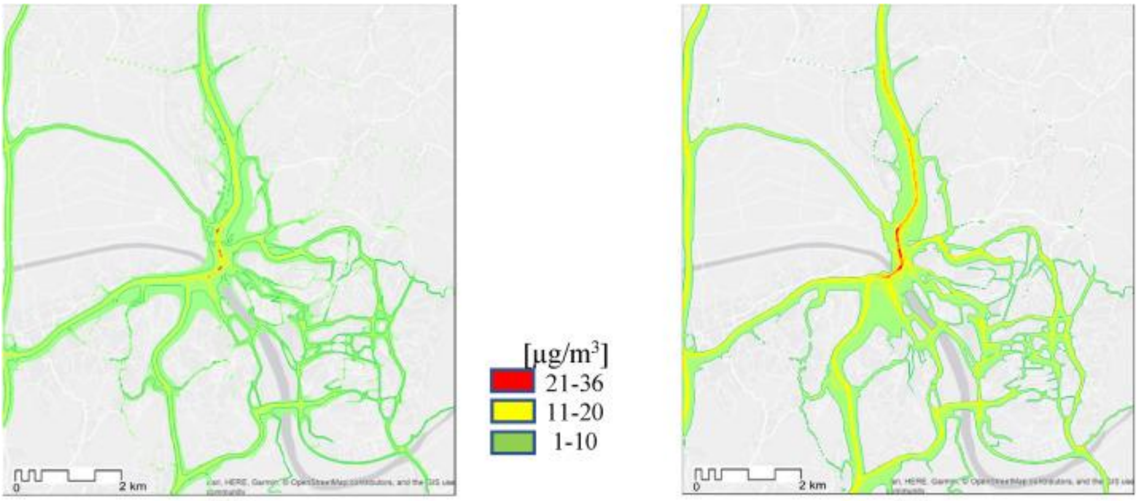


Figure 5.9. PM_{10} exceedances observed at traffic station during 2018.

Spatial distribution of concentrations and the contribution of road traffic to the pollution levels within the study area are also analysed for the days with PM_{10} exceedances. Two examples are presented in Figure 5.10 for 3rd of August and 26th of October. Although the absolute contribution of road traffic to PM_{10} is varying for other days with the exceedances, the spatial pattern of the concentrations is similar (not presented). As expected, higher contribution of road traffic occurs in the vicinity to the main roads resulting in daily average concentration of 36 $\mu g.m^{-3}$ for some grid cells. These data should be analysed taking into account that the limit value for daily average PM_{10} concentrations defined by the legislation is 50 $\mu g.m^{-3}$ and the results presented in Figure 5.10 are focused on local traffic only without taking into account the contribution from other sources.



(a) (b)

Figure 5.10 Traffic local contribution to PM_{10} daily average concentrations on 3rd of August (a) and 26th of October (b) Coimbra urban area estimated by ADMS.

To analyse the model performance, observations and model predictions are presented also as a scatterplot (Figure 5.11) for the period of April-December 2018 (no measurements at AFM are available before April). Both approaches, ADMS+CAMS and ADMS+CHIMERE, underestimate PM_{10} daily average concentrations with a negative mean normalized bias (MNB, -0.2 for ADMS+CAMS and -0.3 for ADMS+CHIMERE) as presented in Table 5.2. The difference between annual average concentrations observed and modelled is about 24%.

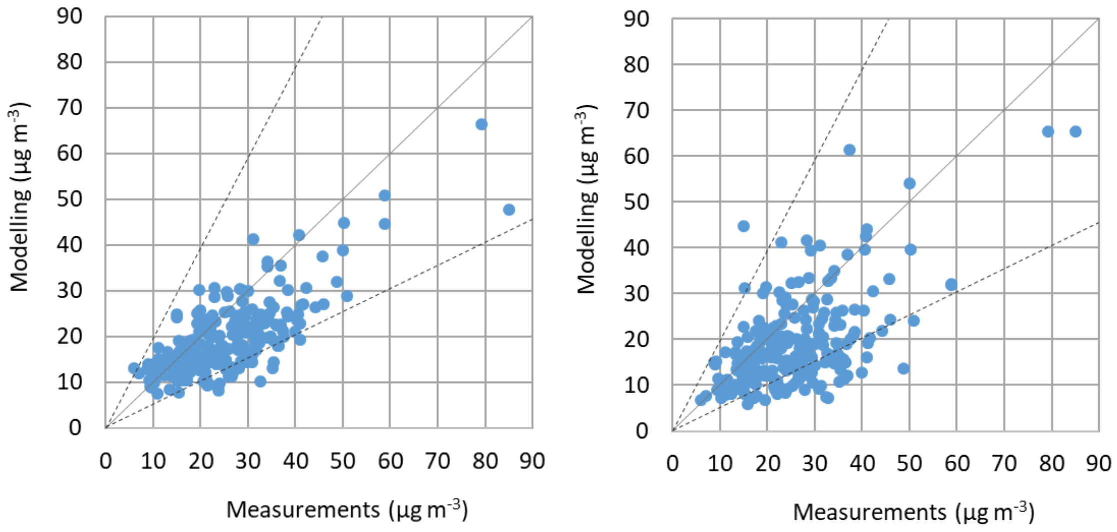


Figure 5.11 ADMS-Roads results for PM_{10} daily average concentrations versus AQ measurements at urban traffic station for April to December 2018 using background concentrations from: a) CAMS and b) CHIMERE.

Table 5.2. Statistical parameters for PM_{10} concentrations: urban scale modelling results and the observations at traffic station (April to December 2018).

Statistical parameters	Measurements	ADMS + CAMS	ADMS + CHIMERE
Average ($\mu g.m^{-3}$)	25.4	19.4	19.0
Max ($\mu g.m^{-3}$)	85.0	66.5	65.4
Min ($\mu g.m^{-3}$)	6.0	7.6	5.8
Correlation coefficient (r)	-	0.8	0.6
BIAS ($\mu g.m^{-3}$)	-	-6.0	-6.4
Mean Normalized BIAS (MNB)	-	-0.2	-0.3
Root Mean Square Deviation (RMSE) ($\mu g.m^{-3}$)	-	9.4	11.5
Fraction of predictions within a factor of two of observations (FAC2)	-	0.9	0.8
Max Error ($\mu g.m^{-3}$)	-	37.3	35.2

The statistical parameters presented in Table 5.2 show that results from ADMS+CAMS are overall better than those from ADMS+CHIMERE, i.e., ADMS+CAMS approach has higher r and FAC2, and lower RMSE. Although the annual statistics reveal lower concentrations obtained with CHIMERE (lower annual average, higher bias), the peaks are better reproduced using the national forecast (maximum error for daily average concentrations is 35.2 $\mu g.m^{-3}$ against 37.3 $\mu g.m^{-3}$ obtained with CAMS background levels) and this is in agreement with the data presented in Figure 5.9. For both approaches, ADMS results for Coimbra satisfy the modelling performance criteria

(MQ_HD=0.536 and MQI_HD=0.648, using CAMS and CHIMERE data, respectively). It is interesting to highlight that MQI obtained for the traffic station with ADMS is better than MQI for the regional data considered as inputs to the Gaussian model.

5.4. Conclusions

In this paper, two existing services at a regional scale were explored and their applicability to the assessment and forecast of PM_{10} concentrations at the urban scale were analysed. Both approaches (CAMS and national forecast based on CHIMERE) show a good statistical performance and enable characterization of the background concentrations for urban scale forecast. In general, both regional models underestimate PM_{10} concentrations for the study area. For the long-term statistics CAMS reveal better performance than national forecast, while episodes related with natural events are better reproduced by CHIMERE.

Urban scale modelling performed with ADMS reveal good correlation (r = 0.8) for daily mean PM_{10} concentrations, but annual mean at traffic station is underestimated about 24%. It should be stressed that contribution of local pollution sources to PM levels is calculated taking into account primary aerosol particles only. Also, negative bias in the background levels provided by the regional modelling contribute to the overall underestimation of PM_{10} at traffic station.

The maximum value of annual mean concentration obtained for PM_{10} from the urban modelling is about 38 $\mu g.m^{-3}$ with the traffic contribution up to 22 $\mu g.m^{-3}$ in the vicinity to the main roads. The spatial pattern of pollution levels obtained by high resolution modelling reveal that only one traffic station, with annual mean PM_{10} concentration of 25.4 $\mu g.m^{-3}$, may not be representative for hotspots in the city. A limited number of the measurement points is also a constrain for the model validation. Thus, the performance of urban scale modelling at only one available traffic station (MQI_HD = 0.536 and MQI_HD = 0.648, using CAMS and CHIMERE background levels, respectively) is better than the regional modelling at background stations (MQI_HD = 0.747 and MQI_HD = 0.930, for CAMS and CHIMERE, respectively). Further investigation is required to analyse comparability of the Quality Indicators obtained from different modelling scales.

Chapter 6

6. Contribution of road traffic to black carbon aerosols at city level – validation of the modelling approach

This chapter is based on the submitted paper: "Pina, N., Almeida, S. M., Alves, C., Tchepel, O. Contribution of road traffic to black carbon aerosols at city level – validation of the modelling approach. Atmospheric Environment".

6.1. Introduction

Urban health and liveable cities require clean air environments. Nonetheless, despite major improvements in European air quality (AQ) due to key policies and technological developments, exceedances of AQ standards are still common in most European cities. Thus, the proportion of urban population exposed to the pollution above the EU limits values is 11% for particulate matter (PM_{10}), 1% for nitrogen dioxide (NO_2), 12% for ozone (O_3) (EEA, 2022c) and these numbers are much high if the new World Health Organisation (WHO) recommendations are considered instead (71%, 85% and 95% respectively). However, what about harmful pollutants, such as Black Carbon (BC), that are still not regulated?

BC, a component of PM, is a product of incomplete combustion and it is not only a key pollutant with adverse effects on human health (Grahame et al., 2014; Highwood and Kinnersley, 2014; Janssen et al., 2011, 2012), but also a short-lived climate forcer that contributes significantly to climate change (Szopa et al., 2021). However, BC is still not regulated in Europe and measurement data are limited. The lack of legislation requirements may help to explain the very small number of urban observational sites (Kunder et al., 2018). The data available are mainly obtained from demanding measurement campaigns at urban sites (Becerril-Valle et al., 2017; Costabile et al., 2017; Helin et al., 2018; Hristova et al., 2022; Küpper et al., 2018; Liakakou et al., 2020; Luoma et al., 2021; Titos et al., 2017; Wyche et al., 2020). In Portugal, few studies have been carried out to gain better understanding of urban BC (see Table 6.1), helping to comprehend the magnitude and temporal variations. Such studies have pointed out that

BC in urban areas is mainly emitted by traffic and is often used as a traffic emission tracer (Hudda et al., 2020; Luoma et al., 2021; Reche et al., 2011). Additionally, biomass burning is also a very important source that contributes to BC urban levels, especially in residential areas during cold months (Luoma et al., 2021). Although the evidence base is insufficient to set an Air Quality Guideline level to provide a basis for legally binding limit values for BC, due to health concerns related to this pollutant, the WHO recommends more actions to enhance further research on its risks and approaches for mitigation (WHO, 2021).

To address the gap on limited BC observations and improve the knowledge on its spatial and temporal variations, different modelling approaches may be used. Based on literature review, statistical modelling, including land-use regression, is mostly applied to estimate and map BC concentrations (Awad et al., 2017; Boniardi et al., 2019; Jones et al., 2020; Kerckhoffs et al., 2016; Liu et al., 2019; Richmond-Bryant et al., 2009; Sanchez et al., 2018; Van den Bossche et al., 2018; Van den Hove et al., 2020). Other approaches are based on machine-learning techniques (Fung et al., 2021; Rovira et al., 2022). However, when no sufficient monitoring data are available, the applicability of statistical modelling approaches is limited.

Atmospheric dispersion modelling is one of the approaches allowing spatial and temporal analysis of BC concentrations. The spatial distribution of BC may be obtained with a high spatial resolution and at a street level (Brasseur et al., 2015; Lugon et al., 2021; Zhang et al., 2019), which is crucial to identify urban hotspots and to implement mitigation strategies. Table 6.2 presents a list of studies based on dispersion modelling and considering different research questions in order to understand the dispersion of BC near a road, to test the ability of the model to predict BC in a specific point, and to understand the BC spatial distribution over a city domain. However, application of the dispersion modelling requires detailed information on local emission sources. Taking into account that road traffic is considered a major contributor to BC pollution in urban areas, quantification of traffic-related emissions is a crucial step.

Table 6.1. An overview of BC (or elemental carbon, EC) concentration measurements in Portugal.

Reference	Location	Period	Approach	BC concentrations
Gamelas et al. (2023)	Lisbon (Portugal)	12/2019 – 11/2020	• A multi-wavelength absorption instrument used to measure BC in $PM_{2.5}$ PTFE filters.	• Mean ($\mu g.m^{-3}$): 2.5 • $BC/PM_{2.5}$ ratio: 0.199 (on a site with influence of high-density traffic motorway and medium-sized industries)
Almeida et al. (2022)	Lisbon (Portugal)	9/2017-10/2018	• Medium-volume samplers (MVS6 Leckel) for PM sampling • Quartz filters were analysed for OC/EC measurements by a Thermal Optical technique	• Outdoor EC concentration ($\mu g.m^{-3}$): 1.4 • Classrooms EC concentration ($\mu g.m^{-3}$): 1.7 • Homes EC concentration ($\mu g.m^{-3}$): 1.1
Martins et al. (2021)	Lisbon (Portugal)	6 – 10/2018 (21 weekdays)	• PM and BC measurements carried out in cars, metro, bus and bicycle • BC concentrations were measured with a portable microaethalometer	• Higher BC mean concentrations registered when travelling by car (inside): 6.1 $\mu g.m^{-3}$ (range 2.6 - 8.4 $\mu g.m^{-3}$) • Outdoor BC mean concentrations when travelling by car: 7.7 $\mu g.m^{-3}$ (range 3.5 - 12.2 $\mu g.m^{-3}$)
Blanco-Alegre et al. (2020)	Braga (Portugal)	1 – 8/2/2013	• Measurements with Aethalometer in a road tunnel	• Mean ($\mu g.m^{-3}$): 21 • Hourly range ($\mu g.m^{-3}$): 0.14 - 49
Malico et al. (2017)	Évora (Portugal)	4/2007 – 2/2015	• Measurements with Multi-Angle Absorption Photometer	• Annual mean ($\mu g.m^{-3}$): 1.3 Range ($\mu g.m^{-3}$): 0.2 - 5.4 (P5 to P95 of hourly values)
Pereira et al. (2012)	Évora (Portugal)	4/2007 – 12/2009	• Measurements with Multi-Angle Absorption Photometer	• Mean ($\mu g.m^{-3}$): 1.8 (summer); 0.9 (winter) Range ($\mu g.m^{-3}$): 0.3 - 5
Alves et al. (2002)	Lisbon and Aveiro (Portugal) Hyytiälä (Finland)	7/1999 (Aveiro) 27 – 30/6/2000 (Lisbon and Hyytiälä)	• High-volume samplers for PM and a Sierra cascade impactor • Quartz filters were analysed for OC and EC measurements	• Mean ($\mu g.m^{-3}$): 0.7 (Aveiro); 1.9 and 2.3 (Lisbon with 2 different instruments); 0.19 (Hyytiälä))
Harrison et al. (1997)	Coimbra (Portugal) Birmingham (United Kingdom) Lahore (Pakistan)	Every fifth day over 10/1992 – 10/1993 (Coimbra)	• High-volume samplers for PM • Quartz filters were analysed by means of thermal/optical techniques to obtain OC and EC concentrations	• Annual mean ($\mu g.m^{-3}$): 3.46 (Coimbra), measured at rooftop (15 m altitude) in the city centre

Table 6.1. (Continued).

Reference	Location	Period	Approach	BC concentrations
Castro et al. (1999)	Urban (Oporto, Coimbra and Aveiro, Portugal; Birmingham, United Kingdom) Rural European locations (Tábua, Anadia and Areão, Portugal; Mace Head, Ireland)	1993 – 1996	• High and low volume samplers for PM • EC and organic carbon (OC) concentrations were determined with a home-made thermo-optical system	• Daily range at Coimbra location ($\mu g.m^{-3}$): ~0.2 - 5 (summer); ~0.5 - 7.8 (winter)

Table 6.2. Selected studies based on BC dispersion modelling (worldwide).

Reference	Location	Period	Approach	BC traffic emissions	BC concentrations
Weger et al. (2022)	Leipzig, Germany	1 – 3/3/2020	• Application of urban dispersion CARDIO model with 40 m grid resolution (downscaled to 10 m resolution) • Validation of the model with observations from 3 monitoring sites for BC • Mesoscale simulations (COSMO-MUSCAT with 14 km to 550m resolution)	• Maximum hourly: 2.4 $g.s^{-1}$ (from TNO-MACC2 over the study domain)	• Hourly observations in roadside site ($\mu g.m^{-3}$): 0.1-2.3 • Fractional bias (FB) with 40m resolution: -0.25 and 0.24 on roadside sites • FB with 550 m resolution: 0.7 and 0.9 on roadside sites
Lugon et al. (2021)	Paris, France	12/4 – 15/5/2014	• Application of a multiscale Street-in-Grid model (SinG) • Inclusion of BC non-exhaust emissions • Comparison with BC observations from the air monitoring station operated by AIRPARIF	• Average BC emissions at the street level: 0.00 - 0.14 $g.s^{-1}$	• Mean at the street level ($\mu g.m^{-3}$): 2.3 - 30.4
Gidhagen et al. (2021)	Curitiba, Brazil	25/6 – 26/8/2016	• Development of emission inventory including industry and road traffic sources (only exhaust emissions) • Dispersion models: regional (BRAMS 5.2 system, 10 km x 10 km resolution); urban (Airvivo system, 200 m x 200 m resolution); street canyon (OSPM at the monitoring site) • Validation with measurements from different methods: EC/OC determined by PM filters analysis and BC measured by aethalometer and microathaelometer.	• Emission factors for public transport: 20 - 288 $mg.km^{-1}.veh^{-1}$	• Observations at street level ($\mu g.m^{-3}$): 5.5 (mean) and 10 (maximum hourly) • Mean street increment ($\mu g.m^{-3}$): 3.2 observed, 1 modelled (improved to 3 after regression analysis)

Table 6.2. (Continued).

Reference	Location	Period	Approach	BC traffic emissions	BC concentrations
Patterson and Harley (2019)	San Francisco, USA	3/2014 – 2/2015	• Application of R-LINE model to assess pollution near-roadway sites • Compilation of vehicle exhaust emissions • Characterization of urban background concentrations with observations	• Daily mean (over different week days): 0.7 - 2.7 kg	• Daily mean (μg.m^{-3}): ~1 • Daily maximum (μg.m^{-3}): ~5 • MBIAS (-0.18 and -0.05 in different sites)
Yang et al. (2019)	Beijing, China	2019	• Development of EMBEV-Link emission inventory, including BC • Combination of emission model with RapidAir dispersion model, however used for NOx predictions only	• Total daily: 5.5 tons (~0.34 kg.km^{-2}, study domain) • Hourly mean: 0 - 0.12 kg.km^{-1} (at the link level, during morning rush hours and midnight)	-
Zhang et al. (2019)	Beijing, China	2009	• AERMOD dispersion model • BC measurements with Aethalometers on a roadside and an urban background sites	• Hourly mean: 0.03 - 0.12 kg.km^{-1}	• Mean (μg.m^{-3}): 2.6 (summer); 3.9 (winter) • Maximum (μg.m^{-3}): ~60
Saha et al. (2018)	North Carolina, USA	1/6 – 2/7/2015 (summer) 28/1 – 20/2/2016 (winter)	• Measurements with fixed stations and a mobile platform • Determination of emission factors • Application of R-LINE dispersion model to assess near road dispersion of pollutants	• Emission factors (kg.fuel^{-1}): 0.035 (winter); 0.045 (summer)	• Mean observations near a motorway (μg.m^{-3}): 1.4 (summer); 0.8 (winter)
Brasseur et al. (2015)	Crown and Belliard Street, Brussels	1/7/2011 – 30/6/2013	• Adaptation and implementation of the Danish Operational Street Pollution Model (OSPM) to predict BC in street canyons (model was denominated $CANS_{BC}$) • Model validations based on BC data from fixed measurement network	• BC traffic emissions corrected with a factor of 2.79	• Hourly range at a traffic location (μg.m^{-3}): 0.5 – 7 • Direct emissions contribute to 36% of BC simulated

The methodologies to quantify BC emissions from road traffic depend on the scale of the study and the level of details required. At local scale, information on vehicle activities, in combination with emission factors, is usually applied and focused on hot exhaust emissions (Brausser et al., 2015; Lugon et al., 2021; Paterson and Harley, 2019). However, some studies indicate that BC inventories based only on hot emissions will result in underestimation of the pollution levels and a correction factor for the emissions is required to explain the observations. For instance, Brasseur et al. (2015) indicate that missing cold start emissions could be one of the reasons for the discrepancy between BC modelling and observation values that could be resolved by application of a correction factor. Moreover, the importance of non-exhaust processes, such as brake wear, tyre wear and road abrasion, has been recently pointed out (Lyu and Olofsson, 2020, Lugon et al., 2021). Thus, in order to avoid emission underestimations, it is important to guarantee the completeness of the BC emission inventory. Additionally, the variability of measurement techniques and procedures to determine the emission factors also contribute to high variability and uncertainties in the BC estimates (Kholod et al., 2016; Rönkkö et al., 2023). Therefore, validation of modelling approaches to study traffic related air pollution is one of the priorities, requiring additional research in this field.

The objective of the present work is twofold: (i) to evaluate the contribution of road traffic to BC pollution levels at urban scale with high spatial and temporal resolution and (ii) to validate the modelling approach using BC observations collected with high temporal resolution to be compatible with the modelling objectives. For this purpose, an integrated modelling approach for quantification of BC traffic-related emissions and its contribution to urban aerosol was implemented for Coimbra city, Portugal. The Traffic Emission and Energy Consumption Model (QTraffic) was extended in order to include BC emission factors and considering road network as line emission sources. These emission data are essential inputs to the urban dispersion simulations configured within the Atmospheric Dispersion Modelling System (ADMS-Roads). To validate the modelling approach, BC observations collected with high temporal resolution during a six-month measurement campaign at an urban traffic station were used.

6.2. Methods and materials

The methodology applied in this study for quantification of BC pollution levels at an urban scale is presented in this topic with a focus on new developments in emission modelling. Additionally, the Coimbra case study is presented, including a detailed description of the modelling inputs and BC measurements used for validation.

6.2.1. Integrated modelling Approach

An integrated modelling approach previously applied to study air pollution at urban scale (Dias et al., 2019a, 2019b; Tchepel et al., 2020) has been extended in this work to assess BC traffic-related concentrations. For this purpose, new developments were required to incorporate BC emissions from road traffic. Overall, the integrated modelling framework consists in the cascade of several models that share an information flow starting from transportation modelling and following by emission and dispersion modelling:

(i) The macroscopic transportation model (VISUM) allows simulating the traffic demand of different transport modes taking into account a defined road network structure by estimating the "Origin-Destination" (O-D) matrix. Through this model, it is possible to estimate the number of vehicles and average speed for each road segment, which is the key information necessary for the emission model.

(ii) The Traffic Emission and Energy Consumption Model (QTraffic) (Dias et al., 2019a) treats roads as line sources and estimates traffic-related emissions by considering EMEP emission factors (EMEP/EEA, 2019a, 2019b) determined according to average speed and vehicle class. These estimations require information on: road network of the study area (type, length, and gradient of each road), vehicle fleet composition (emission reduction technology, engine capacity, engine age, and fuel type), and transport activity data for each road (traffic volume and average vehicle speed) and different vehicle classes (passenger cars, light-duty vehicles, heavy-duty vehicles and buses)

(iii) The Atmospheric Dispersion Modelling System (ADMS-Roads) is an advanced Gaussian plume air dispersion model based on up-to-date physics that uses parametrisation of the boundary layer structure and the boundary layer height to study air pollution caused by road networks. The Monin-Obukhov length similarity theory is used to characterise the hourly meteorological characteristics, such as wind direction, wind speed, precipitation, cloudiness, and others, that control the dispersion of pollutants (CERC, 2017). The urban canopy has a significant impact on the dispersion of pollutants in addition to the weather. A simpler definition of the street canyon characteristics (height and width) near each road source is provided by the advanced street canyon module of ADMS-Roads.

6.2.2. Advances in Traffic Emission and Energy Consumption Model (QTraffic)

The QTraffic emission model (Dias et al., 2019a) was extended in this work in order to include BC traffic emissions, considering both exhaust (hot and cold start) and non-exhaust contributions, based on emission factors and traffic activity data.

6.2.2.1. BC hot emissions

Hourly emissions resulting from fuel combustion under hot engine operation (hot emissions, E_{BChot}, g.h^{-1}) are estimated for each road segment by the QTraffic model based on Equation 6.1 and considering BC as a fraction (*f*) of $PM_{2.5}$ that depends on the vehicle technology:

$$E_{BChot} = L_{road} \cdot \sum_{k} f_k \cdot e_{hot_{PM_{2.5},k}} \cdot N_k \tag{6.1}$$

where $e_{hot_{PM_{2.5},k}}$ is the $PM_{2.5}$ hot emission factor [g.km^{-1}.veh^{-1}] for vehicle technology k, N is the number of vehicles passing on the road segment per hour, and L_{road} is the road segment length [km]. The BC fraction (Table 6.3) and $PM_{2.5}$ emission factors are obtained from EMEP/EEA (2019a). The $PM_{2.5}$ emission factors are defined as a function of vehicle speed, fuel type, engine capacity, and emission reduction technology.

Table 6.3 BC fraction of $PM_{2.5}$ applied to exhaust emissions (EMEP/EEA, 2019a).

Class	Technology	Pre-Euro	Euro 1	Euro 2	Euro 3	Euro 4	Euro 5-6
PC and LDV	Gasoline	0.20	0.25	0.25	0.15	0.15	0.15
	Diesel	0.55	0.70	0.80	0.85	0.87	0.20
HDV	Diesel	0.65	0.65	0.70	0.75	0.15	0.65
L-category (Motorcycles)	2-stroke	0.10	0.20	0.20	-	-	-
	4-stroke	0.15	0.25	0.25	0.25	0.25	-

PC – Passenger cars; LDV – Light Duty Vehicles; HDV – Heavy Duty Vehicles

6.2.2.2. BC cold start emissions

Cold start emissions refer to the additional emissions from the vehicles under cold engine in comparison with hot engine operation. Cold start BC emissions (E_{BCcold}) for each road segment [g.h^{-1}] are estimated by considering the cold distance approach previously implemented in QTraffic (Pina and Tchepel, 2023). This approach enables to determine the number of passenger cars circulating on each road segment with cold engine using information on O-D of trips. Cold start BC emissions for each road segment are calculated based on Equation 6.2, by considering $PM_{2.5}$ cold start emission factors from EMEP/EEA (2019a) and the BC fractions (f) presented in Table 6.3.

$$E_{BCcold} = L_{road} \cdot \sum_{k} f_k \cdot \ e_{hot_{PM_{2.5},k}} \cdot (\frac{e_{cold}}{e_{hot\ PM2.5,k}} - 1) \cdot N_{k,cold} \tag{6.2}$$

where f_k is the BC fraction of $PM_{2.5}$ for technology k, $N_{k,cold}$ is the number of vehicles with cold engine passing per hour on the road segment, $e_{hot_{PM_{2.5,k}}}$ is the $PM_{2.5}$ hot emission factor [g.km^{-1}.veh^{-1}], L_{road} is the road segment length [km], and $\frac{e_{cold}}{e_{hot\ PM2.5,k}}$ is the cold/hot emission quotient for $PM_{2.5}$ that depends on vehicle technology k and ambient temperature, being available for diesel vehicles only (EMEP/EEA, 2019a).

6.2.2.3. BC non-exhaust emissions

Non-exhaust emissions do not originate from combustion processes, but rather from wear and corrosion of vehicle components (e.g., tyre and brakes) and road pavement abrasion. Non-exhaust traffic-related emissions (E_{BCnon}) for each road segment [g.h^{-1}] are estimated by taking into consideration different non-exhaust processes (tyre wear, brake wear and road abrasion) as implemented in QTraffic (Dias et al., 2019b). In the current work, the model was extended to incorporate BC emissions using the following equation:

$$E_{BCnon} = L_{road} \cdot \sum_{p} \sum_{k} f_p \cdot \ e_{non_{PM_{2.5},k,p}} \cdot N_k \tag{6.3}$$

where f_p represents the BC fractions of $PM_{2.5}$ for non-exhaust emissions due to abrasion processes p (see Table 6.4), $e_{non_{PM_{2.5},k,p}}$ is the $PM_{2.5}$ emission factor [g.km^{-1}.veh^{-1}] for each abrasion process and vehicle technology k, N is the number of vehicles passing on the road segment per hour, and L_{road} is the road segment length [km]. It is necessary to take into consideration that $PM_{2.5}$ emission factors are determined as a function of vehicle technology and vehicle speed. For HDV vehicles, the number of axels and load of the vehicle are also considered in the emission factors.

Table 6.4. BC fraction of $PM_{2.5}$ for non-exhaust emissions (EMEP/EEA, 2019b).

	Tyre wear	**Brake wear**	**Road abrasion**
BC/PM$_{2.5}$	0.364	0.067	0.039

6.2.3. Case study

The city of Coimbra (Figure 6.1), selected for this study, is one of the main urban centres in Portugal, after the metropolitan areas of Lisbon and Oporto. Coimbra, with a population around 141 000 inhabitants (PORDATA, 2021), is located in the central region of the country, being 40 km from the coast and crossed by the Mondego River. In the last decades, daily commuting trips reached 250 000 movements per day in the intermunicipal area of Coimbra, 70% of which are carried out by private vehicles (CIRMC, 2016). The local contribution to air pollution resides mostly on road transport, commerce activities and residential sector.

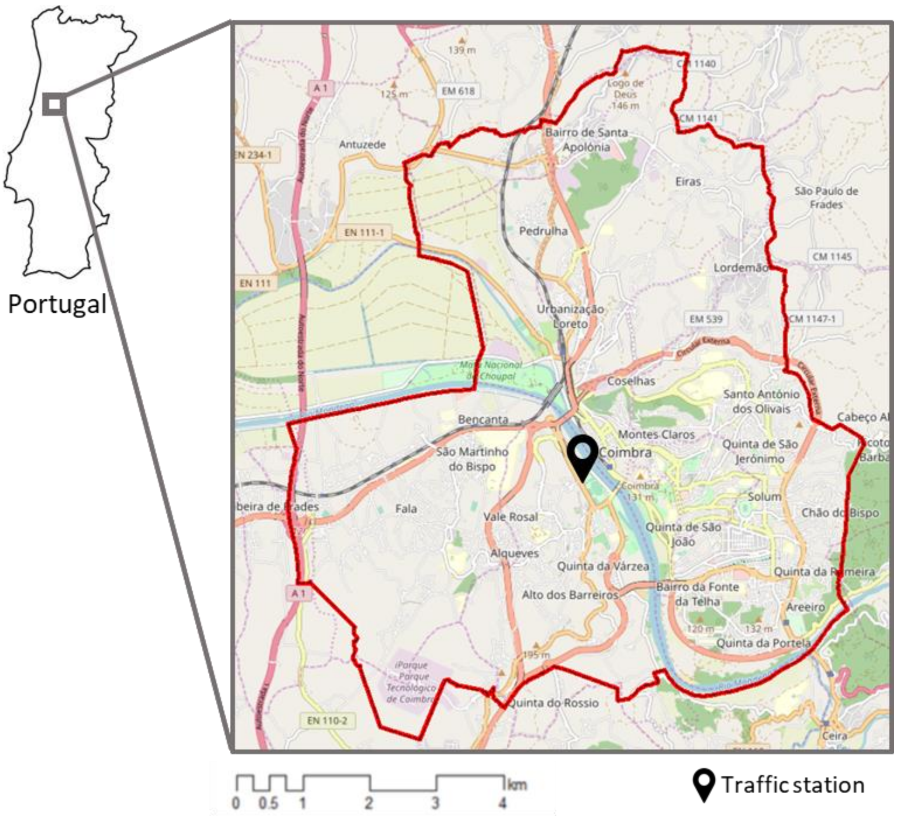


Figure 6.1 Study domain and roadside station within the Coimbra urban area.

6.2.3.1. Application of the Integrated Modelling Approach

To apply the integrated modelling approach for the study area, several input data were compiled. To characterise road traffic, information on number of vehicles and average speed, required by QTraffic for each road segment, were obtained from the VISUM model applied to Coimbra. Information from previous studies (TIS, 2011) was adapted to the current work and calibrated using local data to characterise the supply and demand for transport in the study area. Traffic counts (Figure 6.2) collected during the measurement campaign were considered to define traffic hourly profiles for working days, Saturdays and Sundays. Additionally, information on vehicle technologies (i.e., Euro classes) was obtained from local data as presented in Figure 6.3. The traffic counts took into account the different vehicle types that pass in the specific monitoring place, demonstrating the main contribution of passenger cars (PC, 85%) to the daily trips. Although buses (BUS) and heavy-duty vehicles (HDV) represented a smaller percentage of the daily traffic volume, they were also considered in the emission inventory due to their emission intensity when compared to PC. While BUS and HDV were considered as diesel vehicles, PC where split to 42% of gasoline vehicles, 56% of diesel vehicles and 1% of other types, assuming the national statistics data (INE, 2020).

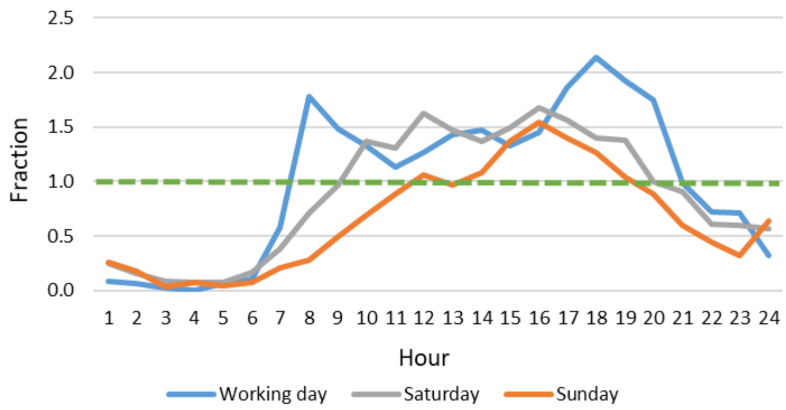


Figure 6.2. Traffic temporal profile for different weekdays (fraction = hourly volume/daily average volume).

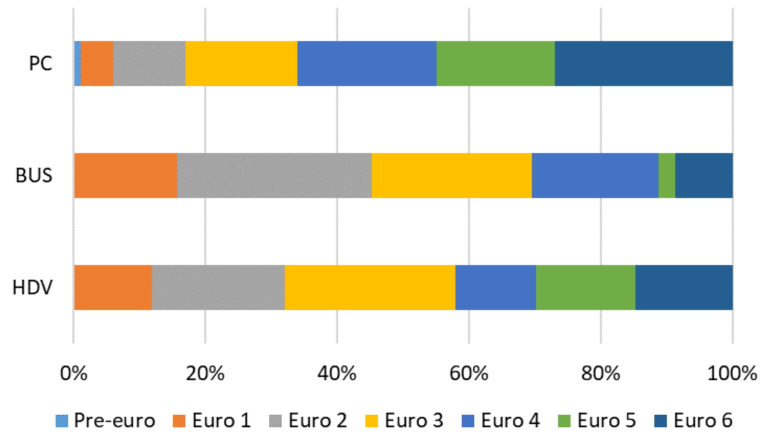


Figure 6.3. Vehicle fleet composition considered for emission modelling.

In addition to traffic emissions, meteorological data, topography and presence of buildings were compiled for air pollution dispersion modelling. Meteorological data from the Portuguese Institute of the Sea and Atmosphere (IPMA, https://www.ipma.pt/pt/index.html%20) for the closest location within the urban area were considered to provide input data required by ADMS-Roads (see Figure 6.4 and 5.5). Figure 6.4 presents the wind rose for the simulation period showing that the wind blew mostly from NW and E/NE and the wind speed was mostly below 4 m.s^{-1} with an average of 2.5 m.s^{-1}.

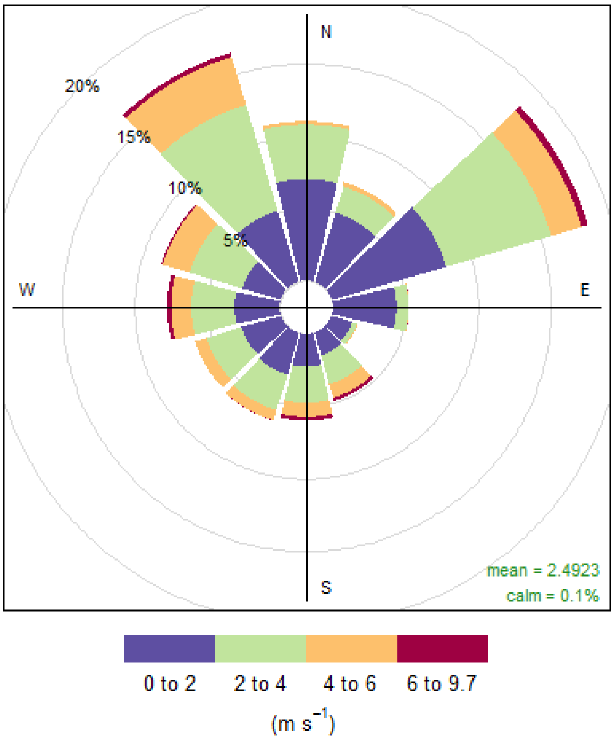


Figure 6.4. Wind rose for the study area for the simulation period (11/01 to 19/06/2019).

Air temperature variations for the study period are presented in Figure 6.5 considering hourly mean and range (i.e., minimum to maximum values) for each month. Additionally, to dispersion modelling, air temperature data were also used for the quantification of cold start emissions.

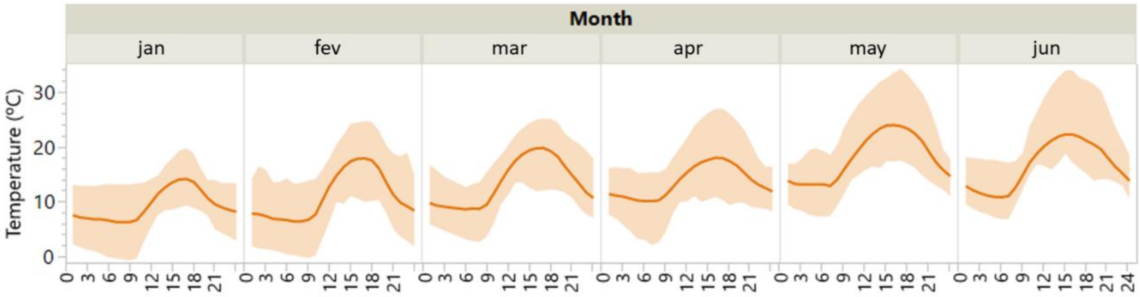


Figure 6.5. Ambient air temperature for the study period (11/01 to 19/06/2019): hourly means (line) and range (band) for each month.

To characterise the pollution dispersion at street level, street canyon dimensions were defined using city map in combination with local data and maximum building height. A regular grid with 30 m spatial resolution was considered for the modelling domain to obtain hourly outputs for pollutant concentrations.

6.2.3.2. PM measurements

A monitoring campaign was implemented for 6 months, from 11th of January to 19th of June 2019, to characterise particulate matter at a roadside location using gravimetric and optical measurements. The traffic station was located near a roundabout (40°12'24.5'' N, 8°26'13.5'' W) at the kerbside of an arterial road that connects interurban and urban roads.

A dual-spot AE33 Aethalometer with seven different wavelengths (λ = 370, 470, 520, 590, 660, 880, and 950 nm) was used to obtain BC concentrations with a 1-min resolution. The contributions of BC from fossil fuel and wood burning were estimated in real-time based on the spectral dependence of light absorption of different sources and using the Aethalometer model (Sandradewi et.al. 2008).

Additionally, PM_{10} samples were taken for 24 h (every 2 days, starting and finishing at mid-night) by a high-volume air sampler (MCV CAV-A/mb) equipped with pre-fired 15 cm diameter quartz fibre filters (Pall Corporation) and running at a flow rate of 30 $m^3.h^{-1}$. To obtain PM_{10} mass loadings, the quartz filters were weighted and then used to determine the carbonaceous content (organic and elemental carbon) through a thermo-optical transmission technique. More details on methodologies used for sampling and chemical analysis of filters have already been reported by Alves et al. (2021), Gonçalves et al. (2021) and Pio et al. (2022).

6.3. Results and discussion

Considering the research objectives, this section is divided into several parts: analysis of BC observations, BC emissions, and BC dispersion modelling results, including comparison with the observations and with the literature.

6.3.1. BC observations

Time series of on-line BC concentrations (originally with 1 min resolution) were obtained for the roadside location during the six-month measurement campaign, providing a unique opportunity to distinguish contribution of the main pollution sources, namely road traffic and biomass burning (Figure 6.6). Two different periods were considered in the analysis due to the contribution from different pollution sources. The cold period, before the spring equinoxes (before March 21), is characterised by a significant contribution from biomass burning to BC levels additionally to road traffic, and the warmer period (after March 21) with BC concentrations mostly derived from road traffic and a very small contribution from biomass burning. According to Luben et al. (2017), BC annual mean concentrations of 1.08–1.15 μg.m^{-3} have statistically significant associations with health.

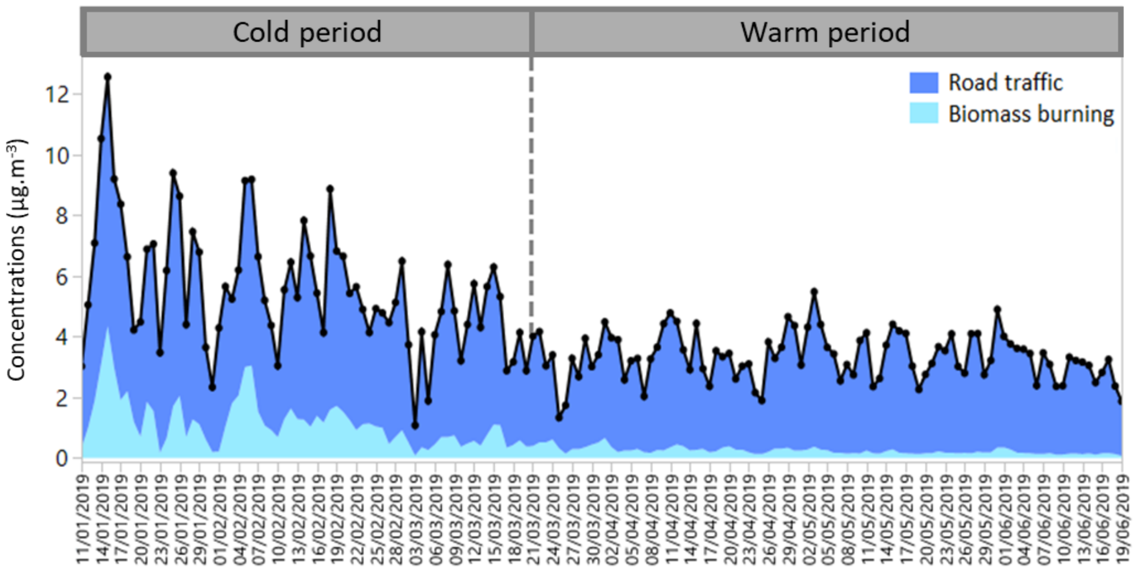


Figure 6.6 Temporal variation of BC daily concentrations and contribution of different source at sampling location (N = 160 days).

As expected, the BC concentrations during the cold period were higher than during the warm period. Throughout the sampling campaign, the hourly mean BC concentration was 4.32 μg.m^{-3} with a minimum of 0.17 μg.m^{-3} and a maximum of 26.36 μg.m^{-3}. BC from traffic activity was 4 to 3 times higher than BC from biomass burning even during the cold period. Due to the objectives of this study, only traffic-related pollution is analysed in the following sections, and BC measurements are used for the validation of modelling results, as described in section 3.3.

To analyse the fluctuations of BC pollution levels in combination with wind speed and direction, a PolarMap for traffic-related BC from the observations is presented in Figure 6.7. The PolarMap was obtained using the openair R package (Carslaw and Ropkins, 2012). This type of plot has shown to be valuable in quickly visualising potential sources and influences at a selected location (Carslaw and Davison, 2023). Thus, Figure 6.7 reveals that BC is mostly coming from northwest, where one of the main entrances to the city by motorway is located.

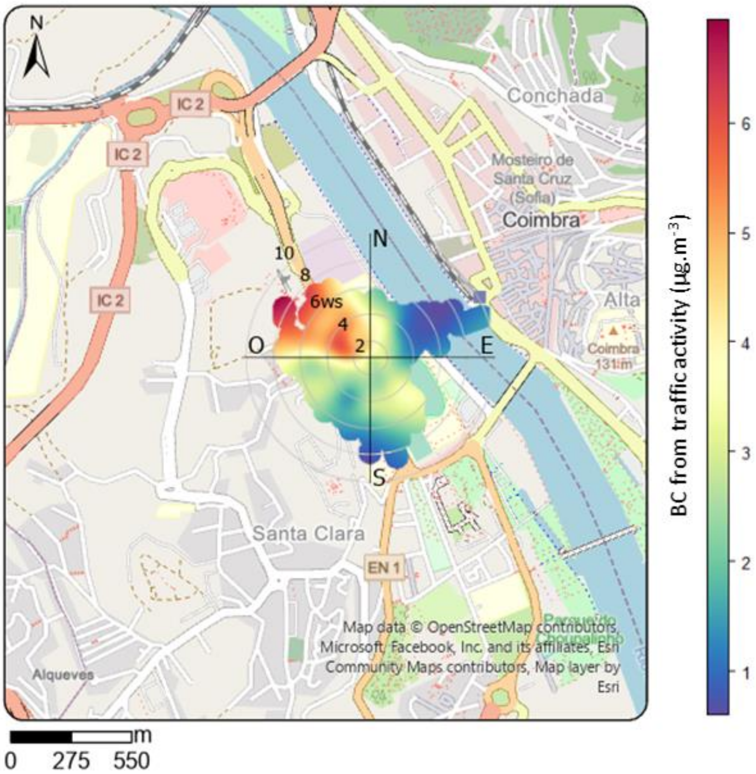


Figure 6.7. PolarMap of hourly mean BC concentrations from road traffic activity at the sampling location.

6.3.2. BC emissions

Based on the new developments and QTraffic model extension, BC emissions were quantified for the study area focusing on spatial and temporal variations. Table 6.5 summarises daily traffic emissions for the urban area of Coimbra, which ranges from 33 to 36 kg.day^{-1} for the whole domain considering different seasons. On average, the contribution of hot exhaust, cold start exhaust and non-exhaust processes to the BC daily emissions are 76%, 11% and 12%, respectively, but some differences could be identified based on seasonal analysis (Table 6.5), particularly for cold start emissions that depend on ambient temperature. Furthermore, Figure 6.8 presents the daily emissions at the road segment level, which achieve 600 g.km^{-1}.day^{-1} in the most congested roads of the city. For comparison with the literature, this value may be converted into an emission rate taking into account the road length, reaching a maximum of 2.68E+04 µg.s^{-1}. Recently, Lugon et al. (2021) obtained mean BC traffic-related emissions ranging from 1.2E+03 to 1.4E+05 µg.s^{-1} for the Paris road network, taking into consideration both exhaust and non-exhaust contributions. Thus, the road emissions predicted for Coimbra are one order lower than those for Paris.

Table 6.5. BC traffic emissions for Coimbra urban area.

BC emissions	Cold period		Warm period	
	kg.day^{-1}	%	kg.day^{-1}	%
Non-exhaust	4.33	12	4.33	13
Hot exhaust	26.54	73	26.54	80
Cold start exhaust	5.25	15	2.44	7
TOTAL	36.14	100	33.33	100

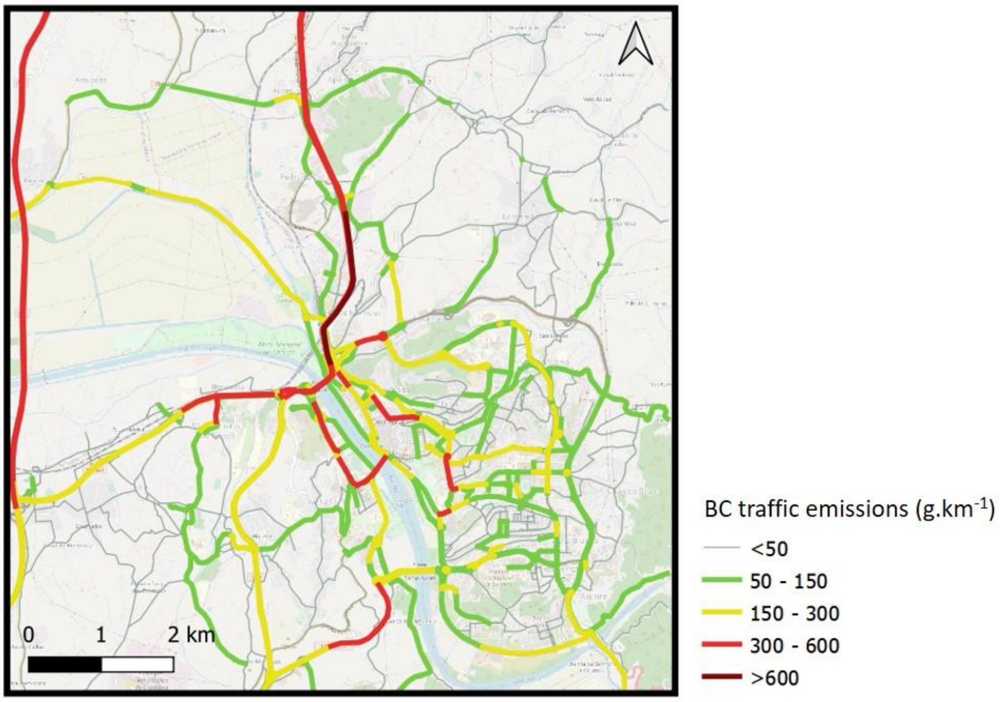


Figure 6.8. BC daily traffic emissions (g.km^{-1}.day^{-1}) for Coimbra city.

6.3.3. BC dispersion modelling results

Atmospheric concentrations of BC obtained from the dispersion modelling and at the measurement point are presented in Figure 6.9. Data are focused only on the contribution from road traffic, as extracted from model outputs and optical measurements. At the selected point, the average daily contribution from road traffic initially obtained by modelling (“Run1”) is 1.5 ± 0.9 μg.m^{-3}, which is significantly lower than the 3.6 ± 1.3 μg.m^{-3} derived from the observations (Table 6.6).

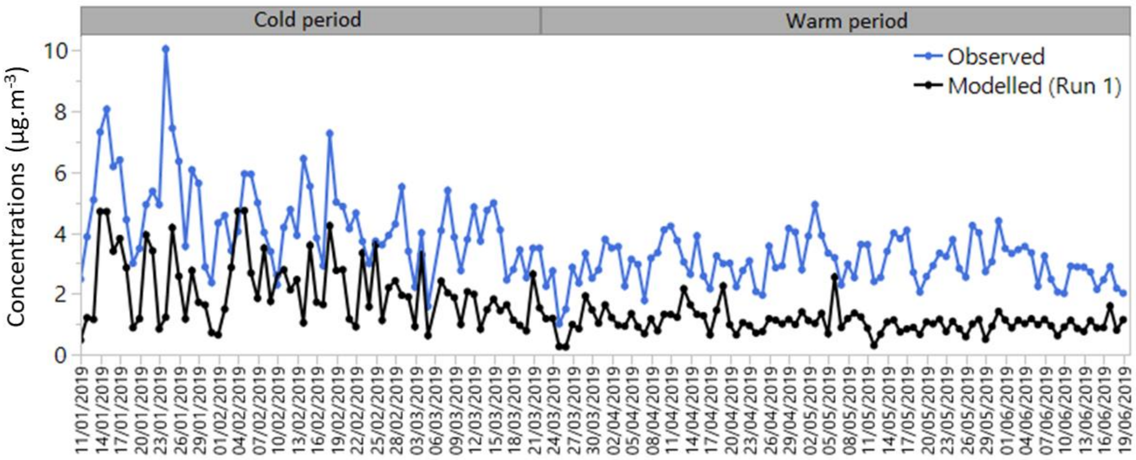


Figure 6.9. Time series of the road traffic contribution to BC concentrations modelled and observed at the sampling location (N=160 days).

It should be stressed that the emission inventory used as input data for Run1 includes already different emission processes (hot emissions, cold start, non-exhaust) and that completeness of the emission estimates was one of the priorities to avoid model underestimation, as previously discussed. However, the emission factors considered in this study are originally based on elemental carbon (EC) measurements, following the methodology reported by EMEP/EEA (2019a, 2019b) and modelling effort at urban/local scale requires more investigation on this point. For this purpose, local measurements obtained during the campaign implemented in Coimbra were considered using parallel measurements for BC and EC concentrations at the traffic location, as described in section 2.3.2. Based on these data, a linear relationship for BC and EC was determined: $BC = 0.93 \cdot EC + 2$, showing that the intercept is different from zero and BC is systematically lower than EC for the data range analysed (Forello et al., 2023). Based on this relationship, the dispersion results were corrected and considered as “Run2”.

Table 6.6. Statistical parameters for traffic related BC concentrations from the observations and modelling.

Concentration (μg.m^{-3})	Observed	Modelled	
		Run 1	Run 2
Average	3.6	1.5	3.4
Median	3.4	1.2	3.1
Minimum	1.0	0.3	2.2
Maximum	10.0	4.5	6.2
Standard Deviation	1.3	0.9	0.9

Run 1 – without correction; Run 2 – BC correction applied

The modelling performance was analysed using statistical parameters defined in Annex. Although the BC concentrations in Run2 are still underestimated (NMB = -0.1), the model evaluation parameters in general are significantly improved in comparison with Run1 (Table 6.7, Figure 6.10) and satisfy more strict performance criteria (-0.3>FB>0.3; NMSE<3; NAD<0.3; FAC2≥0.5; 0.7>MG>1.3; VG<1.6) proposed by Hanna and Chang (2012) and Herring and Huq (2018) for urban areas. However, the correlation between observations and predictions (r = 0.6) indicates a possibility for future improvements.

Table 6.7. Model validation parameters for BC concentrations.

	RMSE	r	NMB	NMSD	FAC2	FB	NMSE	NAD	MG	VG	Max. Error
Run1	2.3	0.6	-0.6	-0.3	0.3	-0.8	0.9	0.4	2.6	2.9	8.8
Run2	1.1	0.6	-0.1	-0.4	1.0	-0.1	0.1	0.1	1.0	1.1	6.9

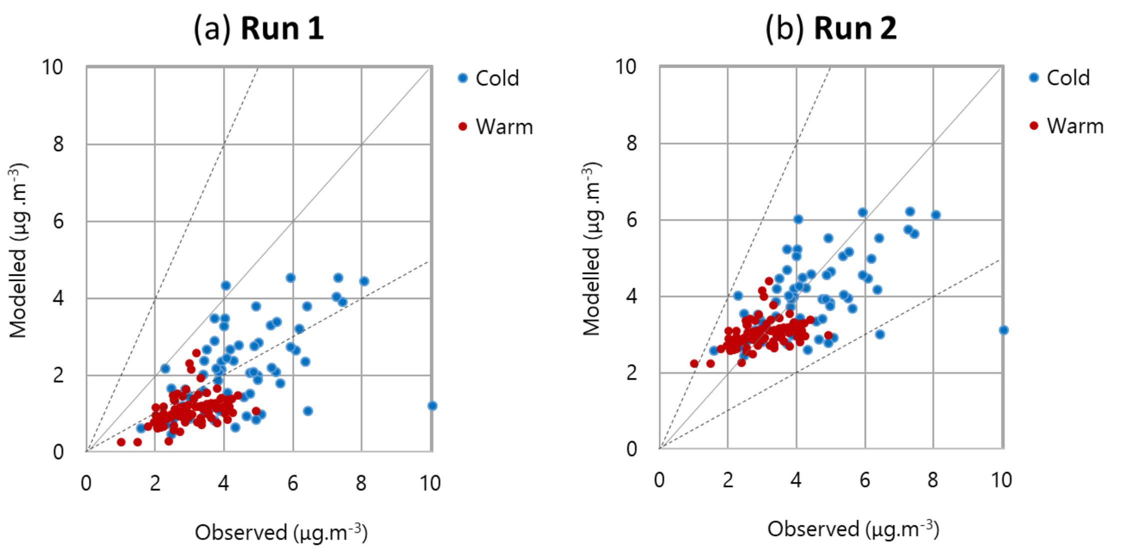


Figure 6.10. Scatterplot of BC traffic-related daily average concentrations (modelling vs observations) at sampling location: (a) model results from Run 1 (before BC/EC correction); (b) model results from Run 2 (after BC/EC correction).

The hourly variations of traffic-related BC concentrations obtained from Run2 and from the observations are presented in Figure 6.11 for different periods (cold and warm) and weekdays (working days or weekends). Overall, the higher road traffic contribution occurs in the rush hours on working days, which is expected since the station was located near an important entrance/exit of the urban area and being part of most commuting trips. There is a clear difference in the magnitude of the pollution levels during the cold and warm periods. In general, the distribution of the concentrations predicted by the model is right-skewed with the median been slightly lower than thus from the observations, except in the evening hours. Moreover, traffic temporal profile for weekends is different from working days (Figure 6.2) with the highest values about 16h but it is not reflected in the model outputs. This analysis may indicate that additionally to traffic emissions, the dispersion conditions considered by the model are very important.

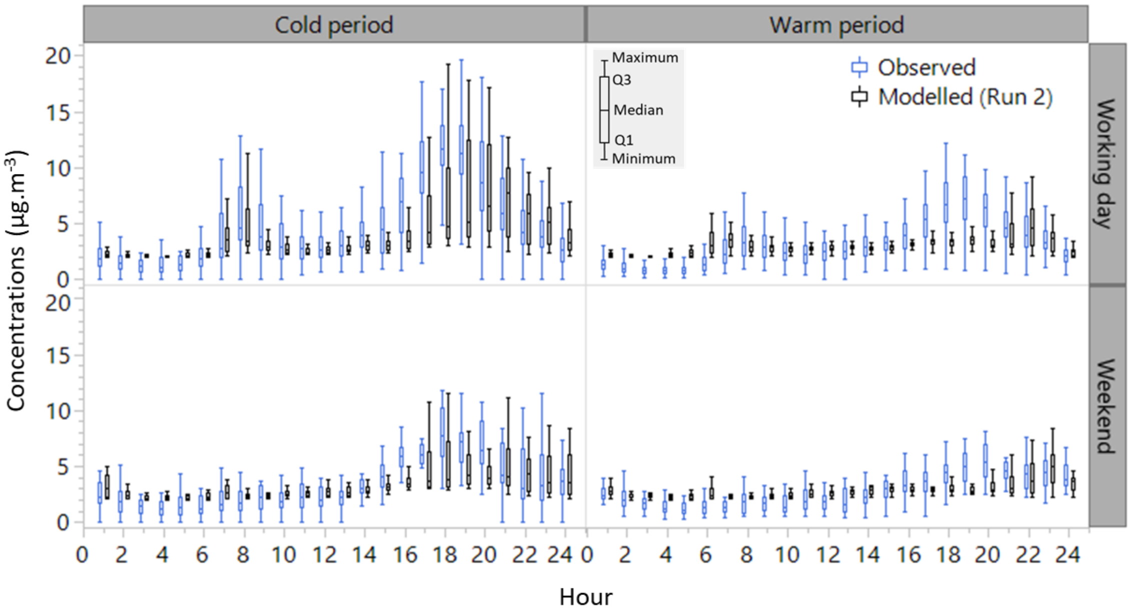


Figure 6.11. Box plot for BC traffic-related concentrations (modelled and observed) for cold and warm period and for weekends and working days (N = 3 811 hours).

The spatial distribution of traffic-related BC concentrations for the modelling domain and the 6-month study period is presented in Figure 6.12. While at the measurement point the BC mean concentration from road traffic activity was 3.4 µg.m^{-3} (Run 2), higher values were found within the domain, achieving maximum of 6.7 µg.m^{-3}. Thus, the spatial analysis highlights that measurements at a specific point may provide limited information and that modelling is very important to explain spatial variability of the pollution levels. In addition to average values, the maximum hourly concentrations for BC were also analysed. In the hotspots identified by the model, the BC maximum hourly concentration was 78.6 µg.m^{-3} in the cold period. Thus, this study demonstrates the importance of considering both spatial and temporal variations of BC for better understanding source contributions and defining mitigation measures.

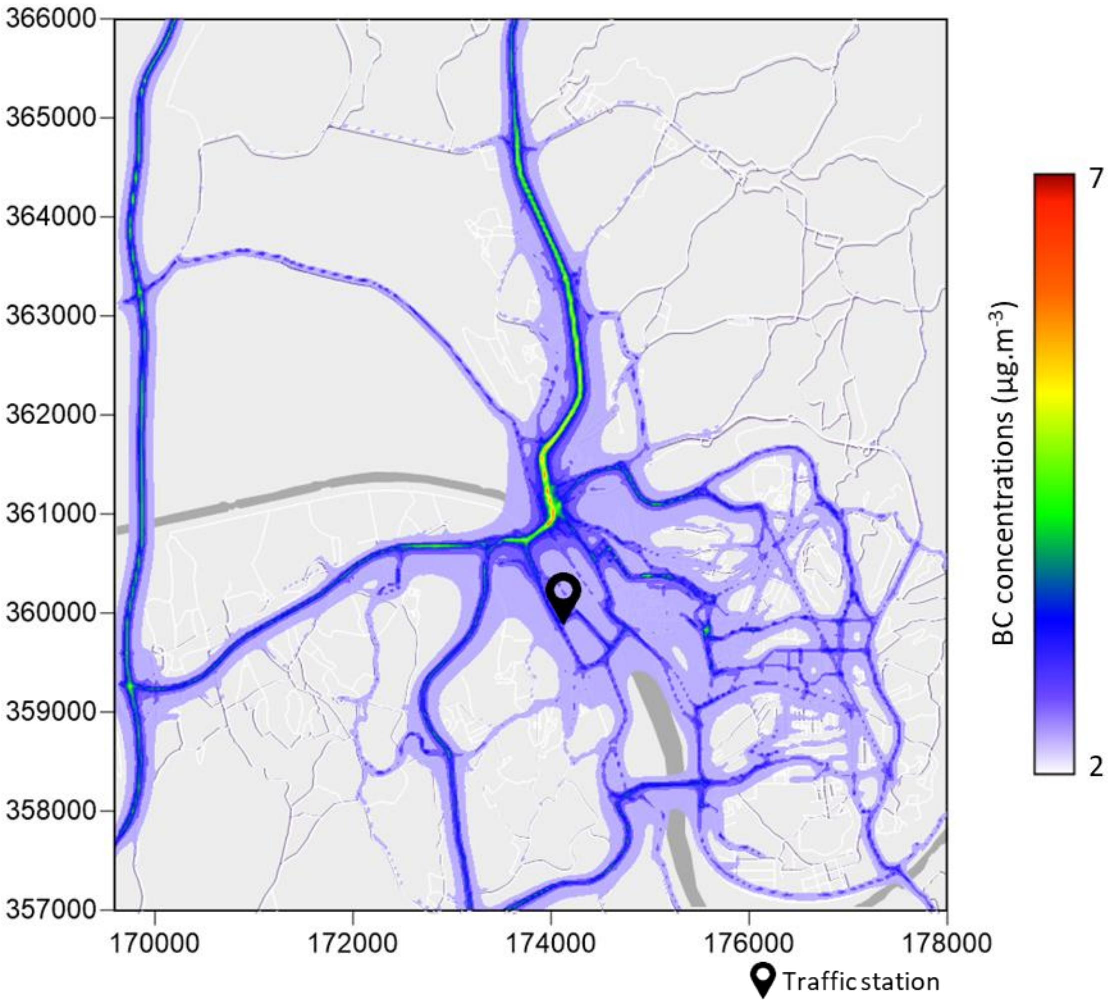


Figure 6.12. Spatial distribution of 6-month average BC concentrations (January - June, 2019).

Overall, the BC daily mean concentrations observed and modelled for Coimbra are in the range of values reported in previous studies for urban areas (Table 6.2). However, the spatial variability of the concentrations is rarely addressed because data are usually obtained from point measurements and a single point is used to characterise BC pollution levels in a city. There are few studies that investigate urban BC focusing on spatial distribution, as discussed in the introduction. To the best of our knowledge, this is the first study in a Portuguese city (Coimbra) where a dispersion modelling approach has been implemented to investigate the spatial distribution of BC with high spatial resolution, since the previous studies were based on point measurements (Table 6.1).

6.4. Conclusions

Regarding the potential effect of BC on human health and the environment, to be aligned with the Sustainable Development Goal 11 ("sustainable cities and communities") established by the United Nations General Assembly in 2015, the urgency of monitoring and managing BC in different cities is crucial. However, BC monitoring requires expensive and complex instruments, resulting in scarce data when compared to regulated pollutants. Thus, modelling procedures appear as a good alternative to address this issue. This study proposes a modelling approach and its validation with observational data to investigate the contribution of road traffic to the urban BC pollution.

The advances in emission modelling allowed to obtain BC concentrations in an hourly and daily basis and with 30 meters of spatial resolution for the entire city. Such disaggregation of traffic data enabled to obtain hotspots in Coimbra city centre where road traffic was estimated to contribute to the BC daily concentrations with 6.7 $\mu g.m^{-3}$ (mean values of a 6-month period). The spatial distribution of traffic-related BC concentrations showed hotspots of maximum hourly concentrations reaching around 78.6 $\mu g.m^{-3}$ in the cold period, while an average concentration for a 6-month period at the traffic monitoring site was 3.4 $\mu g.m^{-3}$. This emphasises the importance of the analysis of BC concentrations at the city scale with high temporal and spatial resolution additionally to point data observations.

The measurement campaign implemented in a crowded traffic location during 6-month allowed to obtain valuable data on the contribution of road traffic to BC pollution levels with high temporal resolution thus providing crucial information for the model validation and helping to improve our knowledge on possible sources of the modelling uncertainties. Underestimation of BC concentrations by dispersion modelling depicted in the literature is frequently related to the incompleteness of the emission inventories. Therefore, it was crucial to validate the proposed modelling approach against the observations. Two datasets were compared on a daily and hourly basis. The validation demonstrated that the (in)completeness of the road traffic emission inventory (considering cold start and non-exhaust emissions, additionally to hot emissions) may not be the main reason for the underestimation of BC concentrations by the dispersion model. Future improvements in emission factors should be considered along with a deeper evaluation of the dispersion conditions considered in the modelling and their influence on hourly fluctuations of the concentrations.

Implementation and validation of the integrated modelling approach for traffic-related BC with high spatial and temporal resolution may contribute to advance the knowledge on BC at urban scale. Also, it is important to denote that the proposed methodology was applied and showed a good performance when validated with real data for a period of 6 months. Furthermore, the proposed approach provided essential input data to study human exposure to urban concentrations of BC and move towards safer and sustainable cities.

Chapter 7

7. Final remarks

Ambient air pollution is a concern in many parts of the world with environmental and health impacts, especially in urban areas where most population are exposed to harmful pollutants. Regarding that the world is becoming a "big urban centre", the monitoring and management of urban air quality turns an urgent matter. Human health is affected by different air pollutants, but some of them have more severe consequences, such as PM, emitted by road traffic in urban context. Thus, improved understanding of PM health effects requires additional information about urban air quality. Moreover, to prevent human exposure to air pollution episodes, it has become urgent to anticipate them and understand the trends. These concerns are in the core of the urban air quality forecast research.

Therefore, the main purpose of the research presented in this thesis was to develop and implement a consistent approach to AQ modelling capable of assess and forecast aerosol particles in the urban atmosphere, especially looking to the road traffic contribution. The development of this modelling system required improvements in terms of both local and background contributions to urban air pollution. Additionally, in order to improve the characterization of the urban aerosol, modelling techniques were combined with the measurements allowing also the prediction of BC concentrations with high spatial and temporal resolution. The main achievements are presented and organized in seven chapters starting with the overall introduction and a literature review of particular topics, traffic-related emissions, air quality management and forecasting, cascade modelling tools applied to urban air quality analysis and forecast.

7.1. Summary of the research findings

"Modelling results are only ever as good as the input data!"
Melody Horan – ADMS User Group Meeting 2020

In light of this statement and with the literature review (Chapter 2), a good urban air quality modelling approach will intrinsically be related to the quality of its required input data. Well, one of the most important input data to predict PM in urban atmosphere is local emissions induced by road vehicles. Therefore, Chapter 3 and 4 were dedicated to improve an emission modelling approach applied to quantify road traffic emissions, focusing on different vehicle processes.

In Chapter 3 a new modelling method to quantify cold start emissions at road segment was proposed based on cold distance concept, allowing spatial analysis of the cold start emissions for line sources while preserving trip Origin-Destination data from transportation modelling. Thus, a cold start emission module was developed and implemented. Additionally, in order to access traffic exhaust daily emissions, both cold and hot emissions for line sources were calculated, which allowed to obtain the significance of cold start emissions in urban areas. Furthermore, the cold start contribution were obtained at a road segment level and taking into account different seasons, daily periods and road types. During winter, high contribution of cold start emissions is demonstrated with median values above 70% for carbon monoxide and hydrocarbons at local roads, where most people leave or work. The modelling tool developed and applied in this study demonstrates the importance of quantifying cold start emissions with fine spatial resolution (i.e. at road segment level). For instance, even for pollutants like NO_x that have a lower cold start contribution (3%, mean value) when the whole urban area is analysed, increases considerably when the analysis is made at the road segment level (8%, mean value near residential and working areas). Therefore, regarding the urgency to manage urban air quality especially due to harmful health effects, there is large variability of cold start emissions contribution at urban roads that should not be neglected. The identification of these variabilities was facilitated by the uniqueness of model to preserve the trip O-D pair information, enabling the allocation of cold emissions to individual road segment of a given path. Moreover, the trip O-D information allows to access cold start contributions of different city zones. Thus, this modelling tool would provide valuable information for traffic pollution studies and quantification of environmental benefits due to emissions (hot+cold) reduction. For instance, it could be innovative to access the contribution of cold start emissions in different climate regions since climate change will affect ambient temperature that directly influences cold start emissions, including CO_2 emissions that are highly affected by cold start processes.

Another important parcel of vehicle emissions occur due to non-exhaust processes. Therefore, Chapter 4 was dedicated to access the contribution of non-exhaust PM emissions to urban air pollution, by applying an integrated modelling approach (i.e. transport-emission-air quality modelling system). Additionally, in this chapter a detailed characterization of traffic-related PM pollution at urban scale was performed by using both an integrated modelling approach and *in-situ* measurements. The *in-situ* measurements, which allowed to understand PM size distribution over time, indicated a higher presence of fine particles during the monitoring campaign due to forest fires and road traffic activity. The relevant measured data regarding PM distribution in 31 size range, meteorology parameters and traffic activity, can be crucial for source apportionment and population exposure studies. The importance of including non-exhaust processes in both PM emission and concentrations quantification was demonstrated, since more than 50% of PM traffic emissions were caused by non-exhaust processes, resulting in an increase of 9 $\mu g.m^{-3}$ in PM_{10} hourly concentrations. Regarding that vehicles emit less and less exhaust emissions due to continues strict regulations, non-exhaust emission quantification will be a crucial component of research in the next years in order to create policies and legislations that allow their reduction.

While the Chapter 3 and 4 were dedicated to improve the quantification of traffic related emissions, allowing the quantification of PM local contribution to urban air pollution; Chapter 5 was focused on advancing the quantification of background contributions. Thus, in Chapter 5 two existing regional scale services (CAMS and a national forecast service based on CHIMERE) were explored and their applicability to forecast PM_{10} concentrations at the urban scale were analysed. In general, both regional models demonstrated a good statistical performance and allowed to obtain background concentrations for urban AQ forecasting. Although the national forecasts by CHIMERE reproduced better the episodes related to natural events, CAMS revealed a better performance for the long-term statistics. These findings are quite important since CAMS provides free forecasting global and regional data that could be used in different case studies worldwide. Additionally, this study highlights the importance of quantifying air pollution with high spatial resolution, since in the urban hotspots PM_{10} annual values achieve 38 $\mu g.m^{-3}$, a value that is around double of regional scale predictions with lower cell resolution, and it is not represented by point measurements (25.4 $\mu g.m^{-3}$) at the traffic monitoring station not located at the specific hotspots. As a result, while urban-scale modelling approaches cannot replace *in-situ* measurements, they are a good option for assessing air quality and, additionally, urban AQ forecast.

This PhD research have gone beyond its initially defined goals by developing, in Chapter 6, a methodology to quantify BC emissions and concentrations at urban scale. BC is a component of PM which has received a great attention in the last years due to its impact to human health and climate change contribution. Actually, WHO have recently recommended more actions to enhance further research on BC's risks and approaches for mitigation (WHO, 2021). However, when compared to regulated pollutants like PM_{10} and $PM_{2.5}$, there is a lack of BC measured data worldwide. Thus, Chapter 6 proposes a

BC assessment approach based on modelling to help overcoming this gap. The modelling approach proposed and implemented in this work was validated with observations collected during a six-month monitoring campaign at an urban traffic station using gravimetric measurements for elemental carbon (EC) and combustion-derived BC concentrations from an aethalometer. The results showed that daily emissions of BC from traffic may achieve more than 600 $g.km^{-1}.day^{-1}$ for some road segments considering the contribution of hot exhaust emissions (76%), cold starts (11%) and non-exhaust emissions (12%). The dispersion modelling results showed that in the monitoring site road traffic contributes with 3.4 $\mu g.m^{-3}$ to 6-month average BC concentrations, but in the hotspots this contribution reaches 6.7 $\mu g.m^{-3}$. Moreover, hourly values at hotspots may achieve 78.6 $\mu g.m^{-3}$. This finding stresses the importance of spatial and temporal analysis of BC with high resolution at city scale, additionally to point measurements. Furthermore, the 6-month monitoring data provided crucial information for the model validation, helping to improve the knowledge on possible sources of the modelling uncertainties. Underestimation of BC concentrations by dispersion modelling depicted in the literature is frequently related to the incompleteness of the emission inventories. However, the validation demonstrated that the (in)completeness of the road traffic emission inventory may not be the main reason of BC's underestimation by dispersion modelling. Therefore, future advancements in emission factors as well as a deeper analysis of the dispersion conditions considered in AQ simulations and their impact on hourly changes of the concentrations should be considered. The modelling approach proposed in this work could contribute to advance the knowledge on cause-effect of BC pollution at urban scale and provide essential input data to human exposure studies, moving towards safer and sustainable cities.

Overall, by achieving all the previously proposed research objectives, the results obtained from this thesis may potentially contribute to advance in understanding urban aerosol particles in the atmosphere, considering road traffic as one of the primary sources of urban air pollution. Actually, road traffic remains a main contributor to urban air pollution in the case study deeply analysed from Chapter 3 to 6 and especially looking to BC, despite improvements in vehicle technologies circulating in the urban area. Moreover, the advances made in the integrated modelling approach provides a promising opportunity for urban AQ assessment and forecast of not only particulate matter but also different traffic–related pollutants. Although the urban AQ forecast system was implemented at Coimbra city, it uses European CAMS data to obtain background contributions; thus, given the globalization of the European atmospheric services, the modelling approach may be used to forecast air quality of different cities if the appropriate input data are available. Furthermore, the PM temporal and spatial variation and its size distribution allows us to provide crucial information for health impacts analysis and climate change research, which support decision making for urban sustainable development.

7.2. Future research

An improvement of the methodology for urban air quality assessment and forecast is a continuous task. Further developments of the integrated modelling system could include PN (Particulate Number) predictions and inclusion of resuspension process, since it might have a significant contribution to the overall PM non-exhaust emissions. The results from the different monitoring campaigns may play a key role in further research, because detailed data were obtained in terms of: NO_x mass; PM mass, surface area and number; BC/EC and organic carbon mass; PM chemical compositions; meteorological parameters; traffic counting data for different vehicle classes and periods.

Given the shift of the literature to emission and air quality estimations with greater spatial and temporal resolution to few meters and seconds, future developments should be made towards integrating microscale traffic simulation and emission data in air quality forecasting.

For future research, the presented modelling system could be extended to address population and individual exposure to harmful pollutants in the urban area. Moreover, not only the impact of aerosols to human health should be addressed but also to building environment and regarding climate change issue. On this topic, addressing individual contribution due to their travel modal choice could be suggested, in a way of understanding the influence in urban/local air quality and associated impacts.

Furthermore, the implementation of the integrated modelling system for AQ forecasting in different case studies, especially looking to places where the human exposure to particulate matter is alarming (e.g. Cape Verde), could be also a line of future research.

Finally, given the health risks associated to poor air quality culminated in the recent UN General Assembly resolution which declared the access to Clean Air as a human right. This will imply a constant AQ monitor and control, thus, a continuous improvement of AQ management systems and approaches is a matter of guaranteeing a human right and saving millions of lives.

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Annex

Table S1 Statistical parameters for model validation (Hanna and Chang, 2012; Janssen et al., 2022).

Parameter	Formula	Range	Desirable
Root mean squared error, RMSE	$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(M_i - O_i)^2}$	-	0
Pearson Correlation Coefficient, r	$R = \frac{\sum_{i=1}^{N}(M_i - \bar{M})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^{N}(M_i - \bar{M})^2}} \cdot \sqrt{\sum_{i=1}^{N}(O_i - \bar{O})^2}$	[-1;1]	1
Normalised mean bias, NMB	$NMB = \frac{\sum_{i=1}^{N} M_i - O_i}{\sum_{i=1}^{N} O_i}$	[-1;1]	0
Normalised mean standard deviation, NMSD	$NMSD = \frac{\sigma_M - \sigma_O}{\sigma_O}$	[-1;1]	0
Fraction of predictions within a factor of two of observations, FAC2	$FAC2 = \frac{1}{N}\sum_{1}^{N} n_i$ with $n_i = \{1\ for\ 0.5 \leq \left\lvert\frac{M_i}{O_i}\right\rvert \leq 2\ else\ 0$	[0; 1]	1
Fractional mean bias, FB	$FB = \frac{2}{N}\sum_{i=1}^{N}\frac{(M_i - O_i)}{(M_i + O_i)}$	[0.5; 2]	0
Normalized mean-square error, NMSE	$NMSE = \frac{\frac{1}{N}\sum_{i=1}^{N}(M_i - O_i)^2}{\bar{M} \times \bar{O}}$	-	0
Normalized absolute difference, NAD	$NMSE = \frac{\frac{1}{N}\sum_{i=1}^{N}\lvert M_i - O_i\rvert}{\bar{M} + \bar{O}}$	-	0
Geometric mean bias, MG	$MG = \exp\left[\frac{1}{N}\sum_{i=1}^{N} ln\left(\frac{M_i}{O_i}\right)\right]$	-	0
Geometric variance, VG	$VG = \exp\left[\frac{1}{N}\sum_{i=1}^{N} ln\left(\frac{M_i}{O_i}\right)^2\right]$	-	0
M_i is a predicted value and O_i is an observed value. σ_M is the standard deviation for predicted values and σ_O is the standard deviation for the observed values.			