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SANDRA SOFIA
SILVA SORTE

ESTRATÉGIAS DE APOIO PARA MELHORAR A QUALIDADE DO AR EM ÁREAS PORTUÁRIAS

SUPPORTING STRATEGIES TO IMPROVE AIR QUALITY OVER HARBOUR AREAS

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Tese apresentada à Universidade de Aveiro para cumprimento dos requisitos necessários à obtenção do grau de Doutor em Ciências e Engenharia do Ambiente, realizada sob a orientação científica da Doutora Maria Alexandra Monteiro, Investigadora Auxiliar no Departamento de Ambiente e Ordenamento da Universidade de Aveiro, coorientação científica do Doutor Carlos Borrego, Professor Catedrático do Departamento de Ambiente e Ordenamento da Universidade de Aveiro e da Doutora Vera Augusta Moreira Rodrigues Investigadora Auxiliar no Departamento de Ambiente e Ordenamento da Universidade de Aveiro. Modalidade alternativa à apresentação de Tese, nos termos do artigo 64.º-A do Regulamento de Estudos da Universidade de Aveiro.

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I dedicate this Thesis to João Peres Ribeiro,
who always believed in me.

o júri

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palavras-chave

portos, emissões, mitigação, dispersão de poluentes, C-PORT, modelação CFD, túnel de vento

resumo

Apesar do seu papel-chave no desenvolvimento económico, os portos marítimos constituem uma ameaça ambiental, com impactes na qualidade do ar, clima local, e saúde humana, devido à emissão de inúmeros poluentes. Episódios de má qualidade do ar a nível local são particularmente preocupantes no caso de portos localizados nas imediações de áreas urbanas densamente povoadas, pondo em risco a saúde dos habitantes locais.

Esta Tese focou-se no impacte das emissões portuárias na qualidade do ar em portos e suas vizinhanças urbanas. O objetivo final foi a produção de recomendações de suporte à tomada de decisão no setor portuário e gestão da qualidade do ar, usando o Porto de Leixões como caso-de-estudo. Após uma revisão do estado-da-arte neste campo, foi desenvolvido um inventário de emissões de alta-resolução, aplicando as duas metodologias mais frequentemente usadas na comunidade científica. Foram compilados dados sobre navios e equipamentos portuários, permitindo a quantificação das emissões e identificação das suas fontes maioritárias. Deste procedimento resultaram recomendações sobre o desenvolvimento de uma nova metodologia harmonizada. Ficou ainda evidenciada a relevância da atualização dos fatores de emissão e dos dados disponíveis sobre as diferentes atividades portuárias.

Dispondo deste inventário de emissões, o C-PORT, uma ferramenta *web* de escala comunitária, foi aplicado pela primeira vez em portos europeus, para simular o impacte das emissões marítimas na qualidade do ar local. A comparação dos valores modelados com medições de campo validou a aplicação desta ferramenta ao caso-de-estudo do Porto de Leixões. A concentração mais elevada de PM10 foi registada no Terminal de Contentores Sul, registando-se também elevada (> 100 $\mu g/m^3$) concentração de NOx junto à autoestrada vizinha. A maior contribuição (cerca de 80 %) para a emissão global de PM10 na área de estudo adveio de fontes de emissão terrestres, enquanto os navios atracados contribuíram com cerca de 50 % das emissões de NOx. Esta Tese inclui a análise de medidas de mitigação capazes de melhorar a qualidade do ar em portos marítimos e sua vizinhança. O caso-de-estudo apresentado foca-se na dispersão de poluentes, com o intuito de controlar a emissão de partículas de petcoke do Porto de Aveiro, e o seu impacte nas comunidades vizinhas. Com esse objetivo, foi estudada, através de simulação física e numérica, a composição, dimensão e posicionamento de uma barreira física. A solução otimizada permitiu reduzir em 74 % – 88 % para as direções de vento mais frequentes/críticas nesta região, estando atualmente implementada no terreno.

keywords

harbor, emissions, mitigation, pollutant dispersion, C-PORT, CFD modelling, wind-tunnel

abstract

Despite their key contribution to economic development, harbours pose environmental threat, affecting air quality, local climate, and human health, due to the release of several pollutants. Poor local air quality episodes are particularly concerning when harbours are located near densely populated urban areas, threatening inhabitants' health.

This Thesis was focused on the assessment of the impact of harbour emissions on the air quality over harbours and their surrounding urban areas, with a final goal of producing guidelines to support decision-making in the harbour sector and air quality management, using Port of Leixões as a case-study. After reviewing the state-of-the-art in this research field, a high-resolution emission inventory was developed, based on the two most used methodologies within the scientific community. Data about ship and cargo handling equipment were compiled, allowing the quantification of emissions and identification of their main sources. The comparison of the two methodologies indicates that a new harmonized methodology is recommended, besides the need of continuous update of emission factors and activity data.

Having the detailed emission inventory, the community-scale webtool C-PORT was applied for the first time in European harbours to simulate the impact of the maritime emissions on local air quality. The comparison of modelled and observed values validated its application for the case study of Port of Leixões. The highest PM10 concentrations were found near the South Container Terminal of Port of Leixões, while NOx concentrations above 100 $\mu g/m^3$ were also found near the highway. Land-based emission sources exhibited the highest contribution (around 80 %) to the PM10 concentrations in the study area, while 50 % of NOx concentration was due to docked ships.

Mitigation measures were investigated and assessed to improve air quality in harbours and their surroundings. In a case-study, pollutant dispersion was addressed, aiming to control fugitive petcoke emissions and their impact on Port of Aveiro's neighbour communities. Optimal structure, size and position of a physical barrier were defined based on numerical and physical modelling, achieving a maximum reduction in petcoke dust reaching the nearby residential area of 74 – 88 % for the most frequent/critical wind directions. The studied barrier has been implemented in the field and monitoring campaigns are currently being carried out to assess its effectiveness.

Publications related to this Thesis

International Journals

1. **Sorte S.,** Rodrigues V., Ascenso A., Freitas S., Valente J., Monteiro A., Borrego C. (2018) Numerical and physical assessment of control measures to mitigate fugitive dust emissions from harbour activities. Air Quality Atmosphere and Health, 11, (5), 493-504. DOI: 10.1007/s11869-018-0563-7

2. **Sorte S.,** Lopes M., Rodrigues V., Leitão J., Monteiro A., Ginja J., Coutinho M., Borrego C. (2018) Measures to reduce air Pollution caused by fugitive dust emissions from harbour Activities. International Journal of Environmental Impacts, 1, (2), 115-126. DOI: 10.2495/EI-V1-N2-115-126

3. **Sorte S.,** Arunachalam S., Naess B., Seppanen C., Rodrigues V., Valencia A., Borrego C., Monteiro A. (2019) Assessment of source contribution to air quality in an urban area close to a harbour: Case-study in Porto, Portugal. Science of the total environmental, 662, 347-360. DOI: 10.1016/j.scitotenv.2019.01.185

4. **Sorte S.,** Rodrigues V., Borrego C., Monteiro A. (2020) Impact of harbour activities on local air quality: A review. Environmental Pollution, 257. DOI: 10.1007/s11869-021-00982-3

5. **Sorte S.,** Rodrigues V., Lourenço R., Borrego C., Monteiro A. (2021) Emission inventory for harbour-related activities: comparison of two distinct bottom-up methodologies. Air Quality, Atmosphere & Health,14, 831-842. DOI: 10.1007/s11869-021-00982-3

International Congresses and Workshops

1. **Sorte S**., Rodrigues V., Ascenso A., Borrego C., Monteiro, A. Emissions from maritime and harbour activities impacting the air quality over the urban area around port of Leixões. Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes (HARMO18), 9-12 October 2017, Bologna.

2. Isakov V., Barzyk T., Arunachalam S., Naess B., Seppanen C., Monteiro A., **Sorte S.** Web-based air quality modeling system for near-port assessments: example of application in Porto, Portugal. Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes (HARMO18), 9-12 October 2017, Bologna.

3. Borrego C., **Sorte S.,** Rodrigues V., Monteiro A., Ginja J, Emissões de petcoque e qualidade do ar na envolvente portuária: o caso de estudo do Porto de Aveiro. XIX Encontro da Rede de Estudos Ambientais em Países de Língua Portuguesa (REALP), 12 - 15 setembro 2018, Fortaleza.

4. **Sorte S.,** Rodrigues V., Borrego C., Monteiro A. 2018. Inventário de Emissões marítimas e portuárias: caso de estudo do Porto de Leixões. CIALP - Conferência Internacional de Ambiente em Língua Portuguesa, 8 - 10 maio 2018, Aveiro.

5. **Sorte S**. Neves A., Rafael S., Rodrigues V., Lopes M., Borrego C., Monteiro A. 2019. Studying mitigation measures to improve air quality in the surroundings of a seaport, 19th International

Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, (Harmo 2019), Bruges, 3 - 6 June 2019

National Conferences

1. **Sorte S.,** Borrego C., Monteiro A. Uma plataforma on-line para avaliação do impacto das emissões marítimas e portuárias. XXI Congresso da Ordem dos Engenheiros, 23-24 novembro 2017, Coimbra.

Interview magazine

1. **Sorte. S.,** Uma plataforma on-line para avaliação do impacto das emissões marítimas e portuárias, Magazine, INGENIUM, vol. 161, novembro 2017.

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List of Acronyms

AE	Auxiliary Engine power
AB	Boiler Energy default
APA	Administration of the Port of Aveiro
APDL	Administration of the Port of Leixões
AQS	Air Quality Stations
AQG	Air Quality Guideline
BC	Black Carbon
CARB	California Air Resources Board
CFD	Computational Fluid Dynamics
CO	Carbon Monoxide
CO_2	Carbon Dioxide
CH4	Methane
CHE	Cargo Handling Equipment
C-PORT	Community screening tool for near-PORT
CMB	Chemical Mass Balance
DFA	Deterioration Factors Adjustment
DR	Deterioration Rate
Disp	Atmospheric stability class
ECA	Emission Control Areas
EEA	European Environment Agency
EF	Emission Factors
EMEP/EEA	European Monitoring and Evaluation Program/European Environmental Agency
ENTEC	Entec UK Limited
EPA	Environmental Protection Agency
ESPO	European Sea Ports Organization
EU	European Union
EU-28	28 EU Member countries
FCF	Fuel Correction Factor
GHG	Greenhouse Gas
GT	Gross Tonnage
HRS	Annual Hours of use
ITF	International Transport Forum
IMO	International Maritime Organization
IVL	Swedish Environmental Research Institute
LF	Load Factor
LFA	Load Factor Adjustment

Lmon	Monin-Obukhov Length
LMIS	Lloyd's Number
MARPOL	International Convention on the Prevention of Pollution from Ships
MCR	Maximum Continuous Rated
ME	Maximum Main Engine power
MLR	Multiple Linear Regression
N	North direction
NBL	Nocturnal Boundary Layer
NE	Northeast direction
NECAs	NO_X Emissions Control Areas
Ni	Nickel
NO_2	Nitrogen Dioxide
NOx	Nitrogen Oxides
NW	Northwest direction
O_3	Ozone
P	power rate of the engine
PAHs	Polycyclic Aromatic Hydrocarbons
PCA/APCS	Main Component Analysis/Absolute Main Component Score
PM	Particulate Matter
PM2.5	Particulate Matter with an aerodynamic diameter smaller than 2.5 µm
PM10	Particulate Matter with an aerodynamic diameter smaller than 10 µm
PMF	Positive Matrix Factorization
RANS	Reynolds-Averaged Navier-Stokes
RCP	Representative Concentration Pathway
RefHt	Reference Height for wind
RL	Residual Layer
RO	Residual Fuel Oil
S	South direction
SBT	Solid Bulk Terminal
SFC	Specific Fuel Consumption
SE	Southeast direction
SECA	Sulphur Emission Control Areas
SO_x	Sulphur Oxides
SO_2	Sulphur Dioxides
SW	Southwest
Temp	Ambient Temperature
$T_{Manoeuv}$	Time spent during manoeuvring
T_{Hotell}	Time spent at hotelling

UNC-IE	University of North Carolina's Institute for the Environment
US/SCG	US Starcrest Consulting Group
uStar	surface friction velocity
V	Vanadium
VOCs	Volatile Organic Compounds
W	West direction
Wd	Wind Direction
WHO	World Health Organization
Ws	Wind Speed
Zimech	Height of the Mechanically generated boundary layer
Zo	Surface Roughness Length
ZH	Zero-Hour emission rate

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Chapter 1

1. Introduction

1.1 Framework

It is undeniable that harbours are key contributors to the social and economic development (Agrawal H. et al., 2008; Eyring et al., 2010). Over the years, the movement of ships has been rising, following the growth of international trade (Georgieva et al., 2007). Despite the economic benefit they provide, activities associated with port operations are also environmentally concerning, with potential effects on local climate, human health, and ecosystems (Lonati et al., 2010). In fact, the increasing movement of ships entails a consequent increase in the release of atmospheric pollutants, which has drawn attention to the environmental impact of these activities, namely in the case of harbours located near densely populated urban areas (Alastuey et al., 2007; Cesari et al., 2014). Besides that, the increasing flow of commercial ships circulating into and out of ports affects not only major ports, but also medium and small-scale ones.

Despite the progress achieved in the last decades regarding air pollution control, owing to the application of strict measures to reduce emissions, several port cities across the world are still facing severe air pollution episodes, with regular exceedances of the European Union (EU) legislation limits and the stricter World Health Organization (WHO) guidelines. According to the European Sea Ports Organization (ESPO), the top environmental priority for harbours is the local air quality, reflecting its importance on the health of port workers and nearby residents (ESPO, 2013). Sulphur oxides (SOx), nitrogen oxides (NOx), particulate matter (PM), carbon monoxide (CO) and polycyclic aromatic hydrocarbons (PAHs) are emitted to the atmosphere as a direct result of maritime activities (Viana et al., 2020). Improved scientific knowledge about these types of emissions remains scarce and there are relatively few studies on the impact that mitigation measures have on local air quality (Borrego et al., 2007; Isakson et al., 2001). Currently, many emission inventories suffer from lack of data about port activity and use outdated Emission Factors (EF). A harbour emission inventory is essential for harbour authorities to quantify the impacts of the maritime activity growth, and to develop mitigation plans to face their environmental impacts (EPA, 2009; SCG, 2019). Without a specific-oriented emission inventory for harbour areas, management entities face strong difficulties to (i) identify and assess opportunities for the implementation of emission reduction measures, and (ii) to quantify the effectiveness of those measures over time, through the monitoring of the achieved reduction in emissions (Liang et al., 2016; Tao et al., 2013). After having a detailed emission inventory, numerical air quality modelling is a crucial tool for the design and management of the harbour activities, since it can assess the impact of planning alternatives on air quality, supporting the evaluating currently implemented measures/scenarios or by testing new ones.

1.2 Objectives

The main objective of this Thesis was to improve the assessment of the impact of harbour emissions on the air quality over the harbours and surrounding urban areas, with a final goal of producing best-practices guidelines, supporting the decision-making process regarding port activities and air quality management.

To achieve this objective, the state-of-the-art in this field was firstly reviewed, identifying the main gaps in the current scientific knowledge. To support the numerical modelling studies performed, a high-resolution emission inventory was made, using Port of Leixões as a case-study. Harbour data, including ship and cargo handling equipment, were thoroughly compiled, allowing the identification of the main sources of emissions, and which pollutants were emitted in higher quantities by this harbour.

Having the detailed emission inventory, the C-PORT model (Isakov et al., 2017) was used (for the first time in European harbours) to simulate the impact of the maritime emissions, considering the most critical pollutants in terms of atmospheric concentration and population exposure over harbour region: NOx and PM10.

The last goal of this work was to study/identify/propose mitigation measures to solve the major air pollution problems originated in harbour areas, using for that numerical modelling and wind tunnel experiments. As an example, the Port of Aveiro was used as a case-study, due to increasing social and health concern over the nearby communities regarding the high levels of petroleum coke (petcoke) particles inside and around the residences. Such particles were coming from the outdoor storage of petcoke inside the harbour, while awaiting transportation to a nearby manufacturing plant. This case-study showcased the applicability of the proposed methodologies for different harbours and realities, to solve issues that are not exclusive to the studied harbours, quite the contrary: such problems are taking place in harbours across the globe.

1.3 Structure of the thesis

This Thesis is divided into 6 chapters, where each objective of the work is addressed. The Chapter 1 presents the framework and objectives.

Chapter 2 aims at reviewing the current knowledge on air quality over harbour areas through measured data analysis. Despite the impact of emissions from shipping activities on air quality, published data on this subject is very scarce. In this chapter, available information is reviewed focusing mainly on PM10, NO_2, SO_2 and PAH emissions.

Chapter 3 presents a highly detailed bottom-up emissions inventory for port activities with focus on case-study of Port of Leixões. This study presented a novel attempt to develop a detailed cargo handling equipment-related emissions inventory for a European harbour. For this purpose, the two most used methodologies by the scientific community and port authorities were applied and compared: EMEP/EEA and US/SCG. This comparison is particularly important to estimate the major

sources of uncertainty associated to this type of emission inventory data and to infer its potential impacts in the air quality modelling applications.

Chapter 4 focuses on the assessment of the relative contribution of different pollution sources to air quality near port areas, namely port activities, shipping emissions, roadway traffic and industry. Port of Leixões was also used as case-study for this work. To this end, a numerical modelling approach based on the web-based research screening tool C-PORT was used. A community-scale webtool developed by the US-EPA, to model emissions related to all port-area activities and predict concentrations of hazardous air pollutants at fine spatial scales in the near-source environment. The work presented in Chapter 4 was the first application of such community-scale webtool in the European context.

In chapter 5, mitigation measures were proposed and assessed to improve air quality in harbours and their surroundings. Pollutant dispersion is addressed, aiming at defining mitigation measures to control fugitive dust petcoke emissions and its impact on the communities in Port of Aveiro's neighbourhood.

Chapter 6 sums the major conclusions of the work and provide some insight into the future challenges and research efforts in this field.

The success and relevance of this PhD work is highlighted/confirmed in several important outcomes, namely the publication of 5 papers in international journals and its presentation in 6 international conferences. Furthermore, the administration of the Port of Leixões recognized the merit of this work, to the point of including some of this work's results in its sustainability report for the year 2019.

Chapter 2

2. Impact of harbour activities on local air quality: a review

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2.1 Introduction

Maritime transport has been growing due to the globalisation of manufacturing activities and the increase of international trade and tourism (Zhao et al., 2013), making harbours key contributors to the social and economic development worldwide (Agrawal H. et al., 2008). In coastal areas, there is a rising concern about the impact of maritime transport and related activities on local air quality. In cases where harbours are located near densely populated urban areas, emissions from ships may have a strong impact, affecting human health of coastal communities and the environment (Isakson et al., 2001; Sorte et al., 2018). The growth of ship movements and consequent release of air pollutants also called the attention to this emission source. SO_X, NOx, PM, CO, and PAHs are emitted to the atmosphere as a direct result of maritime activities. According to global annual estimates, around 70 % of the ships' global emissions are within 400 km of the coast, but they still contribute to the degradation of air quality in coastal areas and harbour cities (Monteiro et al., 2018; Ramacher et al., 2019; Viana et al., 2014).

Despite the progress achieved in the last decades regarding air pollution control owing to the application of strict measures to reduce emissions, several countries are still facing air pollution episodes with regular exceedances of the EU limits and WHO guidelines. In particular, the latest official air quality data released by the European Environment Agency (EEA) in 2018 indicate 19 % of PM10 concentrations above the EU daily limit value considering the reporting air quality monitoring stations in 10 of the 28 EU Member countries (EU-28); PM2.5 concentrations above the EU annual legal limit value were recorded at 5 % of the air quality stations in four Member countries and four other reporting countries (EEA, 2019c).

Furthermore, according to the latest urban air quality database published by the WHO, the great majority of cities worldwide are exceeding the WHO's Air Quality Guideline (AQG) levels for PM10 and PM2.5 (Salameh et al., 2015). The summary report of this database discusses the PM10 levels for available worldwide mega-cities for the last available year in the period 2011-2015. The available data show several mega-cities exceeding the WHO's AQG levels: Delhi recorded annual average concentrations of PM10 above 200 µg/m^3; the cities of Cairo and Dhaka reported PM10 concentrations above 150 µg/m^3, while the cities of Mumbai, Beijing and Kolkata reported PM10 concentrations above 100 µg/m^3 (WHO, 2016).

Strict regulations aiming to control and prevent air pollution from shipping transport were introduced in the Marine Pollution Convention (MARPOL) Annex VI by the International Maritime Organization

(IMO) and entered into force in 2005. Many countries have ratified this protocol to limit NO_X and SO_2 emissions from ships. Several coastal areas have been classified as Sulphur Emission Control Areas (SECA), namely the Baltic Sea, the North Sea, the English Channel and the coastal waters around the United States of America and Canada. Within SECA areas the sulphur content in marine fuels is limited and was set at 1.5 % until 2010, 1 % between 2010 and 2015, and 0.1 % from 2015 (Jonson et al., 2019; Maragkogianni et al., 2016). Moreover, the European Union has established a legal requirement limiting at 0.1 % the sulphur content in fuels used for ships at berth in harbours, implemented since 2010. International legislation to reduce shipping emissions worldwide is mainly focused on the use of low-sulphur content fuel (Contini et al., 2015; Ledoux et al., 2018; Schembari et al., 2012; Xu et al., 2018). Recently, some the Representative Concentration Pathway (RCP) scenarios have been proposed including alternative assumptions on pollution control, in an effort to better understand the role of air pollution control in terms of reference scenario development and the co-benefits from climate policies (Chuwah et al., 2013; Rao et al., 2013).

Air pollutant emissions in harbours come from different sources, from manoeuvring ships to the activity at the dock and at berthing ship. In addition, emissions are also generated while vessels are at berth since not all types of vessels switch off the main engines (Jahangiri et al., 2018; Nunes et al., 2017). Emissions due to harbour-related activities represent only a small fraction of the global emissions associated with shipping (Sorte et al., 2019). Additionally, to the emission sources, many harbours are situated in topographically complex terrain, with limited or inadequately atmospheric dispersion conditions. In addition, coastal sites display specific meteorological patterns with individual and complex characteristics, mainly due to the temporal and spatial scales of the meteorological circulations on those areas, like sea-breezes (Sorte et al., 2019). These specific meteorological patterns of coastal areas, such as land-sea breezes, have a high impact on dispersion, transformation, removal, and accumulation of air pollutants (Anjos and Lopes, 2019). The contribution of ship emissions to local air quality, with specific focus on atmospheric aerosol, has been investigated using numerical models (Gariazzo et al., 2007; Marmer et al., 2009), experimental campaigns (Ault et al., 2010; Contini et al., 2011; Jonson et al., 2015) or using receptor models based on the identification of chemical tracers associated with ship emissions (Contini et al., 2015; Pandolfi et al., 2011; Viana et al., 2008).

Ship emissions in harbours can have a significant impact on local air quality, population exposure and therefore human health in urban areas. Some studies found a high impact from local shipping emissions of nitrogen dioxide (NO_2) exposure in the harbour area of three Baltic Sea harbour cities (50–80 %). While the exposure in the closest urban areas was lower (3–14 % on average). Therefore, the impact of shipping emissions was more accentuated closer to the harbour areas and downwind (Ramacher et al., 2019). In some coastal areas, the contribution of shipping emissions to particulate matter pollution is of high importance, e.g., from 5 to 20 % (Dalsøren et al., 2009). Several studies found that ship emissions can have important effects on air quality and exposure of coastal communities in Europe, Asia, or North America, in locations with high levels of ship traffic, often

located near urban and industrial centres (Contini et al., 2011; Pandolfi et al., 2011; Ramacher et al., 2019; Viana et al., 2015).

Exposure to air pollution has been associated with severe health pathologies, including asthma, lung cancer, cardiovascular diseases, and heart attacks. Ship emissions have been associated with those pathologies (Quaranta et al., 2012; Sofiev et al., 2018; Tian et al., 2013; Yau et al., 2012). For instance, PM emissions from marine vessels activities have been related to an increase of hospitalizations due to cardiovascular episodes (Papaefthimiou et al., 2016). The impact of ship emissions on human health has been estimated in approximately 60,000 annual deaths at global scale, with severe impacts in coastal regions, mostly along European, East Asian, and South Asian coastal areas (Corbett et al., 2007). A more recent study shows that low-sulphur marine fuels will still account for 250,000 annual deaths in 2020 due to the increase in the transport by sea, despite the implemented low-sulphur regulations (Sofiev et al., 2018). Furthermore, population exposure to NO2 ship emissions was found to be consistently associated with total non-accidental mortality, and specific cardiovascular mortality in the harbour of Gothenburg in the Baltic Sea (Stockfelt et al., 2015). Ship exhaust is also one of the major sources of sulphur dioxide (SO_2) emissions in Hong Kong, contributing to 36 % of the ambient SO_2 concentrations, measured by equipment located close to the major shipping harbours (Kwai Chung and Tsing Yi) (Yau et al., 2012). Kilburn et al. (2012) show that emissions from ocean-going ships are associated with 519 premature deaths per year in the Pearl River Delta region, with the majority occurring in Hong Kong.

In summary, maritime transport can have a high contribution to air quality degradation of coastal areas, in terms of global and regional air pollution. Additionally, shipping activities can have a strong impact on local air quality of harbours. Therefore, the main goal of this paper is to thoroughly review and assess the status of air quality over harbour areas through measured data analysis. Despite their impact on air quality, air pollutant emissions data from shipping activities are very scarce in the available literature. To summarise the available data, this review focuses mainly on particulate matter emissions, as well as some gaseous pollutants, namely the NO_2, SO_2 and PAH. The paper is organised as follows: Section 2.2 describes the selected case studies; Section 2.3 presents the impact of harbour areas activities on ambient SO_2, NO_2, PM10, PM2.5 concentrations; Section 2.4 compiles the published results of ship emissions' contributions based on receptor modelling tools and Section 2.5 presents the summary and conclusions.

2.2 Air quality in harbours: case studies

A literature survey was conducted using cross-discipline platforms for research support in different areas - Science Direct, Scopus, and the Web of Science. A set of relevant keywords were used, namely ‘air quality’, ‘harbour activities’, ‘harbour activities and related atmospheric emissions’, ‘ports’, ‘source apportionment’, ’coastal areas’, ’air pollution in coastal areas/ cities’.

Figure 1 shows all the selected case studies focusing on air quality in harbour and/or harbour city areas.

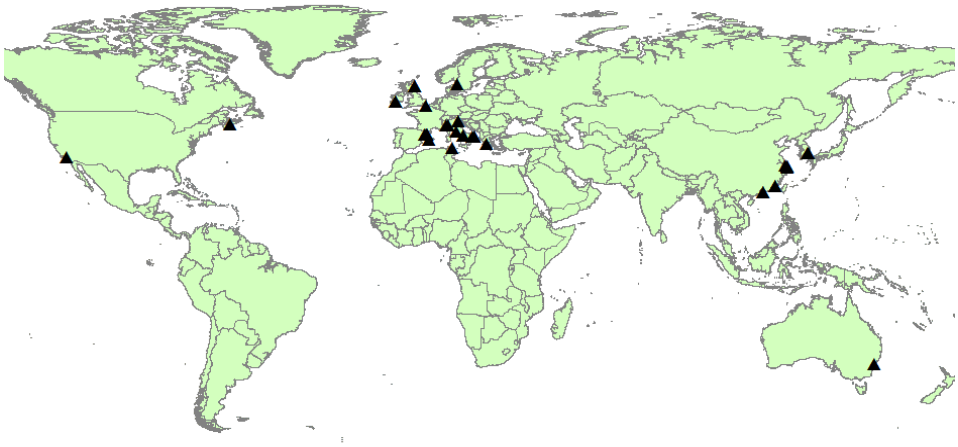

Figure 1. Location of the analysed published studies focusing on air quality over harbours.

Figure 1 presents the compiled case studies, in a total of 66, 9 % located in North America, 18 % in Asia, 2 % in Australia and the remaining 71 % case studies are in Europe. These 71 % case studies are mainly placed in the Mediterranean and North Baltic seas. To the best of the authors knowledge, there is any available study focusing on air quality over harbour areas in South America countries.

2.3 Air quality in harbours: current status

Table 1 identifies the analysed case studies focusing on air quality, summarizing the mean concentrations of the selected pollutants – NO_2, SO_2, PM10 and PM2.5 – followed by the corresponding measurement period.

Table 1. List of the selected case studies focusing on air quality over harbour areas, together with the mean concentrations of SO_2, NO_2, PM10 and PM2.5 recorded for each case study, during a specific period.

	Reference	Case study	Period	NO_2 ($\mu g/m^3$)	SO_2 ($\mu g/m^3$)	PM2.5 ($\mu g/m^3$)	PM10 ($\mu g/m^3$)
Europe	(Ledoux et al., 2018)	Calais harbour (France)	2014.1 - 2014.4	22.0	3.1	NA[a]	25.3
	(Isakson et al., 2001)	Gothenburg harbour (Sweden)	1998.6 - 1998.7	12.0	4.5	NA	NA
	(Amato et al., 2009)	Barcelona urban (Spain)	2005-2007	NA	NA	27.7	40.0
	(Pérez et al., 2016)		2011.02 - 2011.12	NA	NA	18.0	27.0
	(Schembari et al., 2012)	Barcelona harbour (Spain)	2009.8 - 2009.10	31.0	7.1	NA	NA
			2010.8 - 2010.10	27.6	4.2	NA	NA
	(Pérez et al., 2016)		2011.02 - 2011.12	NA	NA	18.0	35.0
	Schembari et al. (2012)	Palma de Mallorca urban (Spain)	2009.8 - 2009.10	53.9	14.1	NA	NA
			2010.8 - 2010.10	76.1	3.9	NA	NA
	(Alastuey et al., 2007)	Tarragona harbour (Spain)	2004.09 - 2005.09	NA	NA	NA	40.1
		Tarragona urban (Spain)	2001	NA	NA	NA	37.4
	(Pandolfi et al., 2011)	Algeciras urban-industrial (Spain)	2003 - 2007	NA	NA	24.2	37.2
	(Merico et al., 2016)	Brindisi harbour (Italy)	2014.7 - 2014.8	18.8	3.1	12.6	NA
	(Merico et al., 2019)	Bari harbour (Italy)	2016	NA	0.83	1.48	1.66
	(Donateo et al., 2014)	Brindisi harbour (Italy)	2012.06 - 2012.10	NA	NA	16.7	NA
	(Salameh et al., 2015)	Barcelona urban (Spain)	2011-2012	NA	NA	19	27
		Marseille urban (France)	2011-2012	NA	NA	17	31
		Genoa urban (Italy)	2011-2012	NA	NA	14	23
		Venice urban (Italy)	2011-2012	NA	NA	30	36
		Thessaloniki urban (Greece)	2011-2012	NA	NA	37	46
	(Gariazzo et al., 2007)	Taranto urban (Italy)	2004	44.6	6.0	NA	50.0
	(Schembari et al., 2012)	Savona harbour (Italy)	2009.8 - 2009.10	14.3	47.2	NA	NA
			2010.8 - 2010.10	14.9	1.6	NA	NA
		Civitavecchia harbour (Italy)	2009.8 - 2009.10	19.4	10.7	NA	NA
			2010.8 - 2010.10	21.2	1.6	NA	NA

[a] NA – Not available

Table 1(cont.). List of the selected case studies focusing on air quality over harbour areas, together with the mean concentrations of SO_2, NO_2, PM10 and PM2.5 recorded for each case study, during a specific period.

	Reference	Case study	Period	NO_2 (µg/m^3)	SO_2 (µg/m^3)	PM2.5 (µg/m^3)	PM10 (µg/m^3)
Europe	(Prati et al., 2015)	Naples urban (Italy)	2012.4	44.1	5.7	NA[a]	27.1
			2012.11	38.8	10.6	NA	31.8
	(Bove et al., 2014)	Genoa urban (Italy)	2011.5 - 2011.10	NA	NA	12.9	NA
	(Murena et al., 2018)	Naples urban (Italy)	2016	56.2	NA	NA	NA
	Contini et al. (2011)	Venice Lagoon harbour (Italy)	2007.3 - 2007.11	NA	NA	17.0	29.1
	(Merico et al., 2017)	Venice Lagoon harbour (Italy)	2002.3 - 2002.4	NA	NA	NA	62
	(Manousakas et al., 2017)	Venice urban (Italy)	2012	NA	NA	12.4	NA
	(Healy et al., 2010)	Cork harbour (Ireland)	2008.5 - 2008.8	NA	NA	9.7	NA
	(Hellebust et al., 2010)		2007.5 - 2008.4	NA	NA	2.8	4.6
	(Marr et al., 2007)	Aberdeen urban (Scotland)	2003.5 - 2003.8	80.8	NA	NA	NA
			2003.11 - 2004.5	107.2	NA	NA	NA
		Aberdeen harbour (Scotland)	2003.5 - 2003.8	56.4	NA	NA	NA
			2003.11 - 2004.5	92.1	NA	NA	NA
	(Gregoris et al., 2016)	Venice harbour (Italy)	2009	NA	NA	19.5	NA
		Venice harbour (Italy)	2012	NA	NA	12.4	NA
Africa	(Schembari et al., 2012)	Tunis harbour (Tunisia)	2009	20.5	7.6	NA	NA
			2010	27.1	9.2	NA	NA
Asia	(Zhao et al., 2013)	Shanghai harbour (China)	2010	63.7	29.4	62.6	NA
	(Xu et al., 2018)	Xiamen harbour (China)	2015.4 - 2016.1	NA	NA	51.9	NA
		Xiamen urban (China)	2015.4 - 2016.1	NA	NA	46.4	NA
	(Mamoudou et al., 2018)	Yangshan harbour (China)	2016	NA	NA	44.0	NA
	(Jeong et al., 2017)	Busan urban (Korea)	2013	NA	NA	26.1	NA
	(Lang et al., 2017)	Qinhuangdao urban (China)	2014.4 – 2015.1	NA	NA	70.1	NA
	(Yau et al., 2013)	Tsing Yi urban (Hong Kong)	2009.8 - 2009.11	NA	NA	25.2	NA
	Yau et al. (2013)	Tsing Yi urban (Hong Kong)	2010.1 - 2010.3	NA	NA	35.5	NA
	Tao et al. (2017)	Zhuhai urban (China)	2014 - 2015	NA	NA	45.0	NA

[a] NA – Not available

Table 1(cont.). List of the selected case studies focusing on air quality over harbour areas, together with the mean concentrations of SO_2, NO_2, PM10 and PM2.5 recorded for each case study, during a specific period.

	Reference	Case study	Period	NO_2 (µg/m^3)	SO_2 (µg/m^3)	PM2.5 (µg/m^3)	PM10 (µg/m^3)
Oceania	(Broome et al., 2016)	Sydney urban (Australia)	2010 - 2011	NA[a]	NA	7	16.4
North America	(Moore et al., 2009)	San Pedro harbour (California)	2007	NA	NA	13.8	43.1
	Moore et al. (2009)	San Pedro urban (California)	2007	NA	NA	16.3	35.6
	(Tao et al., 2013)	Oakland harbour (California)	2008.7-2009.6	NA	NA	7.1	NA

[a] NA – Not available

The average ambient NO_2 concentrations associated with shipping levels ranges between 12 µg/m^3 and 107 µg/m^3, depending on the measurement period, with the highest values located in Scotland, Spain, and China. Several exceedances of the annual limit value of NO_2 were recorded in urban areas close to population clusters, such as Taranto and Naples. In some studies, the air pollution levels were lower around the harbour, when compared with the surrounding urban area. For instance, the case study of Aberdeen (Scotland) showed lower concentrations of NO_2 around the harbour than the concentrations registered in the city centre (Marr et al., 2007). The authors of the study identified as probable cause the height of the emission source, considering the top of the ferry hoppers and the oil service ships. This is enough to spread the hot emissions in a very effective way, not detected locally at a ground level, but affecting further the neighbour urban area (Marr et al., 2007). Similarly, in Gothenburg (Sweden), during summertime, the averaged concentrations measured at an averaged distance of 800 m from the ships of NO_2, in-line with the ship's plume, indicate an average concentration of NO_2 12 µg/m^3 above the urban background levels, while for SO_2 this value was 4.5 µg/m^3, for background levels of 11.3 and 1.6 µg/m^3, respectively (Isakson et al., 2001).

The relatively low values of SO_2 may be due to the efforts of the EU and IMO to restrict ship emissions. 45 % of the harbours reviewed were located under emission control areas, mainly across the coast of Europe, United States of America, and European North Sea. The SO_2 concentrations measured in the different case studies range from 0.83 to 47.2 µg/m^3, considering the distinct sampling periods for different cases. All studies carried in European countries reported a low SO_2 concentration in conjunction with the impact of the EU directive 2005/33/EC, which regulates the SO_2 ship emissions in EU harbours from January 2010 on. The concentration of SO_2 decreased significantly from 2009 to 2010 in EU harbours: 41 % in Barcelona, 72 % in Palma de Mallorca, 97 % in Savona and 85 % in Civitavecchia (Schembari et al., 2012). Moreover, there is also evidence in other European harbours that this strategy contributed to lower SO_2 concentrations, namely at Calais, France (Ledoux et al., 2018), Brindisi, Italy (Donateo et al., 2014; Merico et al., 2016), and Bari, Italy (Merico et al., 2019). Mamoudou et al. (2018) show evidence of a noticeable improvement

of air quality in Yangshan harbour due to the control measures of ship emissions employed in the Yangtze River Delta region (Mamoudou et al., 2018). Some studies showed also that low-sulphur fuels could reduce the shipping contribution to PM2.5 concentration in harbour areas (Contini et al., 2015; Liang et al., 2016), but with limited effects on metals and PAHs concentrations (Gregoris et al., 2016). The study focuses on Brindisi (Italy) (Donateo et al., 2014) indicates the impact on SO_2 concentrations of manoeuvring during the ship's arrival and/or departure. On the other hand, the hoteling phase had limited effects on SO_2 concentration, probably due to the mandatory use of low-sulphur content fuels in European harbours, together with the differences between the auxiliary and main motor emissions, as well as the different engine loads (Merico et al., 2016). No reduction was detected in the non-EU harbour, for instance in cases of study of Tunis and Shanghai (Schembari et al., 2012; Zhao et al., 2013).

Some of studies have also revealed that ship emissions contribute more to fine particles, and especially to ultrafine particles, than to coarse aerosols (Saxe and Larsen, 2004; Viana et al., 2009). Primary particles emitted by ships are predominantly in the sub-micron size fraction, which may support these results (Healy et al., 2009). Ship emissions have been identified as contributors to an increase in particle concentrations and are thus dominated as ultrafine particles (Reche et al., 2011). Murena et al. (2018) show that in the coast of China the PM2.5 concentration at Xiamen harbour differed by less than 20 % from values reported from other harbours such as Shanghai (62.6 µg/m^3, Zhao et al., 2013). Besides that, the $PM_{2.5}$ concentration at this harbour was more than twice the concentration found in other harbours such as Busan, Korea (Jeong et al., 2017), Brindisi, Italy (Cesari et al., 2014) and Barcelona, Spain (Amato et al., 2009). In Shanghai (Zhao et al., 2013), Xiamen (Xu et al., 2018) and Yangshan (Mamoudou et al., 2018) studies point out that ship traffic has a non-negligible impact on primary particles in harbour and surrounding land areas.

Despite being the most studied pollutants in the literature, PM, NO_2 and SO_2 are not the only pollutants affecting air quality over harbour areas. An array of other compounds can be found at significant concentrations around harbours. For instance, Vanadium (V) and nickel (Ni), as well as BC (black carbon) and polycyclic aromatic hydrocarbons are typically emitted by shipping activities, and they are hazardous to human health. As an example, the urban area of the harbour of Venice (Sacca San Biagio), has registered annual average values of 30.7 ng/m^3 in 2009 and 6.3 ng/m^3 in 2012 for gas and particulate PAHs together (Gregoris et al., 2016). In comparison, the monitored air quality levels of the city showed values of 5.4 ng/m^3 and 2.6 ng/m^3, in 2009 and 2012, respectively. The same effect was observed in Brindisi by applying the same double-sampling method (Donateo et al., 2014). Air coming from the harbour/industrial sector was richer in PAHs (5.34 ng/m^3) than air sampled from all directions (3.89 ng/m^3). This result is like the findings in other harbour cities such as Venice (Contini et al., 2011).

2.4 Contribution of harbour emissions to PM

A set of studies addressed the contribution of harbour activities to PM values using source apportionment techniques. These studies focused mainly on the Mediterranean, Asia, and North America city-harbours. These source apportionment studies presented different approaches, namely positive matrix factorization (PMF), main component analysis/absolute main component score (PCA/APCS), chemical mass balance (CMB), multiple linear regression (MLR), and Vanadiun concentration (V).

Table 2 summarizes the contribution of shipping activities to PM concentrations found in each of those studies, with the respective sampling period, and method used. Other emission sectors (e.g., traffic, industry, and natural sources) were also identified and quantified (available on Table S 1 of the Appendix A).

PMF is the most used method for source-apportionment among the selected case studies. In the case study of Busan (Korea), it is possible to see the magnitude of source contribution estimates to PM levels, which differ significantly among PMF, CMB and PCA/APCS models (Jeong et al., 2017). Newly inter-comparison studies, which use different data sets (number of chemical species) and receptor models (PMF, CMB, PCA), suggest that the stability and robustness of the results depend on receptor models, bringing significant differences in the magnitude of the contribution-source (Belis et al., 2014; Cesari et al., 2016; Liang et al., 2018; Viana et al., 2009). Recently, Mamoudou et al. (2018) showed that the V method significantly underestimated (in approximately 1.84 μg/m^3) the contribution of shipping emissions to PM2.5 concentrations at Yangshan harbour, when compared to the PMF method.

The maximum PM2.5 contribution estimated by PMF for ship emissions was 26.1 % in Xiamen, followed by Kwai Chung urban (19 %) and Zhuhai (18 %). For PM10, the major contributors were the harbours of Tarragona (16 %), followed by Patras (15 %) and Barcelona (8 %). The ship emission contributions to PM2.5 differed significantly between European, Asian, and North American harbour cities, ranging from 2-14 %, 1–25 % and 3-29 % of PM2.5, respectively. On the other hand, the ship emission contributions to PM10 in Europe ranged between 3-16 %. In overall, a greater ship contribution was seen in East Asian harbour cities compared to Europe, because of the presence of major hub harbours in that part of the world. The contribution of ships clearly decreased at Yangshan harbour due to the control measures of shipping emissions employed in the Yangtze River Delta, while diesel engine exhaust from ships, as well as diesel vehicles, were still a significant source in this harbour and nearshore areas (Mamoudou et al., 2018).

The source contribution to PM in urban areas is different from those in harbour cities. In Xiamen's harbour, the industry and ship emissions contributions for PM2.5 at harbour/industrial areas were close to double (26 %) compared to the urban ones (13 %) and were primarily caused by refined industrial activities and ship emissions (Xu et al., 2018). The same behaviour was registered in Barcelona's harbour where the contribution of the shipping emissions to PM2.5 (14 %) was approximately double of those at the urban area (6 %). The high concentration of mineral dust measured at the harbour area come from the construction in a new harbour terminal, together with the manoeuvring of bulk solids and the re-suspension of the street dust by light and heavy vehicles. The contribution of other sources of emissions – traffic, industrial and natural – vary according to the type of harbour, namely with the industrial or urban surrounding.

Table 2 List of case studies using source-apportionment methods to estimate shipping contribution to PM values.

	Reference	Location	Period	Method	PM	Shipping (%)
Europe	(Saraga et al., 2019)	Thessaloniki harbour (Greece)	2011.6 – 2012.5	PMF	PM2.5	13
		Thessaloniki harbour (Greece)	NA	PMF	PM2.5	9
	(Scerri et al., 2018)	Malta urban (Republic of Malta)	2016	PMF	PM2.5	5
	(Manousakas et al., 2017)	Patras harbour (Greece)	2011	PMF	PM2.5	10
	Manousakas et al (2017)	Patras harbour (Greece)	2011	PMF	PM10	15
	Cesari et al. (2014)	Brindisi harbour (Italy)	2012.6 - 2012.10	PMF	PM2.5	15.3*
	(Bove et al., 2014)	Genoa urban (Italy)	2011.5 - 2011.10	PMF	PM2.5	11
	(Merico et al., 2017)	Brindisi urban (Italy)	2012	V	PM2.5	2.8
	(Gregoris et al., 2016)	Venice harbour (Italy)	2007 - 2013	PMF	PM10	2.5
	Gregoris et al. (2016)	Venice harbour (Italy)	2007 - 2013	PMF	PM2.5	3.3
	Merico et al (2017)	Rijeka harbour (Croatia)	2014	PMF	PM2.5	10.5
	Merico et al. (2017)	Rijeka urban (Croatia)	2012	V	PM2.5	1.1
	Merico et al. (2017)	Rijeka harbour (Croatia)	2013-2014	V	PM2.5	0.5
	Pérez et al. (2016)	Barcelona harbour (Spain)	2011.02 - 2011.12	PMF	PM2.5	14
			2011.02 - 2011.12	PMF	PM10	8
		Barcelona urban (Spain)	2011.02 - 2011.12	PMF	PM10	4
			2011.02 - 2011.12	PMF	PM2.5	6
	(Amato et al., 2009)		2003 -2007	PMF	PM2.5	6-7
			2003 -2007	PMF	PM10	5
	Pandolfi et al. (2011)	Gibraltar Strait urban (Spain)	2003 -2007	PMF	PM2.5	5-10
			2003 -2007	PMF	PM10	3-7
	Viana et al. (2009)	Melilla urban (Spain)	2007	PMF	PM2.5	14
			2007	PMF	PM10	2-4
	(Alastuey et al., 2007)	Tarragona harbour (Spain)	2004.09 - 2005.09	FA	PM10	16
	(Healy et al., 2010)	Cork harbour (Ireland)	2008.5 - 2008.8	PMF	PM2.5	2
	(Hellebust et al., 2010)		2007.5 - 2008.4	MLR	PM10	< 5*

* Mixed industrial source and shipping

Table 2(cont.) List of case studies using source-apportionment methods to estimate shipping contribution to PM values.

	Reference	Location	Period	Method	PM	Shipping (%)
Asia	Xu et al. (2018)	Xiamen harbour - industrial (China)	2015.4 - 2016.1	PMF	PM2.5	26*
		Xiamen urban (China)	2015.4 - 2016.1	PMF	PM2.5	13*
	Mamoudou et al. (2018)	Yangshan Island harbour (China)	2016	PMF	PM2.5	2.4
	Mamoudou et al. (2018)	Yangshan Island harbour (China)	2016	V	PM2.5	0.23
	Tao et al. (2017)	Zhuhai urban (China)	2014 -2015	PMF	PM2.5	18
	Zhao et al. (2013)	Shanghai harbour (China)	2011	V	PM2.5	4.23
	(Jeong et al., 2017)	Busan urban (Korea)	2013	PMF	PM2.5	7
			2013	CMB	PM2.5	1
			2013	PCA/ APCS	PM2.5	5
	(Yau et al., 2013)	Kwai Chung and Tsing Yi urban (Hong Kong)	2009.8 - 2010.3	PMF	PM2.5	19
			2009.8 - 2010.3	PCA/ APCS	PM2.5	25
North America	(Gibson et al., 2013)	Halifax urban (Canada)	2011.7 - 2011.8	PMF	PM2.5	3
	(Minguillón et al., 2008)	Angeles Long Beach harbour (California)	2007	CMB	PM2.5	< 5
	(Kuwayama et al., 2013)	Oakland harbour (California)	2010.3	PMF	EC	12
	Kuwayama et al. (2013)	Oakland harbour (California)	2010.4 – 2010.5	PMF	EC	29.2
	(Agrawal et al., 2009)	Los Angeles harbour (California	2004	V	PM2.5	8.8

* Mixed industrial source and shipping

A higher traffic contribution for emissions of PM was registered in Europe compared to Asia, with contributions ranging between 16 - 45 % and 7 - 22 %, respectively. The maximum contribution from traffic to PM2.5 emissions was estimated by PMF to be 45 % in Thessaloniki, followed by Barcelona and Patras harbours (36 and 34 % respectively). For PM10, traffic was the main contributor, once again for urban-surrounded harbours; in this case Barcelona and Tarragona, where traffic accounted for 40 and 34 % of PM10 emissions (Alastuey et al., 2007; Manousakas et al., 2017; Pérez et al., 2016; Saraga et al., 2019). The elevated contribution of traffic at the Thessaloniki and Tarragona harbours was attributed to the high number of vehicles, trucks circulation, and the resuspension during dock's loading and unloading in the harbour area (Saraga et al., 2019). However, the road traffic's contribution to the PM2.5 emissions in Barcelona is very similar between the harbour

surroundings and the urban area. This is probably due to the localization of the harbour, with a similar high impact of vehicle traffic in both monitoring sites, both situated close to the main roads in and out of the city (Pérez et al., 2016). Contributions of PM components (Elemental Carbon (EC)) attributed to harbour truck activities decreased from 56 to 23 %, due to the control program, which meets the targets defined in the Emission Reduction Plan for Ports and Goods Movement in California (Kuwayama et al., 2013). After the implementation of the control program, harbour trucks and ships accounted for approximately 23 and 29 % of the ambient EC concentrations at the harbour, respectively.

Industry accounted for 16 % of PM2.5 emissions in Brindisi, followed by 14 and 12 % in Barcelona and Tarragona harbours, respectively. Regarding PM10, once again industry was a major contributor in the Tarragona (12 %) and Barcelona (8 %) harbours. Industry contribution for PM emissions is higher in Asian harbour cities than in European ones, with a maximum of 88 % registered in Yangshan Island, China (Mamoudou et al., 2018). This may be due to the higher industrialization in China or Korea, compared to Europe. Another explanation would be the difference in environmental legislation between Europe and Asia, namely regarding the limit values of PM concentrations.

Natural sources of emissions include marine sea salt, African dust, soil resuspension, sea spray and biomass burning/ fires (not domestic heating). The maximum PM2.5 contribution estimated by PMF from natural sources was 27 % of PM2.5 in the Gibraltar harbour, followed by Busan (18 %) and Barcelona urban (17 %) (Jeong et al., 2017; Pandolfi et al., 2011; Pérez et al., 2016). This is probably due to the limited effect of industries in urban stations since large industrial plants are usually placed in the outskirts of urban areas. Moreover, less populated countries like Gibraltar show less traffic and/or less industrial activity, meaning that a higher share of emissions would be attributed to natural sources (Pandolfi et al., 2011). Contribution from natural sources was comparable between European and Asian harbour cities, ranging from 2-27 % and 2-18 % of PM2.5, respectively.

2.5 Conclusions

The literature review performed in this paper aims to summarize the current knowledge on air quality status locally over harbour areas, focusing on several worldwide harbours. The increase, in the last decade, of available studies evaluating the impact of ship emissions denotes the relevance of this activity sector and its impacts on air quality and consequent human exposure. All the selected studies in this literature review agree on the relevant contribution of shipping and harbour activities in terms of atmospheric emissions and related concentrations of the main critical pollutants, namely PM10, PM2.5, NO_2 and SO_2.

The selected studies indicate a large spatial variability of particulate matter and gaseous concentrations over distinct countries. For instance, the measured NO_2 concentrations ranged from 12 to 107 µg/m^3, while the measured PM10 concentrations varied between 2 and 62 µg/m^3. The maximum concentrations for all the considered pollutants were found in European harbour areas (e.g., maximum concentrations of 107, 47, 37 and 62 µg/m^3 for NO_2, SO_2 (before 2010), PM2.5 and

PM10, respectively), which is mainly due to the high number of available measurements, and not necessarily an indicator of the occurrence of more air pollution episodes over the European harbours. The largest concentrations of PM2.5 were found in Asia varying from 25 to 70 µg/m^3. In addition, in some harbour areas, namely Thessaloniki, Patras, Genoa, Barcelona, Gibraltar and Tarragona, the shipping contribution to PM2.5 concentrations were quantified using source apportionment techniques, indicating a contribution from 10 up to 14 %.

This review highlights the relevance of the maritime transport sector on air pollutants emissions and its impact on air quality and human exposure, over harbour urban areas. Current mitigation strategies in the European territory have proved their efficiency; with decreases of SO_2 concentrations in several harbours (the posterior reductions on the secondary PM are not totally quantified).

The outcomes of this literature review emphasize the further implementation of currently available measures, together with the implementation of new countermeasures especially focused on the emissions of primary particles. We believe that soon other studies will be made available, with new air quality data, from monitoring and/or modelling exercises, covering more air pollutants, and improving our current knowledge in this research field.

Chapter 3

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3. Emission inventory for harbour-related activities: comparison of two distinct bottom-up methodologies

3.1. Introduction

The accelerated growth of the global economy have several negative environmental impacts, such as an increase of air pollutants emissions from distinct sources (e.g., road traffic, shipping sector, aviation sector), which have been contributing to both climate change and air pollution (Crilley et al., 2017; Jacob and Winner, 2009; Karagulian et al., 2015; Monteiro et al., 2018b; Park et al., 2020; Ravindra et al., 2019). Maritime transport sector delivers an important contribution to the worldwide trade and welfare, representing more than 90 % of the international trade in 2019 (ICS, 2014), with an increasing economic relevance of harbours (Puig and Darbra, 2019; Rodrigues et al., 2021; UNCTAD, 2019). Although this important contribution, atmospheric emissions from shipping constitute an important share of the total emissions associated with the global trade (Aulinger et al., 2016; Goldsworthy et al., 2019; Jonson et al., 2015; Russo et al., 2019). Despite being the most energy-efficient way to transport large volumes of cargo over long distances (Jonson et al., 2015; Lindstad and Eskeland, 2015), ships lead to significant levels of emissions of air pollutants into the atmosphere, such as NO_X, SO_X, PM, CO, Carbon Dioxide (CO_2), and Volatile Organic Compounds (VOCs) (Chen et al., 2018; Merico et al., 2016; Sorte et al., 2020). All these atmospheric pollutants have severe impacts (both direct and indirect) on human health and ecosystems (Broome et al., 2016; Graber et al., 2019; Rajagopalan et al., 2018). In particular, CO_2 is of great concern due to its role as a Greenhouse Gas (GHG) and its contribution to climate change (Georgatzi et al., 2020; Martínez-Moya et al., 2019; Ortega Piris et al., 2018; Tsai et al., 2018).
The quantification of these emissions, through the development of emission inventories, is the first step to improve our knowledge about the impacts of harbour and shipping activities (Cesari et al. 2014; Viana et al. 2014; Aksoyoglu et al. 2016; Zhang et al. 2017; Monteiro et al. 2018a; Russo et al. 2018; Goldsworthy et al. 2019). Emission inventories are an important and useful tool for both source contribution assessment as well as input for air quality modelling (Ferreira et al., 2013; Jalkanen et al., 2009; Russo et al., 2018). These tools usually distinguish emission sources into mobile and fix sources. In the case of harbour-related activities, mobile sources are distinguished between on-road mobile sources (e.g. light vehicles, and heavy-duty vehicles) and off-road sources (e.g. cargo handling equipment, heavy-duty trucks, locomotives, and ships) (Sorte et al., 2019). In

particular, mobile emission sources related with port-activities include maritime ships and land-based emission sources (e.g. dredges, tugs and tows, as well as other ships operating within the port area); while, fix emission sources include cargo handling equipment (CHE) (Sorte et al., 2018). Cargo handling equipment refer to cranes, container handlers, forklifts, reach stackers, shovel excavators, and on-road trucks (SCG, 2014).

Due to manoeuvring and hotelling phases, air pollution from ships can be very significant in harbour areas (e.g., Murena et al., 2018). In harbour regions, ships may contribute with 55 % to 77 % of the total atmospheric emissions in those regions (Tzannatos, 2010). The use of auxiliary diesel engines during the hotelling stage contributes mainly to exhaust emissions within the harbour area (Deniz and Kilic, 2010; Tzannatos, 2010).

In the last decades, air pollution associated with maritime transport, in particular shipping emissions, has been addressed by the IMO under the MARPOL. In 2010, IMO has established Emission Control Areas (ECA) and has set limits for SO_X and NO_X emissions (IMO, 2010). In Europe, the maximum sulphur content of the marine fuel used by ships operating in the SECA – the Baltic Sea, the English Channel, and the North Sea – was restricted to 1.0 % in July 2010 and further reduced to 0.1 % in January 2015. Until 2019, the sulphur content of oil to be used in EU ports and Emission Control Areas (ECA) is of 0.1 %, while it is of 1.5 % in non-ECA, EU waters (EU Directive 2005/33/CE), and of 3.5 % in all other conditions. Starting in 2020, the global limits converged to 0.1 % fuels in ECA and port environments and to 0.5 % in all other conditions (Gobbi et al., 2020; Sorte et al., 2020). Regarding NO_X emissions, Annex VI contains a three tier (Tier I, II and III) methodology that indicates the admissible emissions of total NO_X depending on the engine speed. The Tier III, currently in force, applies to new ships (built after January 2016) operating in North American and U.S. Caribbean NO_X emissions control areas (NECA) (IMO, 2016). A harbour emission inventory is essential for harbour authorities to quantify the impacts of the growing maritime activity, and to develop mitigation plans to face their environmental impacts (EPA, 2009; SCG, 2019). Without a specific-oriented emission inventory for harbour areas, the management entities face strong difficulties to identify and assess opportunities for the implementation of emission reduction measures and to quantify the effectiveness of those measures over time, through the quantification of emission reductions (Fu et al., 2017). Bottom-up methodologies were adopted by several organizations to develop emission inventories, such as US/SCG (US Starcrest Consulting Group) and EMEP/EEA (European Monitoring and Evaluation Program/European Environmental Agency) (EEA, 2019a, 2019b; EPA, 2009; SCG, 2015a, 2019a, 2019b).

The main goal of the current work was to develop a highly detailed bottom-up emissions inventory for port activities over the Port of Leixões (Portugal) case-study, accounting not only for maritime transport activities, but also for cargo handling equipment, which is a novelty for European Ports. For this purpose, the two most used methodologies by the scientific community and port authorities were applied and compared: EMEP/EEA and US/SCG. This comparison is particularly important to estimate the major sources of uncertainty associated to this type of emission inventory data and to infer its potential impacts in the air quality modelling applications.

The paper is organized as follow: Section 3.2 briefly describes Port of Leixões' activity and location. The two bottom-up methodologies applied (EMEP/EEA and US/SCG) are described in Section 3.3. In Section 3.4, the main results obtained by the two methodologies are presented and discussed. Finally, Section 3.5 presents the main conclusions and some final remarks.

3.2. The case study of Port of Leixões

The Port of Leixões, an artificial harbour in the vicinity of Porto urban area, in northern Portugal, was selected as a case study for this work. The harbour is bounded by two medium-size cities, the cities of Leça da Palmeira and Matosinhos. The Port of Leixões (41º11'30.73 N, 8º40'56.86 W) is one of the most competitive and versatile multi-purpose ports in the country. It is the largest port in the North region of Portugal, representing 25 % of the Portuguese trade. The Leixões harbour is formed by two curved breakwaters that are 5,240 feet (1,597 m) and 3,756 feet (1,145 m) long. Leixões is mainly an export port, serving virtually all types of ships and cargo, as well as cruises. Within 9 km of the port are the towns of Matosinhos and Leça da Palmeira, with 63/6 km^2 and around 176,000/18,502 inhabitants, respectively. This Port is endowed with quays (a conventional quay for general cargo and solid bulk and a quay for liquid bulk), terminals (terminals for tankers, containers, multi-use, and cruise liners), a yachting marina, a fishing harbour, and specialised facilities (such as for deposits and warehouses).

The Port includes 14 operational terminals, 5 km of quays, 55 ha of embankments and 120 ha of wet area. The Port of Leixões has registered in 2016 a total of 2 717 calls from ships, which corresponded to 32 854 516 Gross Tonnage (GT) and 18 314 852 tonnes of commodities. The container ships (42 %) were the most frequent, followed by tugs (20 %), general cargo (16 %), liquid bulk ships (9 %), dry bulk carriers (5 %), passenger (4 %), ro-ro cargo (2 %) and fishing (2 %), and finally other ships (1 %). There is a total of 68 CHE in Port of Leixões, including 41 % forklifts, 31 % shovel excavators, 15 % reach stackers, 7 % cranes, 3 % loaders and 3 % empty handlers. Figure 2 shows the geographical boundaries of the case study, composed by an 8 km diameter circle around the Port of Leixões, to include the surrounding cities of Matosinhos and Leça da Palmeira (Port-land direction) and the anchorage point of ships while waiting for berth call (Port-sea direction).

Figure 2. Study area - location of the harbour terminals (41º11'30.73 N, 8º40'56.86 W) and anchorage (41° 9'27.98 N, 8°44'33.71 W) of Port of Leixões.

3.3. Emissions' estimation methodologies

Harbour areas exhibit mainly two different types of emissions sources: maritime emissions, which are mainly associated with the diesel engines of the ships, and the land-based emission sources, which include cargo-handling equipment. In this paper, both types of emissions are estimated (Section 3.3.1 and Section 3.3.2) using different available calculation methods, namely the methodologies applied by the i) European Monitoring and Evaluation Programme from the European Environment Agency (EEA, 2019b, 2019a), and ii) US Starcrest Consulting Group report (SCG, 2019). The US/EPA methodology was recently revisited by the Starcrest Consulting Group report, which proposes updated methodologies based on the progresses achieved since the last report published in 2009 (EPA, 2009; SCG, 2019).

In both cases, the emission inventories include as atmospheric pollutants the NO_X, PM10, PM2.5, SO_X, CO, VOC, black carbon (BC), and as GHG the CO_2, N_2O, methane (CH_4).

3.3.1 Cargo handling equipment emissions

CHE includes a diverse group of equipment according to the cargo handled and the task performed, suited for handling with cargo arriving and/or departing by ship, truck, or train, which can include liquid, break bulk and dry bulk, and containers. The majority of CHE includes off-road equipment, which is not allowed to operate on public roads.

In this study, the CHE emissions inventory considers mainly the equipment available at bulk terminals and container terminals, including both CHE powered by diesel and gasoline. The data provided by the port concessionaires includes equipment type, the engine's rate power, the model and year of the equipment, engine fuel type, together with data on the cumulative running hours, and the operating hours (on an annual basis).

The methodology applied to estimate the CHE emissions considers the latest version of the EMEP/EEA (EEA, 2019b), and US/SCG (SCG, 2019) guidelines, represented by the Equation 1 and Equation 2, respectively.

$$E_i = N \times HRS \times P \times (1 + DFA) \times LFA \times EF_{Base} \quad (1)$$

$$E_i = N \times HRS \times P \times LF_i \times EF_i \times FCF \quad (2)$$

$$EF_i = ZH + (DR \times CH) \quad (3)$$

Where,

E indicates the emissions of a specific pollutant i (tonne/year),
N is the number of CHE type (units),
HRS is the annual hours of use (h/year),
P is the power rate of the engine (kW),
DFA is the deterioration factor adjustment,
LFA is the load factor adjustment,
LF is the load factor (%),
EF_{Base} is the base emission factor (g/kWh),
EF is the emission factor, grams of pollutant per unit of work (g/kWh),
FCF is the fuel correction factor (dimensionless),
ZH is the zero-hour emission rate (g/kWh),
DR is the deterioration rate (g/kWh^2) and
CH is the cumulative hours account for the number of hours in which the CHE engine has been turned on (h).

One of the main differences between the two methodologies is related to the fact that the EMEP/EEA methodology estimates emissions based on the deterioration factors adjustment (DFA), while the US/SCG methodology estimates based on the Fuel Correction Factor (FCF). The deterioration factor adjustment of EMEP/EEA methodology depends on the CHE' power range and on the technology (for more details see the Appendix B - App. 1.3). On the other hand, the fuel correction factor of US/SCG methodology is used to adjust EF associated with the fuel used, reflecting changes in fuel properties over time (for more details see Appendix B in the Table S 4). Engine load factors depend

on the average operational level of an engine in each application as a fraction or percentage of the engine manufacturer's maximum rated horsepower. Since emissions are directly proportional to engine horsepower, load factors are used in the inventory calculations to adjust the maximum rated horsepower to normal operating levels. Additional details of load factors for both methodologies are included in Table S 2 and Table S 3 of the Appendix B App. 1.1.

Additionally, emission factors are also a source of uncertainty when applying different methodologies. EMEP/EEA methodology uses fixed EF established according to technology level, engine size class (according to EU emission directives for non-road mobile machinery) (EEA, 2019b); US/SCG methodology uses EF computed according to Equation 3, where ZH and DR are listed according to technology level and engine size class (SCG, 2019). Additional details of EF for both methodologies are included in Table S 7 and Table S (Appendix B).

3.3.2 Shipping emissions

Regarding shipping emissions within the harbour area, for both methodologies, the estimation of emissions was based in distinct parameters such as the ship movement information, the activity data, and the emission factors per ship activity (e.g., manoeuvring, hotelling at anchorage or hotelling at berth).

Ships have three main sources of emissions on-board: the main engines, auxiliary engines, and boilers. Ships operate in two different operational modes - manoeuvring and hotelling. Manoeuvring mode includes the segment during harbour transit and docking; hotelling comprises the hotelling at anchorage while waiting for berth call, and the hotelling at berth during loading/unloading (Song and Shon, 2014). A third operational mode exists but was left out of the scope of this work – at sea moving. The reason for this choice was that the focus of this work was put in the immediate urban vicinity of the Port, to understand and highlight the relevance of quantifying port-related emissions and the impact they may present to the living of those urban communities.

The majority of the engines are turned on whenever the ship is in motion (transiting at any speed and manoeuvring), and are normally turned off during hotelling operations, while the auxiliary engines are turned on during all the stages (Song and Shon, 2014). During hotelling, the auxiliary engines are responsible to keep the same power and any refrigerated containers cold while the ship is being worked.

In this work, both methodologies were applied to Port of Leixões using the ship activity-based methodology. All the needed input data were provided by the administration of the Port of Leixões and include data on ship movement data, as well as technical information on the engine size, power installed, and technology, or fuel use and time spent in each activity – manoeuvring, hotelling at berth and hotelling at anchorage. Some ship characteristics were confirmed on the SeaWeb website (SeaWeb, 2018).

Equation 4 and 5 presents the methodology usually named as Tier 3 (which is ship activity-based) in the EMEP/EEA (EEA, 2019a), while the Equation 6 and 7 is the method developed by US/SCG (SCG, 2019).

$$E_{Manoeuv} = T_{Manoeuv} \ (ME \times LF_{ME} \times EF + AE \times LF_{AE} \times EF + AB \times EF) \quad (4)$$

$$E_{Hotell} = T_{Hotell} \ (AE \times LF_{AE} \times EF + AB \times EF) \quad (5)$$

$$E_{Manoeuv} = (Load_{ME} \times T_{Manoeuv} \times EF \times FCF + Load_{AE} \times T_{Manoeu} \times EF \times FCF + Load_{AB} \times T_{Manoeuv} \times EF \times FCF) \quad (6)$$

$$E_{Hotell} = (Load_{AE} \times T_{Hotell} \times EF \times FCF + Load_{AB} \times T_{Hotell} \times EF \times FCF) \quad (7)$$

Where,

E is the emissions by mode (tonne/year),
$T_{manoeuv}$ is the average time spent during manoeuvring (h),
T_{hotell} is the average time spent at hotelling (h),
ME is the maximum main engine power (kW),
AE is the auxiliary engine power (kW),
AB is the boiler energy default (kW),
LF is the engine (ME and AE) load factor (%),
EF is the emission factor (g/kWh),
Load is the maximum continuous rated (MCR) propulsion engine power times Load Factor (LF) and FCF is the fuel correction factor (dimensionless).

One difference between the two equations is the EF is parameter that will contribute to the differences between both methodologies. This parameter expresses the emitted quantities from the engines of the ships during the different activities, and function of the engine size and technology and power installed or fuel use (EEA, 2019a; EPA, 2009).

The types of fuel used by the ships (for ME, AE and AB) in both methodologies were assigned according to the type of ship based on information from Entec (2010) (Appendix B App. 2.1) (ENTEC, 2010). Some of these data were confirmed on the SeaWeb platform. However, there is still a great deal of uncertainty associated with the definition of engine / fuel type profiles based on the fleet of ships used in the Entec UK Limited 2010 study.

For EMEP/EEA methodology engine/fuel type profiles have been obtained from the Lloyd's number (LMIS) dataset from Entec (2010) study which considered 14 255 ships (ENTEC, 2010). The EF of SO_2, CO_2, HC and the specific fuel consumption (SFC) for main and auxiliary engines were taken from the Entec 2002 study (ENTEC, 2002), while those of CO, CH_4 and N_2O were derived from the study performed by IVL Swedish Environmental Research Institute (IVL, 2004). EF for PM10 depend on the fuel type and sulphur content. These factors were taken from the ICF (2009) study. PM2.5 EF were obtained from a PM10 to PM2.5 conversion factor of 0.92 (ICF, 2009). Uncertainties on emission factors are generally between 20 % and 50 % for the different pollutants (ENTEC, 2002).

For the US/SCG methodology the emission factors were obtained from the ENTEC 2002 study, except for PM, CO, and greenhouse gas emission factors. The PM emission factors were provided by the California Air Resources Board (CARB) study from 2007 (CARB 2007). The study performed

by the IVL Swedish Environmental Research Institute's in 2004 was the source for PM emission factors for gas turbine and steamship ships, as well as for CO and the greenhouse gas emission factors (CO_2 and N_2O). CH were assumed to be 0.2 % of HC emission factors based on the IVL data. The emission factors for base fuel (residual fuel oil/heavy fuel oil (HFO) with 2.7 % sulphur content) and fuel currently being required by CARB regulation (sulphur marine distillate/marine gas oil (MDO/MGO) 0.1 % sulphur content) are shown in.Table S 16.

In the US/SCG methodology the EF are function of the engine type, the NOx control following the IMO regulations (Tier 0 to Tier III) and the sulphur content fuel used. The emission factors for base fuel (residual fuel oil (RO) with 2.7 % sulphur content), together with the current fuel requirements imposed by the European Union directives (MDO/MGO 0.1 % sulphur content) are shown in Table S17 (SCG, 2019). EMEP/EEA methodologies do not differentiate the emission factors as function of fuel's sulphur content and IMO Tier (Appendix B – App. 2.4 in Table S 16).

Another relevant difference between the two equations is the consideration of FCF only in the US/SCG methodology. The emission estimates are adjusted by fuel correction factors accounting for differences in fuels or other factors, comparing to the conditions of emission factor's development (SCG, 2015). Additional data about FCF are included as Appendix B – App. 1.3 (in Table S 14). As previously discussed, the emission factors are appropriate for engines using residual fuel with average sulphur content of 2.7 %. Table S13 lists the fuel correction factors used when the emission factors that are based on 2.7 % sulphur fuel are used for engines that burned fuel with a lower sulphur content. The FCF are applied to propulsion engines, auxiliary engines, and auxiliary boilers if they have switched fuel from the default residual fuel (2.7 % average sulphur content) to a lower sulphur content fuel.

In addition to the FCF and the EF, the LF is also different between the two methodologies. LF relates the engine's power output at a given speed and the engine's MCR power (SCG, 2015b). The Propeller Law establishes that the propulsion engine load is function of the cube of the ratio of actual speed to the ship's maximum rated speed is applied in the estimation of the propulsion engine load factor (SCG, 2019). In Appendix B – App. 2.2 the load factors for both methodologies are shown.

3.4 Results and discussion

3.4.1 Cargo handling equipment emissions

Cargo handling equipment is mainly used in the general cargo and container terminals. There is a total of 68 CHE at Port of Leixões, including 28 forklifts, 21 shovel excavators, 10 reach stackers, 5 cranes, 2 empty handlers and 2 loaders.

The percentage of total NO_x, VOC, CO, N_2O, NH_3, PM10, PM2.5, BC, CH_4, SO_X and CO_2 emissions from cargo handling equipment estimated using the EMEP/EEA (Equation 1) and US/SCG (Equation 2) methodologies are shown in Figure 3, respectively. Figure 4 presents the estimated emissions generated by CHE depending on the equipment type used in the Port of Leixões during 2016. Table S 17 and Table S 18 (in Appendix B – SM3.1) show the CHE emissions by equipment, for all the pollutants listed (in tonnes).

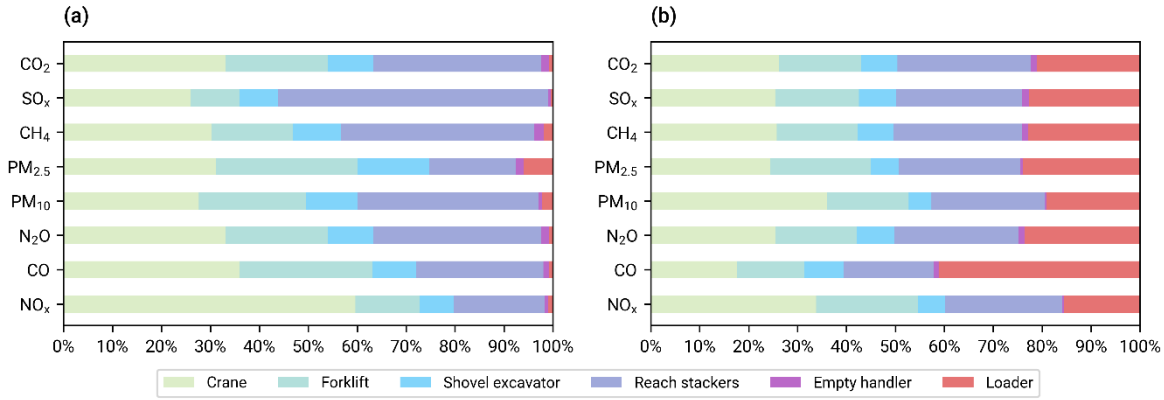

Figure 3. Emission shares of CHE of principal pollutants calculated based on EMEP/EEA (a) and US/SCG (b) methodology.

Results from EMEP/EEA methodology show that cranes have the highest share of the harbour's CHE emissions (25 to 60 %, depending on the pollutant), followed by reach stackers (14 – 55 %) and forklifts (9 – 29 %) (Figure 3 (a)).

Cranes contribute the most for total NO_X emissions (60 %), while reach stackers contribute the most for total SO_X (55 %). However, there is a difference of 53 % between the largest contribution and the lowest contribution of NO_X, with the difference of only 16 CHE. This is because terminal operators replaced their equipment according to technology levels such as EU Stage IV - Tier 4. Table S 18 shows also that CHE emitted a total of 46993 tonnes/year CO_2, 89 tonnes/year NO_X, 46 tonnes/year CO, 6 tonnes/year VOC, 5 tonnes/year PM10, 2 tonnes/year PM2.5 and 2 tonnes/year SO_X, for the EMEP/EEA methodology (see Table S 17).

Zhang et al., 2017 reported a lower contribution of CHE to NO_X, SO_X, and CO_2 emissions in the Port of Nanjing (around of 92 %) compared to our results for the Port of Leixões.

Results from US/SCG methodology show that the CHE generates several emissions, with high emissions of NO_X, as pollutant, and CO_2 shows the greatest contribution of GHG. PM emissions are lower than other atmospheric pollutants emitted by CHE. The cranes have the highest share of the

harbour's CHE emissions (18 to 36 %, depending on the pollutant), followed by loaders (16-41 %) and reach stackers (18-28 %) (Figure 2(b)). Considering the methodology US/SCG results show that CHE emitted a total of 7143 tonnes/year of CO_2, 50 tonnes/year of NO_X, 18 tonnes/year of CO, 3 tonnes/year of HC, 3 tonnes/year of PM10, 2 tonnes/year of PM2.5, 2 tonnes/year of SO_X and 0.33 tonnes/year of CH_4 (see Table S 18).

3.4.2 Shipping emissions

Emissions of the major pollutants (NO_X, PM10, PM2.5, SO_X, CO, VOC, HC) and greenhouse gases (CO_2, N_2O, CH_4) during manoeuvring, and hotelling, both at anchorage and berth were estimated for all categories of ships for 2016, considering both methodologies (i.e., EMEP/EEA and US/SCG). Table S 19 and Table S 21 summarize the total emissions of air pollutants by ship type, operation mode and engine type are summarized (in tonne). Figure 4 illustrates the percentage of emissions for each pollutant for various ship types based on EMEP/EEA (a) and US/SCG (b) methodologies.

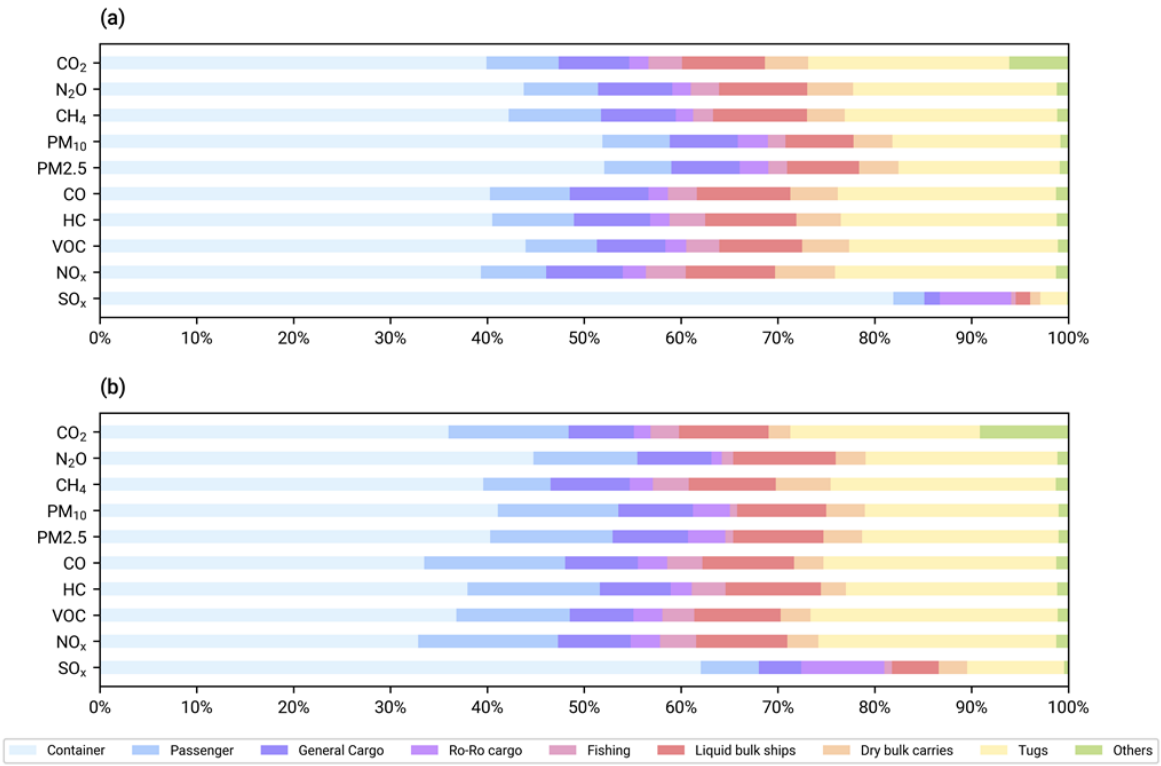

Figure 4. In-harbour ship emissions by various ship type, based on EMEP/EEA (a) and US/SCG (b) methodologies.

Results from EMEP/EEA methodology show approximately 39 to 82 % (depending on the pollutant) of the harbour's total ship emissions attributed to container, while 3 to 23 % of the total ship emissions are attributed to tugs, 2 to 8 % to general cargo ship, 3 to 10 % to passenger ship, 1 to 10 % to liquid bulk ship, 2 to 7 % to Ro-Ro ship, 1 to 6 % to dry bulk carriers, and up to 6 % to others ships (Figure 4 (a)). Ship emissions in Port of Leixões account for 117 030 tonnes/year of CO_2 emissions, 2 201

tonnes/year of NO_X emissions, 914 tonnes/year of SO_X emissions, 54 tonnes/year of PM10 emissions and 43 tonnes/year of PM2.5, as well as various other emissions (see Table S 19). On the other hand, results from US/SCG methodology show the share of emissions (%) for each pollutant and the distinct types of ship: containers ship have the highest percentage of overall emissions, like the findings from the EMEP/EEA methodology, contributing with 33–62 % to the total emissions. Contribution rates from tugs, liquid bulk carriers, general cargo and passenger ships ranged as follow 10-26 %, 5–11 %, 4–8 % and 6-15 %, respectively (Figure 4(b)). Table S 20 shows the total emissions of the different pollutants, namely 102036 tonnes/year of CO_2, 1055 tonnes/year of NO_x, 207 tonnes/year of SO_X, 104 tonnes/year of CO, and 30 tonnes/year of PM10.

Figure 5 illustrates the contribution of shipping emissions for operating modes in harbour based on EMEP/EEA (a) and US/SCG (b) methodologies.

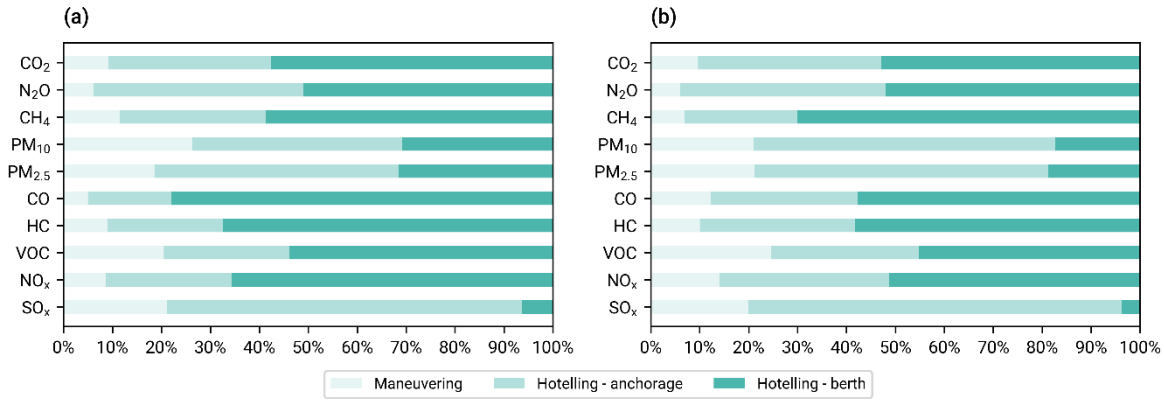

Figure 5. In-harbour ship emissions by operating mode, manoeuvring, hotelling at berth and hotelling at anchorage, based on EMEP/EEA (a) and US/SCG (b) methodologies.

Both methodologies revealed that emissions during hotelling at berth mode were dominant, ranging from 78 % with the EMEP/EEA methodology, and from 70 % from US/SCG, depending on the pollutant type.

This higher contribution during hotelling is consensual. Works have been published reporting dominant contribution from hotelling, either using EMEP/EEA methodology (Saraçoğlu et al. 2013; Nunes et al. 2017; Alver et al. 2018) or US/SCG methodology (Deniz and Kilic, 2010; Song and Shon, 2014; Zhang et al., 2017b). More information regarding these references may be found in appendix B – App. 4. Hotelling emissions stand out possibility due to two major reasons. The first one is the choice of emission factors. In light with the European Union directives (2005/33/EC), since 2010 all ships calling at an EU harbour should use low sulphur fuel (0.1 %) during hotelling. This led to updates on the emission factors for SO_X, namely a sulphur content of 0.1 % m/m in the marine fuel oil, which is also reflected in EF for PM10. In this work, compliance with that normative (meaning EF 0.1 % S) led to a reduction of 96 % and 85 % in SO_X and PM10 emissions, respectively.

Accordingly, (López-Aparicio et al., 2017), also concluded low sulphur (0.1 %) fuel may led to reductions of 90 % of SO_X emissions and 10% of PM10 emissions. In opposite, hotelling was the main contributor to NO_X and SO_X emissions using EF 0.5 %S (Saraçoğlu et al. 2013; Song 2014; Song and Shon 2014; Nunes et al. 2017; Zhang et al. 2017; Alver et al. 2018). The second reason for the different conclusion on the main emitting activity lies on the utilization (or not) of the main engine during hotelling. In this work, and according to the procedures adopted at Port of Leixões, main engine was considered turned off during hotelling. The studies have also considered that during the hotelling the main engines are turned off but failed to consider the boiler emissions (Alver et al., 2018; Deniz and Kilic, 2010; Saraçoğlu et al., 2013; Song, 2014; Song and Shon, 2014; Zhang et al., 2017b) Nunes et al., 2017 did consider boiler emissions but assumed that the main engine was turned on during hotelling.

Figure 6 presents the shipping emissions considering the distinct types of engines, in the Port of Leixões, based on EMEP/EEA (a) and US/SCG (b) methodologies.

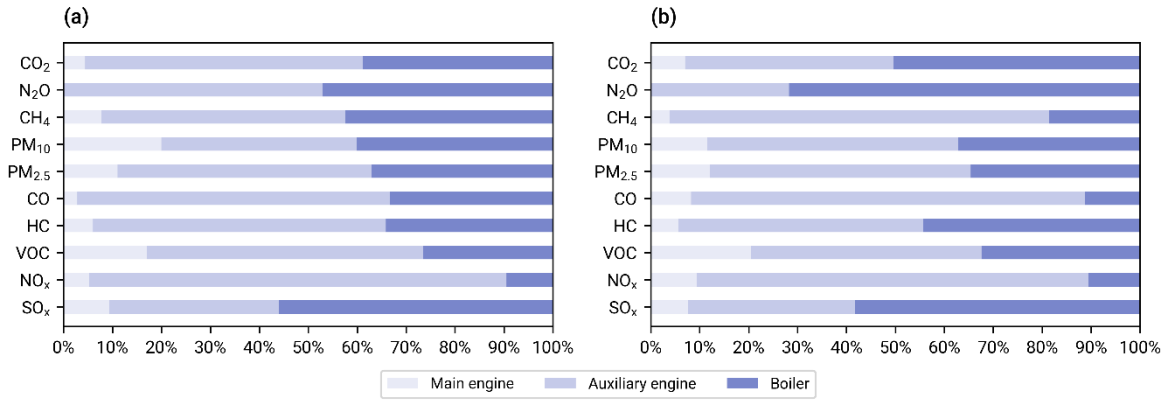

Figure 6. In-harbour ship emissions by various engine type, based on EMEP/EEA (a) and US/SCG (b) methodologies.

Both methodologies revealed that emissions from main and auxiliary engines were dominant. Apart from N_2O and SO_X emissions, to which boiler contributed with 45-70 % and 55-59 %, the share of main and auxiliary engines was above 50 % for all pollutants. The obtained results are in line with those from Song 2014, which presents a study based on the SCG 2010 report (SCG, 2011). On the other hand, Alver et al., 2018 showed greater contribution from the auxiliary engine to NOX, SOX, PM10 and HC emissions, compared to the main engine, using load factors of 40 % for ME and 75 % for AE in manoeuvre mode, and 0 % for ME and 75 % for AE in hotelling mode; in this work, the same load factor was applied for ME, but lower load factors were applied for AE (50 %) in manoeuvre and 40 % in hotelling, according to EMEP Report (see Table S 11) (Alver et al., 2018).

3.5 Emission inventory of CHE and shipping in harbour areas: comparison between EMEP/EEA and US/SCG methodologies

To better identify and comprehend the differences between the two methodologies, Table 3 summarizes the differences between the CHE emission estimates obtained from the two methodologies (EMEP/EEA and US/SCG). To perform this comparison, EMEP/EEA was taken as reference and the relative difference of the emissions obtained by US/SCG methodology was computed according to Equation 8. Positive values represent that EMEP/EEA estimate higher values when compared to the US/SCG, while negative values indicate the opposite (i.e., lower values estimated by the EMEP/EEA methodology).

$$emission\ reduction = \frac{(E_{EMEP/EEA} - E_{US/SCG})}{E_{EMEP/EEA}} \quad (8)$$

Table 3 Relative difference (%) of the total emissions computed by EMEP/EEA, in comparison with US/SCG, for different CHE.

	NO_X	CO	N_2O	PM10	PM2.5	CH_4	SO_X	CO_2
Crane	68	81	72	31	32	-210	22	87.9
Forklift	11	80	71	59	39	-263	-36	87.8
Shovel excavator	56	64	70	77	65	-167	23	87.9
Reach stackers	27	73	73	67	-22	-142	63	87.9
Empty handler	76	65	73	75	78	-142	-120	87.9
Loader	-888	-1817	-1027	-251	-349	-245	-608	-4306

High differences between emission factors from the two methodologies produce relevant difference on the total emission estimates. The emission factors in the US/SCG methodology depend on a parameter designed as zero-hour emission rate by fuel type, as well as the CHE engine type, model year, and cumulative hours (Equation 3).

The equipment presents different cumulative hours. In 2016, the total operating hours are as follows: 8793 h for 5 cranes, 6745 h for 28 forklifts, 4284 h for 21 shovel excavators, 2707 h for 10 reach stackers, 385 h for 2 empty handlers and 857 h for loaders. Besides the different accumulated hours associated to the different equipment, another factor contributing to discrepancies between methodologies is the inclusion of distinct variables between methodologies, namely the deterioration factor adjustment in the EMEP/EEA methodology and the fuel correction factor in the US/SCG methodology.

Due to such differences, EMEP/EEA estimates higher total emissions (for all CHE combined) of NO_x, CO, N_2O, PM2.5, PM10, SO_X and CO_2 (up to 85 %), and lower emission of only CH_4 (264 %), comparing to US/SCG. Cranes and forklifts generate the highest CO emissions when following the EMEP/EEA methodology.

In addition, the main difference for loader may be due to the use of a piece of equipment from 1981, with 147 kW engine power and 195 hours of use. As the emission factors are chosen based on the

level of technology and the engine power of each CHE, the Loader presents the highest emission factors in both methodologies. However, and unlike EMEP/EEA, the US/SCG methodology applies emission factors estimates based on cumulative hours of usage (equation 2), instead of listed emission factors. Therefore, those emission factors have a higher impact on the total emissions estimates in cases of usage of such obsolete technology for long periods.

Table 4 and Table 5 present the relative differences (%) found in the total annual emissions of ships, per type of ship (Table 4) and operation mode and engine type (Table 5Table 6). Table 4 addresses different ship types and Table 5 addresses different operation mode and engine type.

Table 4. Relative difference (%) of the total emissions computed by EMEP/EEA methodology, in comparison with US/SCG methodology, for different ship types.

	Container	Passenger	General Cargo	Ro-Ro cargo	Fishing	Liquid bulk ships	Dry bulk carries	Tugs	Others
SO_X	83	57	38	74	62	27	37	18	32
NO_X	60	-3	55	39	57	51	75	48	53
VOC	52	9	47	21	45	40	64	32	41
HC	11	-53	12	-3	11	1	47	7	10
CO	48	-11	42	5	23	38	61	33	38
PM2.5	51	-16	30	18	73	21	37	23	27
PM10	57	2	40	33	77	28	46	37	33
CH_4	35	49	27	7	-26	36	0	26	22
N_2O	-10	-50	-7	40	57	-25	30	-1	-1
CO_2	21	-45	20	24	26	6	56	18	-30

Table 5. Relative difference (%) of the total emissions computed by EMEP/EEA, in comparison with US/SCG, listed by operation mode and engine type.

	SO_X	NO_X	VOC	HC	CO	PM2.5	PM10	CH_4	N_2O	CO_2
Manoeuvring	10	-19	2	-18	-83	9	35	62	-2	5
Hotelling - anchorage	-1	1	4	-43	-33	4	-18	51	-4	-1
Hotelling - berth	45	43	31	8	45	53	54	25	-8	17
Main engine	19	-35	-2	-2	-126	11	52	68	0	9
Auxiliary engine	6	31	32	11	6	18	-5	2	43	32
Boiler	0	18	0	-38	74	26	24	72	-61	-22

Table 4 shows that the general cargo ships, containers, and fishing are the ones that reveal the highest EMEP/EEA emissions compared with US/SCG emissions. On the other hand, the tugs, passenger, and other ships show the highest US/SCG emissions, when compared to the EMEP/EEA. Since tugs and other types of ships have the same emission factors, it can be concluded that this variation may be due to the number of ships, GT, and power engine.

Among the considered pollutants, EMEP/EEA methodology estimates higher total emissions (for all types of ship combined) of SO_X, NO_X, VOC, PM2.5, PM10, CH_4, HC, CO, and CO_2 (up to 46 %), and lower emission of only N_2O (up to 7 %), comparing to US/SCG. In fact, HC is the pollutant with the

highest emission estimate by US/SCG methodology – more than twice (53 %) the estimate of EMEP/EEA.

Regarding operation mode, the highest discrepancies are found for CO, CH_4 and PM10 (Table 5). The emissions estimate of VOC, PM10 and CH_4 always present positive differences, meaning higher estimate by EMEP/EEA.

Regarding the type of engines, the highest discrepancies are found for CO, CH_4 and PM10 particularly considering boiler and main engine. The emissions estimate of SO_X, PM2.5 and CH_4 present positive differences, meaning higher estimate by EMEP/EEA. These differences are mainly due to different emission factors and load factors recommended by each methodology, as well as the introduction of fuel correction factors used by US/SCG methodology as adjustment of the emissions estimates. Regarding EMEP/EEA methodology, the load factors were 40 % for ME and 50 % for AE in manoeuvre mode, while for hotelling mode the LF were 0 % for ME and 40 % for AE. Regarding US/SCG methodology, the load factors were 15-27 % for ME and 33-80 % for AE in manoeuvre mode, while for hotelling mode the LF were 0 % for ME and 10-64 % for AE (see Appendix App. 2.2).

3.6 Conclusions

This work highlighted and discussed the differences obtained when different emission's estimation procedure (EMEP/EEA and US/SCG methodologies) are used to estimate emissions associated to port activities, CHE, and shipping.

Such differences lead to very distinct emission's estimate. Regarding CHE, the EMEP/EEA methodology is based on the deterioration factors adjustment, while the US/SCG methodology uses fuel correction factor; moreover, different methodologies for emission factors definition also contributed to the discrepancies found in CHE emissions. EMEP/EEA estimates higher total amount (for all CHE combined) of NOx, CO, N_2O, PM2.5, PM10, SO_X and CO_2 (up to 85 %), and lower emission of only CH_4 (264 %), comparing to US/SCG. Cranes and forklifts generate the highest CO emissions when following the EMEP/EEA methodology. Regarding shipping, the main differences between the two methods are related to the very distinct emission factors and load factors, plus the consideration of fuel correction factor in only one of the methodologies (US/SCG). Among the considered pollutants, EMEP/EEA methodology estimates higher total shipping emissions (for all types of ship combined) of SO_X, NOx, VOC, PM2.5, PM10, CH_4, HC, CO, and CO_2 (up to 46 %), and lower emission of only N_2O (up to 7 %), comparing to US/SCG.

Despite all these discrepancies, the results of both emissions inventories have shown that shipping activity contributes the most for global emissions, especially during hotelling at berth operations, for the Port of Leixões case study (2016 year).

This study points out that it is of the utmost importance to standardize a common methodology for all ports to estimate emissions coming from ships and from CHE.

The results achieved highlight the need for each harbour authority to draw up its own emissions' inventory based on their reality and using not only the most recent emission factors but also to consider emission factors that attend to the regulations on sulphur emissions by the maritime sector. Emission factors are usually pointed out as the weakest link in ship emission inventories since they continue to be derived from very limited data. Emission testing of ship is an expensive and difficult undertaking, thus data on real emissions are relatively rare. An array of reasons may influence a measurement, such as different port/ship technologies (e.g., the adoption of cleaner technology), ship speed and load, or even different emission testing protocols. The shortage of real measurements on ship emissions strongly influences the emission factors adopted from each data source. Literature points to uncertainty in the order of 20 % - 50 % in emission factors for the different pollutants (ENTEC, 2002).

This work hinted that the EF used by EMEP/EEA method for CHE emissions may raise less doubts and probably are the most recommendable, since they account for technology level and engine size class (according to the guidelines of emissions for non-road mobile machinery), rather than just ZH and DR, as defined by US/SCG method. On the other hand, regarding shipping, the US/SCG report may be more adequate considering the legislation in force since January 2020, given that it differentiates the emission factors according to the sulphur content of the fuel and the IMO level. Therefore, we suggest the development of a new harmonized methodology combining both EMEP/EEA and US/SCG for the aspects where each one performs best.

Besides emission factors, two other major issues need harmonization: (i) whether to consider the boiler as emission source or not, and (ii) to define if the main engine should be considered turned on or off, while hotelling. Moreover, it is still unclear in the reports which type of engine should be considered for in each operation mode. Finally, higher-resolution input data on fuel consumption and EF could help reducing some of the described uncertainties in emission's inventory.

Chapter 4

4 Assessment of source contribution to air quality in an urban area close to a harbour: case-study in Porto, Portugal

The content present in this chapter has been published as:

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4.3 Introduction

Ports are a critical feature of the world's economy. Ports serve as critical hubs for the continual flow of agriculture, energy, and consumer products from coastal communities to the inland areas (Viana et al., 2015). Despite the economic benefit they provide, activities associated with port operations are also an environmental concern with potential effects on local climate, the weather, human health, and ecosystems (Lonati et al., 2010; Stockfelt et al., 2015).

As multi-modal transportation hubs, ports can be significant sources of air pollution due to the maritime transit, manipulation, and storage of materials in bulk and containers, and due to land transport associated with these activities which produce a significant release of atmospheric pollutants that can cause air quality problems (Alastuey et al., 2007; Almeida et al., 2012; Moreno et al., 2007).

Port emissions affect the residents of neighbouring communities, especially sensitive population groups, including children and old people (Corbett et al., 2007). In several ports located in Europe, studies have shown that the primary air pollution concern is emissions from dust and fumes that occur from everyday operational activities in harbours (Corbett et al., 2007; Pérez et al., 2016; Tian et al., 2013). Despite strict measures to reduce air pollutants, several European countries still face air pollution episodes regularly exceeding the established legal limits values. Europe's most troublesome pollutants regarding human health are PM, NO_2 and ground-level ozone (O_3) (EEA, 2018; EEA, 2015). Most EU member states, mainly in urban agglomerations where human exposure is also higher, have reported exceedance of the NO_2 thresholds (EEA, 2018; EEA, 2015). The annual NO_2 limit value continues to be widely exceeded across Europe, with around 10 % of all the reporting stations recording concentrations above the standard limit in 2015 in 22 countries out of the EU-28 (EEA, 2016). In Portugal, over the last few years, air quality problems have been detected, particularly concerning PM10 (Borrego and Costa, 2011; Tchepel et al., 2010) and NO_2 in the northern region (Borrego et al., 2004).

According to the European Sea Ports Organization (ESPO, 2013), the top environmental priority for seaports is the local air quality, focusing on the health of the workers and nearby residents. With the

projected increase of shipping activities, air quality in and around ports is gaining emphasis, especially for ports surrounded by high-density residential areas. Improved knowledge on this type of emissions remains scarce and there are relatively few monitoring and experimental data available to quantify the contribution of ship emissions to local air quality (Isakson et al., 2001; Sorte et al., 2018). Since emissions from harbour-related activities can have a significant impact on air quality, the study of microclimate conditions and resulting pollutant dispersion patterns in port areas was of the utmost importance. Coastal areas, where harbours and city ports are located, experience specific meteorological patterns, like sea and land breeze phenomena, playing an important role in the dispersion, transformation, removal, or accumulation of air pollutants (Baumgardner et al., 2006).

According to the "Shipping Emissions in Ports" report, issued by the International Transport Forum (ITF), shipping emissions in ports accounted for 0.4 million tons of NO_X, 0.2 million tons of SO_X and 0.03 million tons of PM10 worldwide during 2011. Around 85 % of the emissions come from container ships and tankers. Although container ships have short port stays (few hours to a few days), their emissions are very high (Merk, 2014). Approximately 230 million people are directly exposed to shipping emissions in the top 100 world ports (Merk, 2014). These emissions have increased at a large pace over the last few decades and are expected to continue to increase further in the near future.

Data from the Los Angeles County Health Survey revealed that Long Beach communities near the Ports of Los Angeles and Long Beach (two large ports right next to each other and ranked top 2 in the U.S.) experience higher rates of asthma (2.9 % on average), coronary heart diseases and depression, compared to other communities in Los Angeles (HIP, 2010). Additionally, the California Air Resources Board attributed 3,700 premature deaths per year to port activities and shipment of goods (THE, 2012). On a global scale, calculations suggest that shipping-related PM emissions are responsible for approximately 60,000 cardiopulmonary and lung cancer deaths every year, with most deaths occurring near coastlines in Europe, East Asia, and South Asia. These data show the impact of ports on air quality and human health. Thus, it is of absolute importance to study the dispersion behaviour of pollutants in the vicinity of ports, so adequate minimization measures can be taken.

Several studies have tackled the contribution of ships to the local air quality using different approaches. It has been investigated using models with a specific focus on atmospheric aerosol (Gariazzo et al., 2007; Marmer et al., 2009); or using experimental analysis at a high temporal resolution (Contini et al., 2011; Donateo et al., 2014); or even using receptor models based on the identification of chemical tracers associated with ship emissions (Bove et al., 2014; Cesari et al., 2014; Viana et al., 2009). Although the potential impact of ship emissions on air quality is known from model studies at a more aggregated level, the knowledge based on the attribution of local air quality problems to ship emissions in areas close to shipping lanes is rather limited, and there is a clear need to improve the observation-based knowledge.

To help community groups assess the impacts from port activities, researchers at the University of North Carolina's Institute for the Environment (UNC-IE) in collaboration with the U.S. Environmental Protection Agency (EPA) have developed a research grade-screening tool for near-port

assessments. The Community screening tool for near-PORT (C-PORT) assessments (Isakov et al., 2017) is designed to provide a platform for air-quality modelling and visualization that can inform users about potential local air quality impacts in the vicinity of ports (Arunachalam et al., 2015) in the U.S.

This paper presents a case study for assessing the relative contribution of various sources such as port activities, shipping emissions, roadway traffic and industry to air quality near the Port of Leixões, in northern part of Portugal. In this study, we expanded the C-PORT tool, which was initially focused on U.S. ports alone. Thus, this work presents the first case study for ports outside the U.S. This tool allowed the assessment of the air quality impact for different types of emission sources to be considered when simulating dispersion of pollutants in port and adjacent areas. One relevant feature of the different emission sources is their height of release, which causes different patterns of pollutant dispersion in port areas. Area sources or roads and rails (line sources) emit typically at ground level, while (some) point sources and ships-in-transit (line source) emit typically at greater heights, leading to plume rise. Special attention was given to the critical pollutants monitored in the study region, namely NO_X and PM10, which have been the focus of air quality plans due to the continuous exceedances measured in the last years.

Section 2 describes the case study, including local meteorological and air quality characterization. The modelling approach (based on C-PORT tool) is presented in Section 4.3, together with modelling setup and input data description. The main results are presented in Section 4.4, while Section 4.5 provides the main conclusions.

4.2. The Port of Leixões case study

The Port of Leixões has become a crucial point for Europe's shipping lines. Due to its geographic location, it is one of the main operation centres in Portugal. This port is situated in the northern part of Portugal, in the North-West corner of the Iberian Peninsula, about 2.5 km north of the River Douro and near the city of Porto, being surrounded by the towns of Leça da Palmeira (to the North) and Matosinhos (to the South). Matosinhos and Leça da Palmeira belong to the metropolitan Porto area, with 130,984 inhabitants and 18,502 inhabitants, respectively (INE, 2011).

Representing 25 % of the Portuguese foreign trade by sea and moving 16.4 million tons of goods per year, Leixões is mainly an export port, serving virtually all types of ships and cargo, as well as cruise ships. With 5 km of quay, 55 ha of embankments and 120 ha of wet area, Leixões is equipped with the most updated information systems for ship traffic control and management. In Figure 7, the simulation domain of C-PORT is shown, with the identification of the main emission sources (industrial; port and road) and monitoring (meteorological and air quality) stations.

It is worth noting that the simulation domain comprised another source of emissions that may affect air quality in the studied area. This source was a refinery facility with distillation capacity around 4.4 Mt/year (second largest in Portugal), located north of the Port of Leixões and connected to the tanker terminal by several pipelines of approximately 2 km length.

Figure 7. Geographical/simulation domain of the study area of Porto of Leixões, with the locations of the port terminals, refinery and surrounding urban area.

The following Section details the meteorological and air quality characterization that was performed for the study area, based on monitoring data.

4.2.1. Meteorological characterization

There were two meteorological monitoring sites in the study region, including an onsite meteorological station located inside the harbour (see Figure 7). Wind roses for this onsite station, for a 3-year period from 2014 to 2016, are shown in Figure 8.

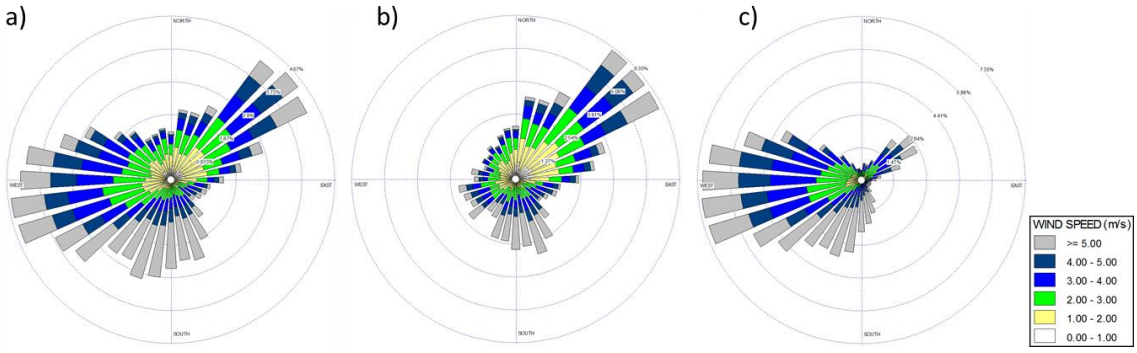


Figure 8. Wind roses for the meteorological station during a 3-year period (2014 to 2016) considering a) all hours, b) daytime hours: 00 a.m. to 9 a.m., and c) night-time hours: 10 a.m. to 8 p.m.

The presence of local sea/land breeze features can be clearly seen in the wind roses for daytime and night-time periods (Figure 8 (b) to Figure 8 (c)). The most common wind patterns showed a diurnal pattern, with winds blowing from the West (W) – Southwest (SW) quadrant (from ocean to land) during the daytime and from Northeast (NE) wind (from land to ocean) during the night-time. This diurnal pattern suggested that pollutant concentrations over the urban area close to the Port of Leixões would be higher during the day, but pollutants emitted from the port during night-time were expected to be dispersed and transported over the sea. The pattern of dispersion of pollutants emitted during the night-time and daytime periods was different due to differences in wind speed and stability. Higher wind speeds were typically experienced during the daytime.

Atmospheric stability conditions played a crucial role in understanding the air quality impacts of port activities and will be discussed further below.

4.2.2. Air quality characterization

There were three monitoring stations, part of the monitoring network maintained by the Portuguese Environmental Agency (http://qualar.apambiente.pt), located inside the study domain: 1) Seara; 2) João Gomes Laranjo; and 3) Custóias, as shown in Figure 7. These stations continuously measure hourly data for the main atmospheric pollutants (PM10, SO_2, NOx and CO). QualAr database provided continuous measurements based on 1 h averages with data registry every 15 min. Air quality monitoring stations were placed according to specific legislation to guarantee the representativeness of the Portuguese territory. Besides these stations, PM10 concentrations measurements were made inside the Port (for the 4-year period from 2013 to 2016). All these air quality monitoring sites were highlighted in Figure 7.

The 95 % confidence intervals for the annual average of PM10 and NO_2 observations from the air quality network are shown in Figure 9. The data collection efficiency was > 80 % for all sites presented in Figure 9. The Figure also includes PM10 annual average concentrations from onsite observations (2013-2016 period).

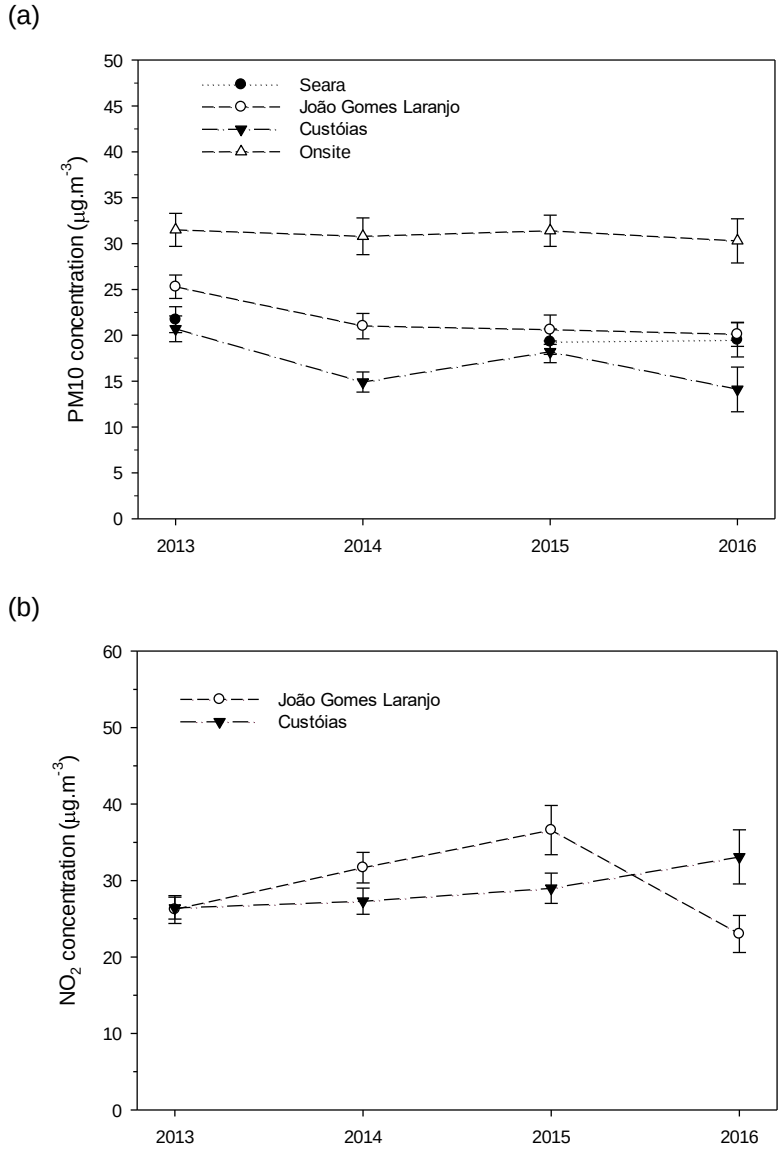


Figure 9. The 95 % confidence intervals for the annual average of PM10 (a) and NO_2 (b) observations from the air quality monitoring network, and PM10 observations from the onsite monitoring station (inside port area).

PM10 concentrations at the Seara and João Gomes Laranjo sites were within 19-21 µg/m^3 range. These two stations are classified as urban sites and located closer to the port terminals than the Custóias suburban site. Concentrations at Custóias were between 14-18 µg/m^3, about 25 % lower than at the urban sites. Similar differences between João Gomes Laranjo (urban) and Custóias could be seen for NO_2 in 2014 and 2015. Nevertheless, for 2016 the Custóias (suburban) station had higher

concentrations than the João Gomes Laranjo (urban) site. This inversion in the NO_2 trend observed in João Gomes Laranjo station compared to Custóias may be due to some shift in the urban dynamics, namely the introduction of a toll charge in the highway near this station, and/or changes in traffic patterns due to support of public transport in the Porto municipality. On the other hand, Custóias station is representative of the suburban area of Matosinhos city, which has been undergoing rapid urbanization. In fact, we recommend that the classification of this monitoring station as "suburban" environment should be re-assessed to verify if it is still applicable.

Table 6 presents the number of exceedances to the hourly limit value of PM10 (50 µg/m^3) and NO_2 (200 µg/m^3) measured at the air quality stations from 2013 to 2016.

Table 6. The number of exceedances to the PM10 and NO_2 limit values, from 2013 to 2016 at the air quality stations.

	PM10 daily exceedances				**NO_2 hourly exceedances**			
Station	**2013**	**2014**	**2015**	**2016**	**2013**	**2014**	**2015**	**2016**
Onsite	36	42	44	27	NA[a]	NA	NA	NA
Custóias	17	1	2	0	0	0	0	0
João Gomes Laranjo	16	2	0	3	5	0	0	0
Seara	7	NA	0	0	NA	NA	NA	NA

[a] NA – Not available

No exceedances to the hourly limit value of NO_2 were registered, except for João Gomes Laranjo station, in which 5 exceedances were recorded in 2013. All the air quality stations were complying with the requirements of the EU Directive 2008/50/EC, which states that the number of exceedances to the hourly limit value could not exceed more than 19 times in a year.

On the other hand, according to the same legislation, daily maximum PM10 concentration of 50 µg/m^3 must not be exceeded more than 35 days per year. In this work, this limit was compiled in the Onsite station, in 2016.

4.3. Modelling approach

4.3.1. Description of C-PORT

As a research grade-screening tool, C-PORT is designed to be an easy-to-use computer modelling tool for exploring the range of potential impacts that changes to port operations might have on local air quality. C-PORT predicts concentrations of multiple primary pollutants: CO, SO_2, NO_X, PM10 and selected Mobile Source Air Toxics (benzene, formaldehyde, acetaldehyde, and acrolein) at fine spatial scales in the near-source environment with access through an easy-to-use web-based platform. C-PORT can also be used to model air quality concentrations based on representative emissions and meteorological conditions, such as summer rush hour traffic within a stable atmosphere. The key model inputs include emissions and meteorology, and model outputs are presented as geospatial maps. Users can run the model with the included default data or input their

own locally derived values. C-PORT was constructed with an intended purpose of calculating differences in annual averaged concentration patterns and relative contributions of various source categories over the spatial domain within about 10 km of the port. However, this tool also has some limitations, such as it does not include atmospheric chemistry to account for secondary pollutants such as ozone or secondary aerosols and does not account for local variations in terrain. Further, C-PORT is not intended to assess model predictions for specific hours (e.g., a specific date and time). Meteorological inputs include hourly observations of wind speed and direction, ambient temperature, and other atmospheric boundary layer parameters needed for dispersion modelling. These data were processed through AERMET (https://www3.epa.gov/scram001/metobsdata_procaccprogs.htm), a meteorological data pre-processor for AERMOD. Subsequently, using the methods described in Isakov et al (2017), the typical hourly inputs for five different atmospheric conditions related to stability class for each of two seasons is identified from the annual dataset. For Portugal applications, C-PORT uses hourly weather measurements from the onsite monitoring site that is nearest to the study location to calculate the representative hours. C-PORT allows the user to simulate short-term (hourly) or long-term (annual) concentrations. For short-term, the user can model any of the five representative meteorological conditions: 1) Stable, 2) Slightly Stable, 3) Neutral, 4) Slightly Convective, and 5) Convective), and for each season (Winter & Summer), and an annual average option, based on 100 representative meteorological hours for each station, is also available. These 100 hours include a combination of 5 wind speeds, 4 wind directions and 5 stability conditions. The dispersion algorithm is run explicitly for the 100 hours, and then weighted by frequency (how often these 100 hours occur in the annual dataset) to estimate the annual averages. This method called the METeorologically - weighted Averaging for Risk and Exposure (METARE) is described further in (Chang et al., 2015).

C-PORT allows the user to upload custom inputs for port terminals, ships-in-transit, roadways, rail, and point sources. The required parameters include source locations (latitude and longitude in decimal degrees) and annual emissions (in tons/year) for multiple pollutants: NOx, CO, SO2, PM2.5, PM10, EC2.5, OC2.5, benzene, formaldehyde, and acrolein. For ships-in-transit and point sources, stack parameters are required: stack height (m), stack diameter (m), stack temperature (deg. K), and stack exit velocity (m/s).

C-PORT includes dispersion algorithms for area, point, and line sources related to freight-movement activities and emissions from the port terminals. The dispersion code for area and point sources is based upon model formulations used in AERMOD (Perry et al., 2005), while the road and rail are modelled as line sources, based upon an analytical approximation that is used in the C-LINE modelling system (Isakov et al., 2017).

The dispersion algorithm of C-PORT allows for the vertical distribution of emissions above the surface for point sources, reflecting the stack heights of ships and refinery. The model assumes that the concentration distributions in the vertical and horizontal are Gaussian except for convective conditions, in which case, we used a bi-Gaussian distribution (Isakov et al., 2017).

The dispersion algorithm for line sources is designed to calculate near-source pollution profiles representing emissions from roadway traffic and rail. This tool represents a highway as a set of line sources located at the centre of each lane of the highway. Each line source is represented as a set of elemental point sources (Isakov et al., 2017). C-PORT modelling system is the dispersion algorithm that calculates near-source pollution gradients for buoyant line sources.

The dispersion algorithm is designed to specifically model moving line sources such as ships in transit (Isakov et al., 2017). Assuming that the averaging time for the calculation is long compared to the transit time of the ship, we can model the moving ship as a line source laid along its path. This source has buoyancy corresponding to the exhaust gases of the ship (Isakov et al., 2017). The dispersion algorithm is designed to efficiently model area sources representing emission sources such as dray trucks or rubber tire gentry at port terminals. As in AERMOD, an area source is treated as a polygon. The emissions from the area source are distributed among a set of line sources that are perpendicular to the near surface wind (Isakov et al., 2017).

The roadways emissions in C-PORT were consistent with the C-LINE web-based model (Isakov et al., 2017) that estimates the air quality impacts of traffic emissions for roadways in the U.S. Roadway emissions in C-PORT are calculated based on a combination of road network, traffic activity and emissions factors. A road network is the system of interconnected roadways, and a description of their types (e.g., principal arterials such as interstates). Traffic activity describes the number, types, and speeds of vehicles on a given roadway and for a given time.

4.3.2. Modelling inputs and setup for Porto case study

Several scenarios were defined and simulated to estimate the relative contributions of the different source sectors, namely harbour, roadway traffic, and industry to the air quality over the Port of Leixões case study. For these simulations, the modelling domain defined covered an area of approximately 10x10 km^2 with spatial resolution of 40 m (Figure 7). We provide a brief description of the model input data, namely meteorology and emissions data below.

Meteorological data

Hourly meteorological observations from the onsite meteorological station were used to create the meteorological inputs in C-PORT for 2016, using the same methods as in Isakov et al, 2017. The typical hourly inputs for five different atmospheric conditions related to the stability class (stable; slightly stable; neutral; slightly convective and convective) and for each of the two seasons – winter and summer - are identified from the annual dataset. Table 7 summarizes these meteorological inputs that include hourly observations of wind speed (Ws), direction (Wd), surface friction velocity (uStar), height of the mechanically generated boundary layer (Zimech), Monin-Obukhov length (Lmon), surface roughness length (Zo), reference height for wind (RefHt) and ambient temperature (Temp), and the respective atmospheric stability class (Disp).

Table 7. Meteorological inputs data (for year 2016).

Season	Disp	Wd (º)	Ws (m/s)	uStar (m/s)	Zimech (m)	Lmon (m)	Zo (m)	RefHt (m)	Temp (K)
WINTER	Stab	209	0.8	0.172	674	32.7	1	10	284
	sStab	209	2.9	0.688	1404	521.1	1	10	284
	Neutral	209	11.1	1.929	4000	-3456.3	1	10	284
	sConv	209	3.9	0.688	1439	-529.1	1	10	284
	Conv	209	1.8	0.369	574	-47.8	1	10	284
SUMMER	Stab	241	0.8	0.172	228	32.8	1	10	291
	sStab	241	2.4	0.568	1045	354.9	1	10	291
	Neutral	241	5.4	1.325	3641	3273.8	1	10	291
	sConv	241	3.9	0.709	1430	-234.6	1	10	291
	Conv	241	1.8	0.375	585	-41.7	1	10	291

In both seasons, we found typical wind directions from Southwest, characterized by higher wind velocities during the winter period (and mainly for neutral atmospheric conditions). As expected, the highest boundary layer heights were found in neutral conditions (reaching 4000 m in winter) and the lowest for stable conditions with values inferior to 300 m in summer season. The difference estimated in surface temperature between the two seasons is only 7 degrees.

Methodologies for port-related emission input

In order to support the local air quality modelling study, a local-scale emission inventory has been developed for the Port of Leixões case study.

Emission sources were categorized as mobile sources (e.g., automobiles, trucks, buses and ship in transit), point sources (e.g., a refinery and ship hoteling), and area sources (e.g., cargo handling equipment and bulk material stored). Mobile sources were further categorized as on-road sources (e.g., automobiles, trucks, buses) and non-road mobile sources (e.g., construction equipment, cranes, yard trucks, locomotives, and marine vessels). Mobile source port-related emissions were generated by ships and by land-based sources at ports. Marine emissions came primarily from diesel engines operating on ships, tugs, and other ships operating within a port area.

Land-based emission sources included CHE, such as terminal tractors, cranes, container handlers, reach stacker, backhoes and forklifts, as well as heavy-duty trucks and locomotives operating inside a port area. The harbour-related emissions with high spatial and temporal resolution were estimated. The Port of Leixões is equipped with 14 operating terminals, with the container terminal having the highest port traffic. In 2016, there was a record of 2,717 ship calls corresponding to a total of 32,849,816 Gross Tonnage (GT). A summary of the ship and cargo activity in the Port of Leixões is shown in Table 8.

Table 8. Number of ships and cargo in the Port of Leixões (for year 2016).

Terminal	Number of Ships	Number of cargo	Units
South container	1288	434,604	(TEU)
North container		132,535	(TEU)
Solid bulk	379	2,567,999	(tons)
Liquid bulk		8,352,890	(tons)
Roll on Roll Off	134	705,033	(tons)
Cruise	85	79,065	passengers

Shipping emissions

A bottom-up methodology to estimate emissions, proposed by the European Monitoring and Evaluation Programme/European Environment Agency (EMEP/EEA), was used to prepare emissions input for the model.

Regarding shipping emissions (marine emissions), the activity-based method was based on ship movement information and involved the application of emission factors to a particular ship activity, namely manoeuvring or hoteling. Those emission factors expressed the emitted quantities for the operational status of the ship's engines during each activity, depending on engine type and size, engine nominal power, fuel type and time spent in port (EEA, 2016).

Fugitive Emissions

The storage of materials in open-air storage was another main area source (considering land-based emissions sources). Emissions from those stockpiles stored at terminals were estimated based on the methodology developed by the US Environmental Protection Agency (EPA, 2009). This methodology was proposed for loading/unloading operations of particulate material cargo. Additionally, an emission reduction factor was applied to the General Cargo and Bulk Terminal of Leixões, which was previously developed and applied by Borrego et al. (2007). This reduction percentage was due to the application of containers and windbreaks around bulk piles, since it promoted a decrease of the total amount of particulate emissions from the terminal, through the diminishing of the wind velocity in the pile surface.

Road mobile Emissions

Non-road mobile sources (cargo handling equipment) were divided into groups by engine fuel type, and pollutant emission factors were established based on the equipment model year, as well as the engine power (EEA, 2016).

Finally, on-road sources (roadway emissions) were estimated using the TREM (Transport Emission Model for Line Sources) model (Borrego et al., 2004), based on estimated origin/destination (O/D) matrices, traffic counts available for the Porto urban area, the vehicles' average speed in each main route, and statistical data from the Porto vehicle fleet. This emission model has already been

extensively applied in Portugal and in the Porto region, exhibiting good agreement when compared/validated against observational data (Borrego et al., 2012; Relvas et al., 2017).

Figure 10 below shows the percentage of each source (ships, trucks, locomotives, and CHE) that contributed to the overall Port of Leixões emissions.

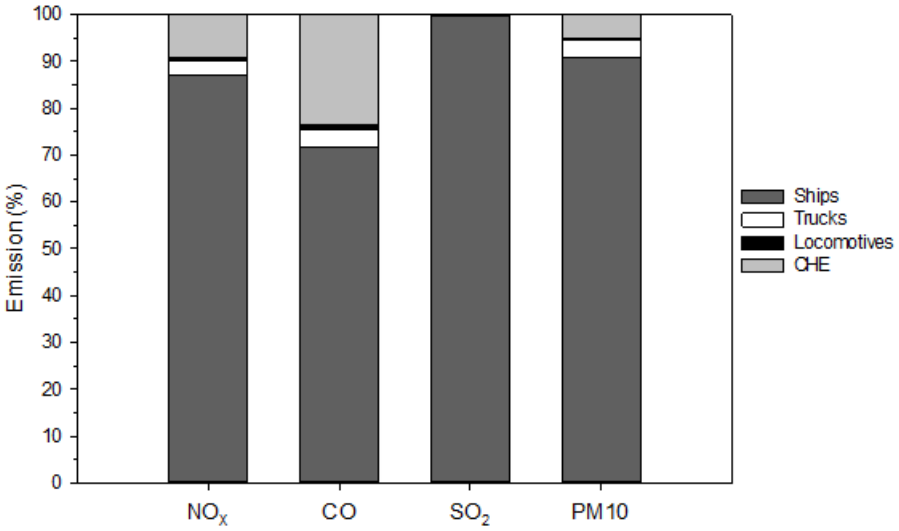


Figure 10. Relative contribution by each source category for the harbour emissions, at Port of Leixões, in 2016.

The dark gray bars highlight the fact that ships are by far the biggest source of emissions at the Port of Leixões. CHE is the second biggest source, followed by trucks and trains. Trucks and trains contribute less than 11 % and 3 % of the port's emissions, respectively.

4.4. Air quality modelling results

The analyses of the C-PORT modelling results include the model evaluation using observations and several sensitivity tests regarding meteorological conditions and sources contribution. The background concentrations, required by the model, were obtained throughout the regional modelling simulation done for the Portugal domain in the scope of the forecasting system operating daily (Monteiro et al., 2007; http://previsao-qar.web.ua.pt/). The background values used were 5 µg/m^3 NO_X and 6 µg/m^3 PM10, and the total concentrations were obtained by adding these to the C-PORT based local source contributions.

4.4.1 C-PORT tool results

In order to evaluate the model performance, Figure 11 shows the observed and modelled concentrations at each monitoring point.

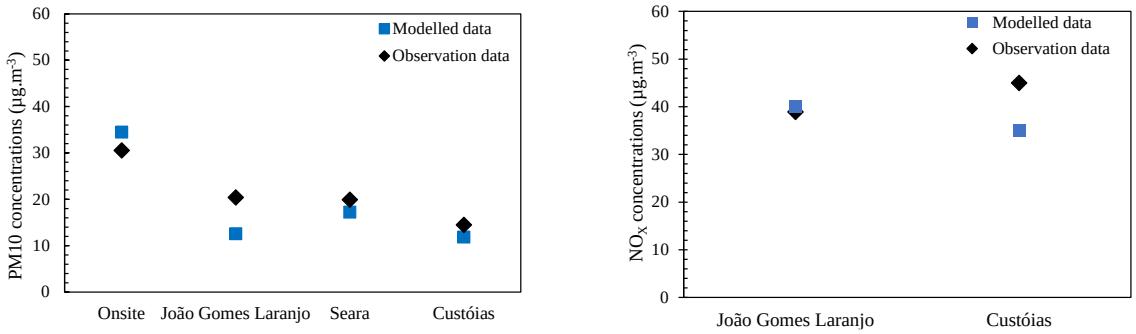


Figure 11 Observed and modelled annual averages of PM10 (left) and NO_X (right) at the air quality stations. The modelled values represent the range of concentrations simulated in the grid cells in and around the one containing the monitoring site.

The PM10 concentrations measured in the air quality stations are close to the values obtained by C-PORT, aside from the João Gomes Laranjo (urban) station, which presented a difference between the modelled and observed concentrations of 10 µg/m^3. Furthermore, modelled data ranges allowed the evaluation of spatial variability of the modelled results nearby the main receptor. Port of Leixões' onsite station showed the widest range of modelled concentrations, corresponding to the solid granulate and the containers terminals emission sources. Regarding the other stations, Seara and Custóias, the concentrations measured in the air quality stations were also within the range of the modelled concentrations by C-PORT.

In relation to NOx, it was possible to observe the spatial variabilities of the simulated values when compared to the observed values in the two stations. The model can perfectly capture the NOx magnitude values at the urban station - João Gomes Laranjo.

Figure 12 and Figure 13 show the modelled annual averages of PM10 and NO_X together with the observed values for the air quality stations (AQS) nearby the Port of Leixões, respectively.

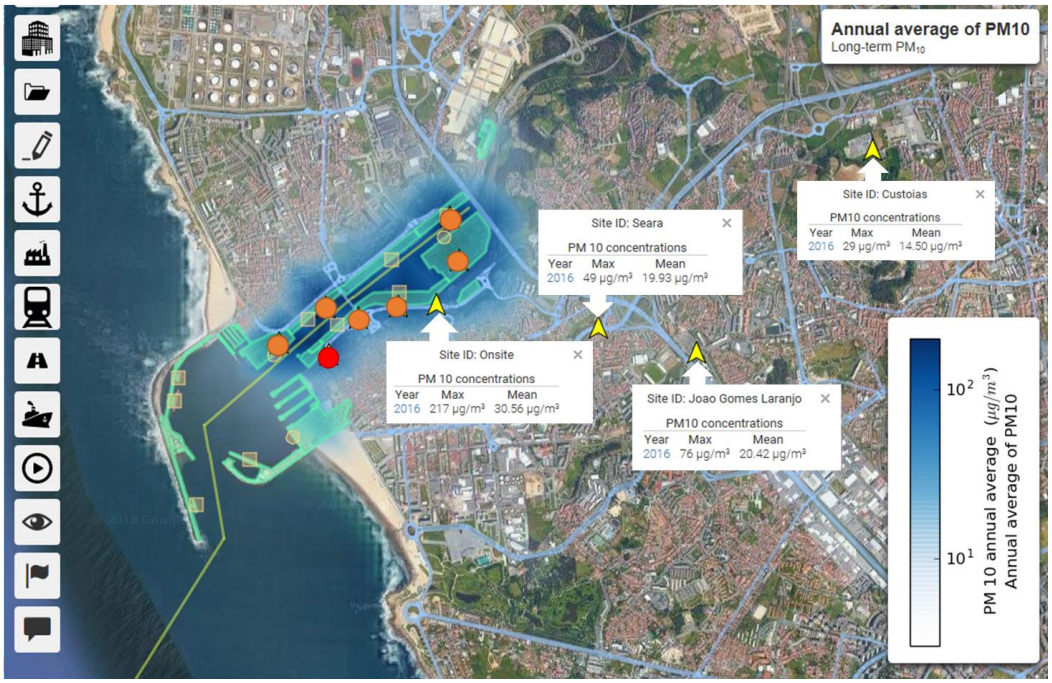


Figure 12. Contour map of the annual average PM10 concentrations obtained with C-PORT tool, comprising: the measured values at the distinct AQS locations (yellow triangles), the receptors with concentrations higher than the annual legal limit value of 40 µg/m^3 (red and orange markers) and docked ships (yellow squares).

Figure 13. Contour map of the annual average NO_X concentrations obtained with C-PORT tool, comprising: the measured values at the distinct AQS locations (yellow triangles), the receptors with concentrations higher than the annual legal limit value of 40 µg/m^3 (red and orange markers) and docked ships (yellow squares).

The highest PM10 concentrations simulated with the C-PORT tool were found within the Port of Leixões area, mainly in the South Container Terminal. The maximum value of PM10 concentrations of 263 µg/m^3 (red marker) was recorded in the receptor located outside the port, first line of habitations. There were six receptors within the entire domain with concentrations higher than the annual legal limit value of 40 µg/m^3 (orange markers), all of them located within the harbour, mainly in the container terminal and solid bulk conventional quays.

The maximum value of NO_X concentrations of 122 µg/m^3 (red marker) was located near the entrance of the southern container terminal and highway. There were twenty-six receptors within the entire domain with concentrations higher than the annual legal limit value of NO_2 - 40 µg/m^3 (approximately 21 ppb) (orange markers), located mainly at the quay and along the highway. European Air Quality Directive 2008/50/EC doesn't establish any limit for NOx for human health protection, this emission source lead to the surpassing of the NO_2 annual limit-value of 40 µg/m^3, which may be used as a reference value (NO_2 is in average 60-70 % of NO_X).

4.4.2. Influence of the meteorological conditions

Sensitivity tests were performed to assess the impact of the meteorological conditions (atmospheric stability, wind direction, sea-breeze circulation) on the pollutants' dispersion patterns, and to further identify the conditions that are responsible for worst-case pollution episodes.

Atmospheric stability condition was one of the key parameters influencing the dispersion phenomena. Therefore, distinct atmospheric stability conditions were tested with the C-PORT tool to determine the conditions more critical or favourable to promote pollutants dispersion over this study area, with particular attention to impacts in the surrounding urban area.

C-PORT simulations were performed for wind directions ranging from West-Southwest to West, corresponding to the typical average conditions of summer season, corresponding to the typical average conditions of summer season, considering all the point sources (i.e., the refinery and the docked ships).

Convective atmospheric stability conditions demonstrated that the refinery plume dispersion follows the horizontal direction because of high dispersion conditions and characterized by an intense vertical mixing. The convective stability class would be the best possible scenario (least pollutant concentration impacting the population) since it was characterized by a greater turbulence capable of dispersing the pollutants quicker, resulting in wider plumes, with lower ground-level concentrations along the average wind direction. On the opposite side, the stable class was characterized by a lower dispersion rate, and it produced greater ground-level concentrations near the emissions source and along the average wind direction. Neutral atmospheric stability conditions represented the plume gradually expanding in the horizontal direction, symmetrically both to the left and right.

Based on results obtained with C-PORT, the atmospheric stability that potentiated higher concentrations, over the port and surrounding urban area, is the slightly stable condition. With the stable class simulation, the refinery plume was not visible, which is probably due to the stack height of 100 m; while hoteling ship emitted smoke at 20 m. Smoke emitted by the refinery was at the top

of the Nocturnal Boundary Layer (NBL) or in the Residual Layer (RL), which under stable atmosphere conditions was rarely dispersed down to the ground because of the limited turbulence.

In coastal areas such as Port of Leixões, the dispersion of pollutants is mainly driven by the local maritime breeze system. There are several studies showing the important role of sea breezes on pollution dispersion (Bouchlaghem et al., 2007; Damato et al., 2003; Ledoux et al., 2006).

To evaluate the impact of the sea breeze circulation on the pollutant's concentration field, the different dispersion fields were simulated with C-PORT, considering all emissions inside harbour (i.e., area and point sources), for daytime (WSW direction) and night-time (NE direction). All these simulations were performed for slightly stable conditions.

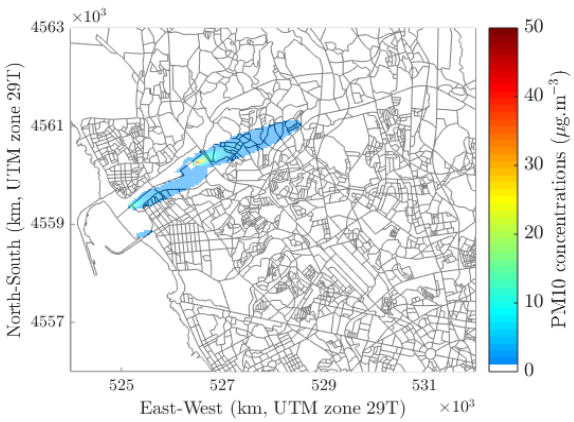
The influence of the sea breeze could be seen with the different spatial patterns of pollutant concentrations over the study area – pollutants emitted from the port during night-time were expected to be dispersed and transported over the sea. Moreover, the rate of dispersion of pollutants emitted during night-time and daytime periods was different due to the differences in wind speed and stability (wind speed was expected to be higher during daytime), as show in Table 7. The highest PM levels were recorded during daytime sea breeze. The lowest NOx dispersion led to hotspots close to the main sources of emissions. The stagnation of air masses over the area favoured the accumulation of pollutants in the surrounding urban area.

4.4.3. Source contribution analysis

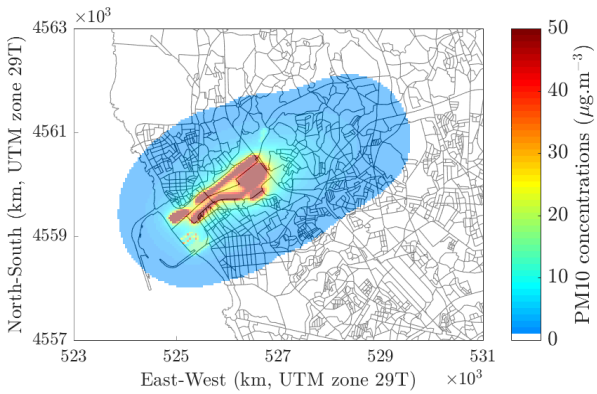
Together with the contour map, C-PORT tool also displayed relative contributions linked with the distinct source categories distributed over the domain, namely marine, land-based, roadways and refinery emissions, both for PM10 and NO_X concentrations.

Figure 14 and Figure 15 show the short-term contributions of the different sources over the study area, to PM10 and NO_X concentrations, respectively, considering WSW wind conditions, daytime, and slightly stable atmospheric class.

(a)



(b)



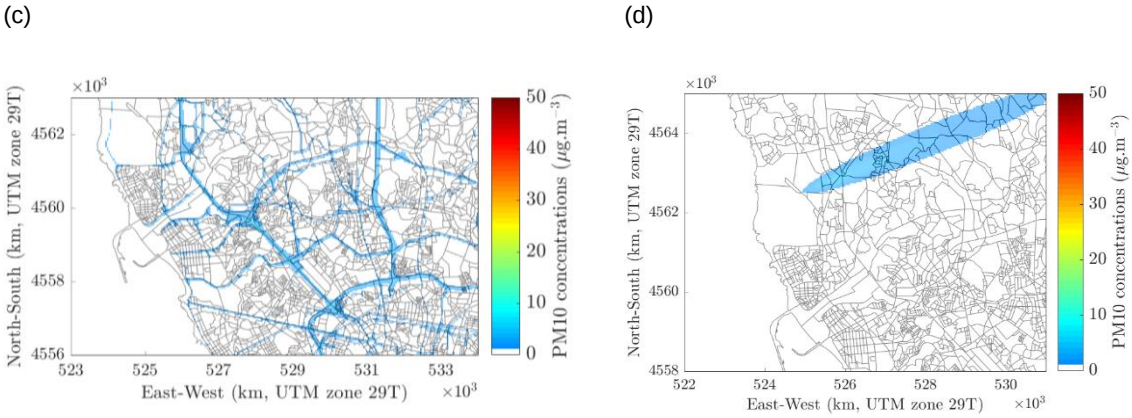


Figure 14. Short-term contributions of the different sources to PM10 concentrations estimated with C-PORT above regional background: a) marine emissions (including ship in transit and point source); b) land-based emissions; c) roadway emissions and d) refinery emission.

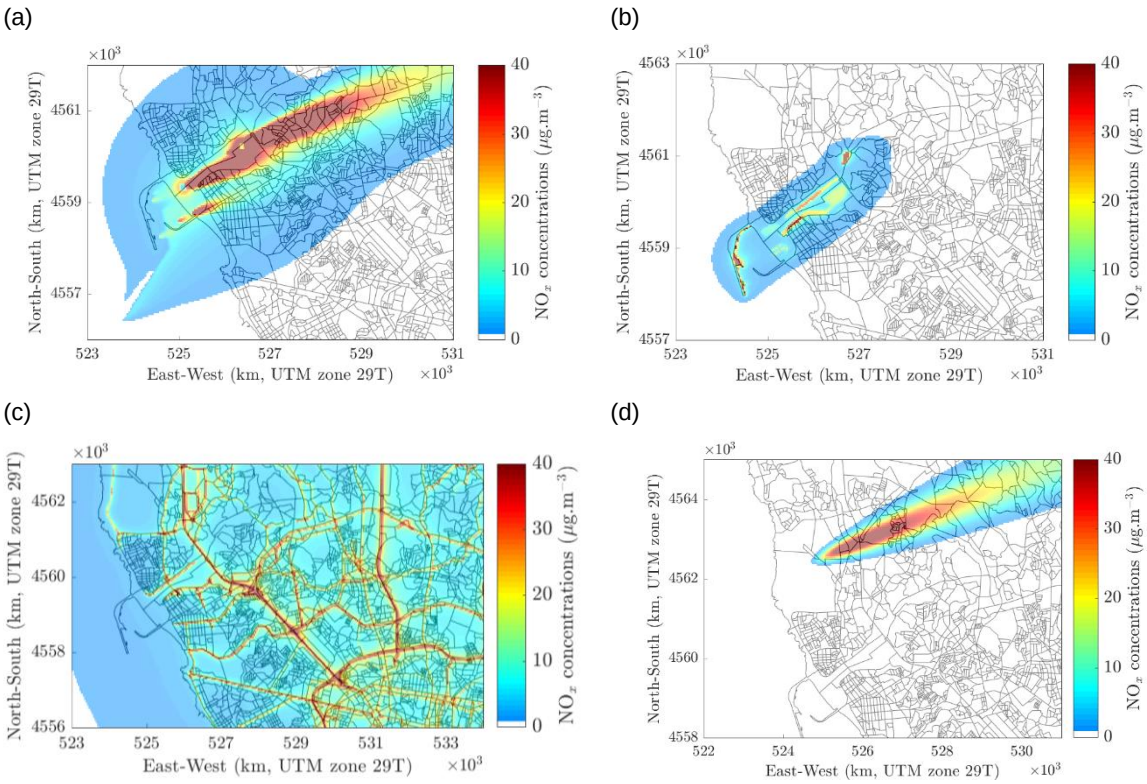


Figure 15. Short-term contributions of the different sources to NO_X concentrations estimated with C-PORT above regional background: a) marine emissions (including ship in transit and point source); b) land-based emissions; c) roadway emissions and d) refinery emission.

All the short-term simulations clearly point towards hoteling ship and refinery industry, indicating that PM10 emissions occurred in very narrow plumes from the stacks. This was since the hoteling ship emitted at a higher altitude (up to 20 m above ground, depending on the stack height). The same behaviour could also be seen in Figure 14(d)) where the emission source (refinery) was emitted at a height of 100 m. As seen in Figure 14(b)), the dispersion does not point towards any specific direction, indicating that PM10 originate from several diffuse sources inside the harbour, including cargo-handling equipment, bulk material transport in trucks and trains and bulk material outdoor storage. All these sources emitted at ground level, causing low dispersion and high concentrations close to the emission sources – i.e., general cargo and solid bulk and container terminal. All short-term simulations for PM10 concentrations point out that area sources exhibited the highest impact over the study area.

Regarding NO_X, C-PORT results suggested that the marine emissions (Figure 15(a)) were the main contributors of NO_X. Although the European Air Quality Directive 2008/50/EC doesn't establish any limit for NOx for human health protection, this emission source lead to the surpassing of the NO_2 hourly limit-value of 200 µg/m^3, which may be used as a reference value (NO_2 is in average 60-70 % of NO_X). NOx effects reached inland at a distance above 5 km from the container quay. The plumes released during docked ships were distinctly visible on NO_X, but the ship in transit inside the harbour had limited effects on NO_X.

The land based, roadway and refinery emissions respect the legal limit, showing maximum NO_X concentrations of 88 µg/m^3 (Figure 15(b)), 142 µg/m^3 (Figure 15(c)) and 56 µg/m^3 (Figure 15(d)).

Figure 16 and Figure 17 show the annual contributions of the different sources over the study area to PM10 and NO_X concentrations, respectively.

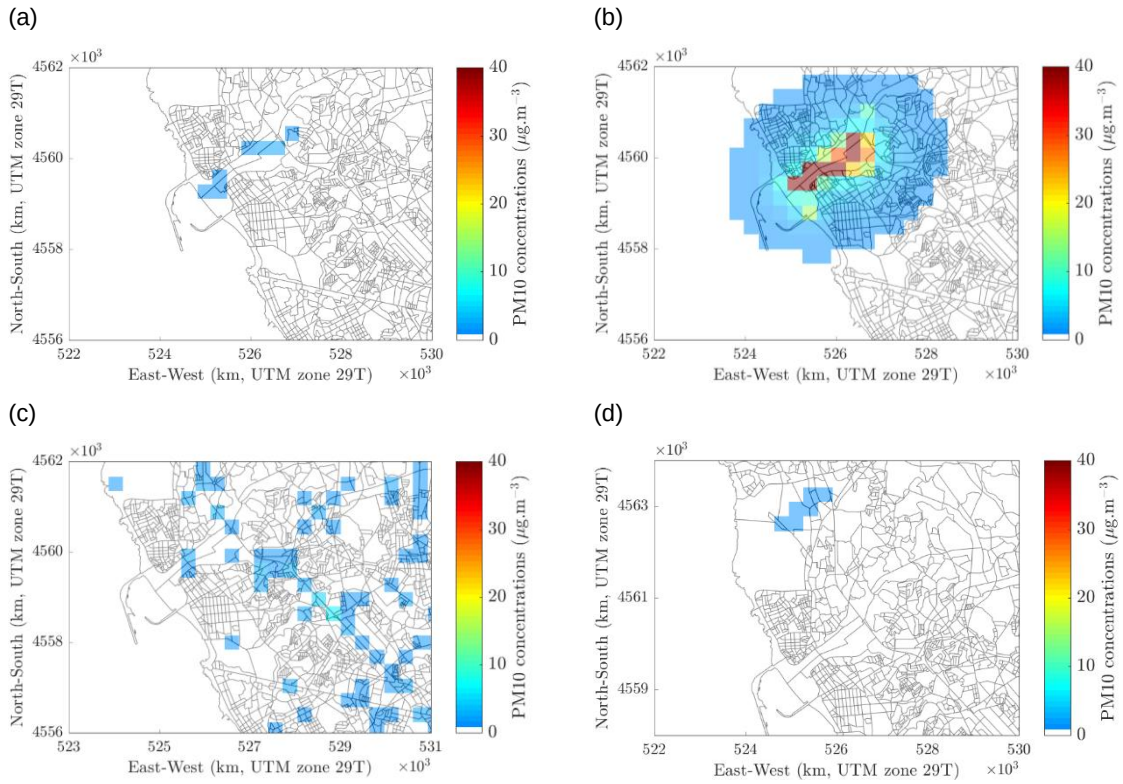


Figure 16. Annual average contributions of different sources to PM10 concentrations estimated with C-PORT above regional background: a) marine emissions (including ship in transit and point source); b) land-based emissions; c) roadway emissions and d) refinery emissions.

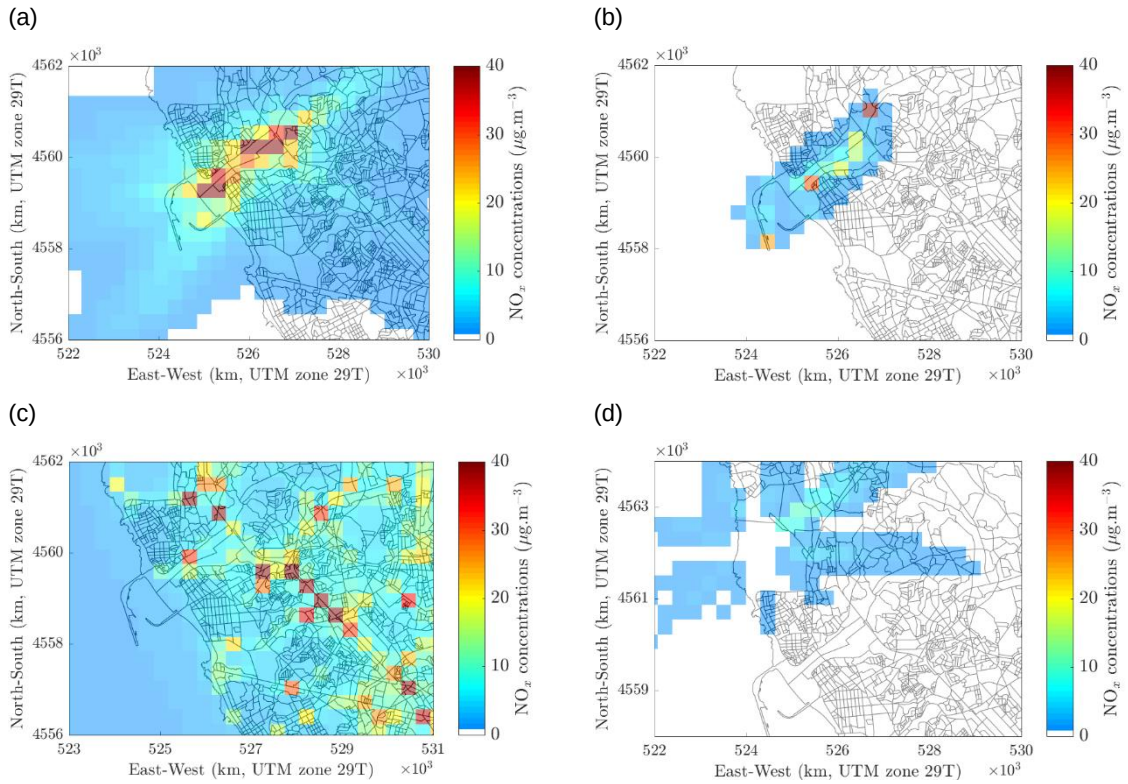


Figure 17. Annual average contributions of different sources to NO_X concentrations estimated with C-PORT above regional background: a) marine emissions (including ship in transit and point source); b) land-based emissions; c) roadway emissions and d) refinery emissions.

From the annual average concentration patterns obtained in Figure 16, C-PORT tool identified the land-based emission sources as the main ones contributing for the total PM10 concentrations, with an estimated contribution of 76-84 %. Roadway and refinery (located close to the port) contributed 3-4 % and <1 %, respectively. Marine activities had a negligible contribution of 27 % from docked ships and < 1 % from ships in transit.

Model results pointed out that these land-based emissions (Figure 16(b)) contributed to a maximum value of PM10 around 260 µg/m^3, with five receptors registering a concentration above the legal limit value of 40 µg/m^3, established by the European Air Quality Directive 2008/50/EC. While both the marine activities and the refinery had a negligible contribution for the PM10 concentrations in the study area. The maritime emissions (Figure 16(a)) led to a maximum of 2.4 µg/m^3 of PM10 concentrations, while the refinery (Figure 16(d)) led to a maximum of 1 µg/m^3 of PM10. Regarding the roadway contribution (Figure 16 (c)), model results exhibited a maximum value of PM10 concentrations around 6 µg/m^3, close to the main routes.

Regarding the NOx concentrations modelled for 2016, docked ships were the main contributors to the higher NOx concentration (55-73 %), followed the by roadway emissions with a contribution between 2035 %. Ships in transit contributed with less than 1 %, which could be justified by the extended hoteling time while the ships were docked. The maximum NOx concentration of 93 µg/m^3 was linked with marine emissions (Figure 17(a)). This value was due to the high number of ships

docked in the south container terminal. Figure 17(a) presents 4 receptors over mainly the channel leading to the Port of Leixões with a concentration above the legal NO_2 annual limit value of 40 µg/m^3, once again used as reference in the absence of a NOx limit value. The maximum concentrations from the land-based sources (Figure 17 (b)) were about 66 µg/m^3.

Regarding land-based emissions source, Figure 17 (b) highlights the existence of two hotspots, in the entrances of the harbour (waterway and terrestrial). Highest land-based emissions sources in this region were centred on the trucks' entrance, used by approximately 830,000 trucks in 2016. Road-traffic (Figure 17(c)) presented a strong influence in the area as an emission source, leading to high values of NOx concentrations close to the main roads. The maximum value of NOx concentration was recorded southeast the port area over the main motorway (A28) and was higher than 90 µg/m^3. Figure 17(c) presents four receptors over the main motorway with a concentration above the NO_2 annual limit value for human health protection (40 µg/m^3) and over 8 receptors inside the study domain. Finally, the annual average plume from the refinery (Figure 17(d)) had a maximum value of 10 µg/m^3.

4.4.4. Population exposure

The population exposure was estimated considering the annual average concentrations of PM10 and NO_X, and the local population distribution by each computational grid cell. The study domain included a total of 374,144 residents.

Figure 18 and Figure 19 show the average population potentially affected by concentrations above 40 µg/m^3 for both PM10 and NO_X. Black squares represent the grid cells with concentration above the respective annual limit values.

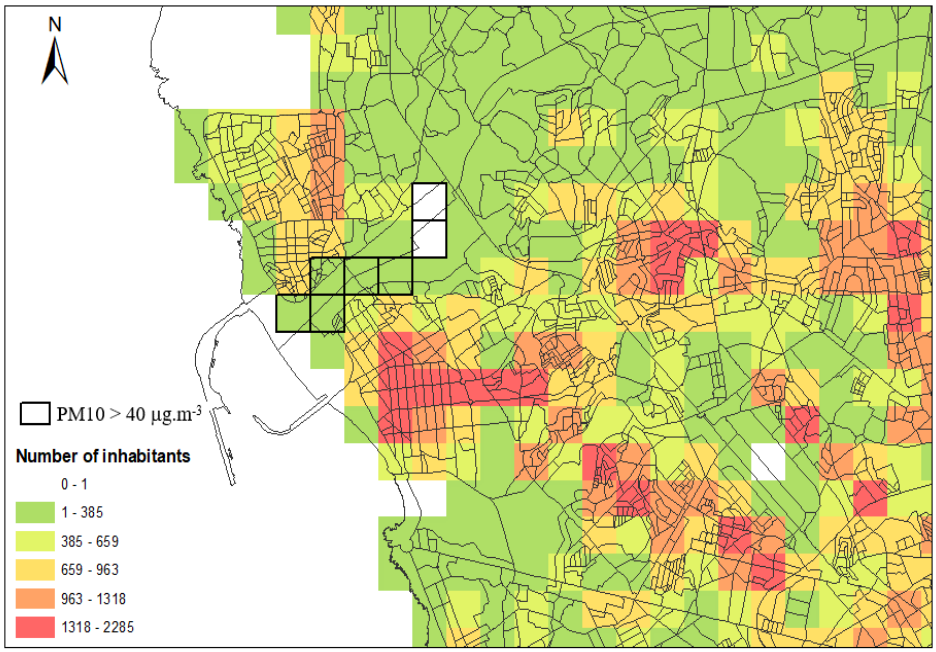


Figure 18. Number of inhabitants and annual average population potentially affected by contributions of all port-related sources of PM10 (marine emissions; land-based emissions; roadway emissions and refinery emissions).

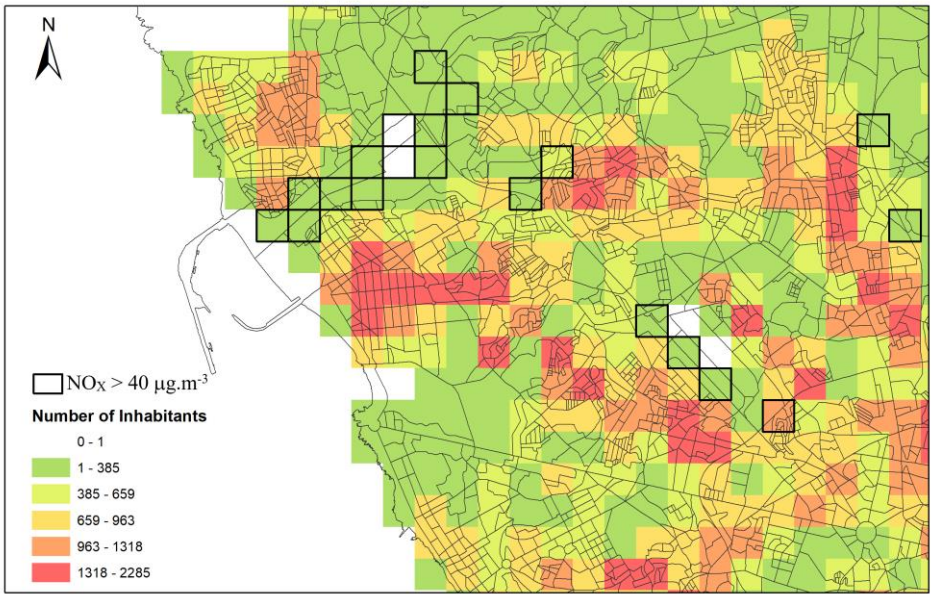


Figure 19. Number of inhabitants and annual average population potentially affected by contributions of all port-related sources of NO_x (marine emissions; land-based emissions; roadway emissions and refinery emissions).

As expected, there were low exposure values for both PM10 and NO_X, since most of the cells that exceeded the legislated limit value were located where there was little population. The population potentially affected by PM10 concentrations above the EU limit value accounted for 567 inhabitants. On the other hand, the population potentially affected by NO_X concentrations above 40 µg/m^3

accounted for 5,657 inhabitants. For both pollutants, cells with concentrations greater than 40 μg/m^3 were located mainly in the port terminals (Figure 18 and Figure 19).

4.5. Conclusions

The Port of Leixões, near the city centre of Porto's metropolitan area, is the largest port infrastructure in the Northern Region of Portugal and one of the most important in the country. The impact on Port of Leixões' air quality was investigated with particular emphasis on the population of the surrounding urban area.

The C-PORT air quality modelling platform, designed to study urban-scale air pollution due to port-related sources, was developed by UNC-IE and U.S. EPA, based on dispersion modelling algorithms, and optimized for rapid execution through an intuitive web-based interface. We expanded C-PORT for a first application outside the U.S., to Port of Leixões in Portugal, characterizing local-scale air quality in and around the port, and performing source apportionment to understand dominant source sectors. First, the impact of different meteorological conditions and the influence of wind directions/sea breeze on local air quality were assessed. Results pointed out that air pollutant dispersion is dependent on meteorological conditions, with slightly stability atmospheric conditions exhibiting the most critical situation for PM10 and NO_X dispersion. The dominant wind direction, a diurnal sea breeze from WSW is responsible for the transport of pollutants over the surrounding urban area. During night-time periods, the dispersion pattern is completely different and promotes the accumulation of pollutants over the port area.

The C-PORT modelling tool was also applied to estimate the relative contributions of various source sectors to outdoor air quality concentrations, including port (terminals, ships, and roads), roadway traffic, and industrial (refinery) sources that potentially affect the port vicinity, including the local urban community.

The land-based emission sources, (including trucks, railways, cargo handling equipment and bulk material stored) at the Port of Leixões exhibit the highest contribution (approximately 80 %) to the levels of PM10 concentrations in the study area. Marine activities and the refinery (located close to the port) have a negligible contribution. Regarding NOx, the docked ships are the main source with a contribution above 50 % for NOx concentration values, with ships in transit contributing below 1 %, justified by the extended hoteling time while the ships are docked.

Future work will include the CFD based modelling of microscale air quality in which the dispersion of the air pollutants will be computed applying a Lagrangian approach. This will identify the obstacles that lead to the formation of additional hot spots from the rearrangement of vertical flow structures, and for a better understanding of the air quality problem in urban hot spots in the immediate vicinity of the harbour.

Chapter 5

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5. Measures to reduce air pollution caused by fugitive dust emissions from harbour activities

5.1. Introduction

Recently, emissions from maritime transport have become an important air quality issue, raising interest among the scientific community, stakeholders, and decision makers, in particular for ports surrounded by high-density residential areas (Contini et al., 2015). According to the European Sea Ports Organization, the top environmental priority for seaports is the local air quality, reflecting its importance on the health of port workers and nearby residents (ESPO 2013). Detailed knowledge about the type of emissions from shipping remains scarce (Healy et al., 2010), encompassed by the lack of experimental data quantifying the contribution of ship's emissions to local air quality (Borrego et al., 2007; Isakson et al., 2001). Marine ships' emissions come primarily from diesel engines operating on oceangoing ships, tugs and tows, dredges, and other ships operating within a port area. Land-based emission sources include essentially cargo handling equipment. Emissions from cargo unloading and handling activities are also considered, since they have an important contribution for the total emissions (Song and Shon, 2014).

Petroleum coke (petcoke) is a granular residue that poses considerable threat to the environment and human health. It is a dark solid material composed primarily of carbon, usually showing limited amounts of elemental forms of sulphur, non-volatile inorganic compounds, and heavy metals, such as V or Ni (McKee et al., 2014). Petcoke is generally stable under normal conditions and chemically inert; however, it has the potential to become flammable or explosive in enclosed spaces (Caruso et al., 2015). In 2014, petcoke accounted for almost 30 % of the materials loaded, unloaded, and handled in the North Portuguese harbours. Due to its relatively low cost when compared to coal, it supplies energy for industrial combustion processes such as cement manufacturing (Dourson et al., 2016). There is a growing global demand for the supply of petcoke in a vast array of industries. Thus, outdoor storage in stockpiles in harbours is becoming a common practice, even in harbours located

in the surroundings of urban communities. In such cases, fugitive dust emissions from petcoke endanger the local population's health (Dourson et al., 2016). Moreover, petcoke particles may also have negative impact on buildings (Dourson et al., 2016). There is still a lack of understanding concerning health and environmental effects of fugitive petcoke dust and most authors agree that further investigation is an important requirement (e.g., (Dourson et al., 2016), especially considering that petcoke production and usage continue to increase. Toxicological studies performed recently suggested that petcoke acts generally, as an inert dust with human health risks associated to high-level or long-term dust inhalation, such as chronic pulmonary inflammation or respiratory tract irritation (Caruso et al., 2015; McKee et al., 2014). According to the EEA (2013) components of PM such as heavy metals or PAH, which are contained in petcoke, are known carcinogens and directly toxic to living cells. (Keil et al., 2016) reported a reduction of the immunoglobulin-M antibody production, while (Salnikow et al., 2004) found that the inhalation of Ni present in petcoke and residual oil fly ash leads to hypoxic stress and induces the secretion of pro-inflammatory cytokine.

Studies performed by the US Environmental Protection Agency have reported that petcoke dust has negligible biodegradation in soils, atmospheric photo-oxidation, and volatilization, being also poorly soluble in aquatic environments with neutral pH (USEPA 2015). Caruso et al. (2015) studied the effect of pelleted petcoke in water (at 1 g/L loadings) and reported a 7.1 % inhibition on algae growth, although no adverse effects on aquatic invertebrates or fish were recorded. Increase in the concentration of heavy metals in the aquatic environment due to petcoke leaching was also reported, with noteworthy effects to local aquatic species (Caruso et al., 2015; Jinnagara Puttaswamy et al., 2014). Baker et al. (2012) analysed trace metals in wetlands constructed using petcoke and other consolidated waste sediments in the Alberta tar sands (Canada), founding high concentrations of Ni and V, attributed to petcoke, at levels that are toxic to local invertebrate species. Their results supported the findings of Puttaswamy et al. (2014), who found high levels of Ni and V in leachates collected from shallow and deep lysimeters in the same region. Both lysimeters were buried in petcoke and covered in glacial till (deep lysimeter) or peat (shallow lysimeter), and the concentrations measured were toxic to freshwater species.

Petcoke's dispersion rate depends not only on the meteorological conditions of the storage place but also on the intrinsic characteristics of the material, namely moisture content, size, density, and mechanical properties (Dourson et al., 2016).

Fugitive petcoke dust emissions can be minimized through the application of obstacles, such as walls, fences, or shrubs, upwind from the petcoke piles, which is a realistic and relatively low-cost approach in reducing emissions. Novak et al. 2015 suggested that changing pile arrangement space as well as pile configuration could reduce dust emissions in the open storage yard. Increasing of the particles' moisture content may also reduce fugitive dust emissions.

This work aimed to find control measures that can mitigate fugitive dust petcoke emissions from harbour activities. The main goal was to find a way to minimize the impact of those petcoke emissions on the communities in the harbours' neighbourhood. A real situation involving a Portuguese port area

was studied, focusing on the implementation of real scale control measures, which is one of the main features of this work.

The work is structured as follows: Section 5.2.2 describes the case study framework; Section 5.2.3 mitigation and management measures; Section 5.2.4 and Section 5.2.5. describe the study of the type of barrier and its optimum location; assessment of local air quality using a CFD model is discussed in Section 5.2.7. The summary and conclusions are given in Section 5.2.8.

5.2. Case-study framework

With 350 m of quays and 15 ha of embankments, the Solid Bulk Terminal (SBT) at the port of Aveiro is prepared with appropriate handling equipment with crane capacity up to 120 tonnes. A movement of about 2 million tonnes is recorded, with large areas dedicated to uncovered and covered storage of materials. In 2014, the Administration of the Port of Aveiro (APA) handled 424 ton of petcoke in its SBT, which corresponds to a 1.5 % increase when compared to the previous year. Due to its proximity to one of Portugal's main cement production plants, Port of Aveiro's SBT is one of the country's major petcoke entry points. The port is located in the vicinity of Aveiro, within the town of Gafanha da Nazaré, which has a population around 15000 inhabitants. The nearest residential area is located at approximately 1 km to the south of the Port of Aveiro's SBT. The location of the SBT, the urban community, and the measures points is shown in Figure 20

Figure 20. Map of the study area, with the town of Gafanha da Nazaré. The blue dot indicates the SGT, the red dotes P1, P2 and P3 indicate the air quality measurement points. The red square indicates the residential area located southeast the seaport included in the modelling study, and the blue square indicates the industrial area.

To characterize the typical meteorological conditions of the studied area, an analysis of meteorological data (namely wind data) was performed using hourly data from an 8-year period (2006-2013) recorded at the University of Aveiro's meteorological tower.

This meteorological tower is located roughly 5 km southwest from the studied area. This tower is representative of the surrounding environment (25 km), belonging to the national meteorological monitoring network. Figure 21 shows the wind rose obtained from the data measured at 10 m height.

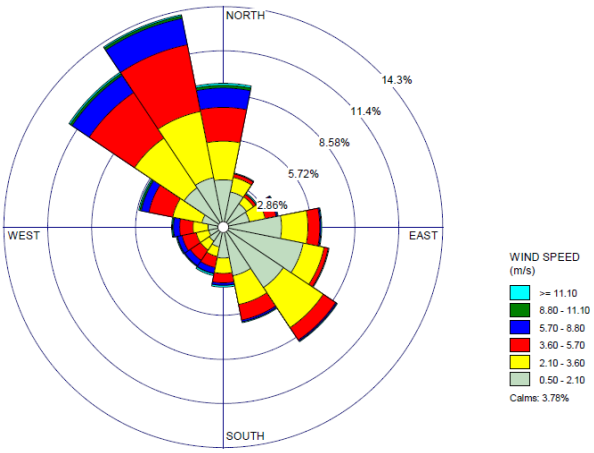


Figure 21. Wind rose for 2006-2013, obtained from measurements performed at 10 m height in the Meteorological Tower of the University of Aveiro.

Typically, the wind blows from the second and fourth quadrant, mainly from Northwest (NW), North (N) and Southeast (SE) directions. Mean wind speeds range typically from 0.5 m/s up to 11 m/s. N and NW winds were recorded during approximately 35 % of the measurement time. The nearest urban communities to the Port of Aveiro are located South (S) and Southeast (SE) from the port, thus the wind directions that may affect the populations are in the 4th quadrant (W-N quadrant).

Experiment this work were performed considering the typical wind directions registered in Aveiro to determine the position and dimension of the solid barrier that would lead to the greatest minimization of petcoke dispersion. Different spatial configurations were tested in the WT, as well as a reference scenario, for three wind directions (N, S and NW), and speeds (3, 7 and 11 m/s).

5.3. Mitigation and management measures

Several technical and management measures can be adopted to reduce emissions and control the dispersion of air pollutants into the atmosphere. Nowadays, several ports and harbours have adopted appropriated environmental management plans to mitigate the adverse impacts of its activities. The control and/or reduction of dust and particulate matter emissions from petcoke at the port of Aveiro needs to consider the different operations involved from arrival to departure of the material: the unloading activity using a closed claw; the outdoor storage in piles, the loading of trucks or trains using tractor shovel, and the transport to its destination in heavy trucks or trains.

The following actions could be considered additionally to the construction of fixed barriers:

- To spray of water over the bulk material to control the fugitive emissions during loading and unloading processes;
- To enclose the sources either fully or partially, if possible. In the case of Aveiro's port, covering the piles could prevent the dispersion of particulate matter;
- To reduce the time of outdoor storage;
- To transport bulk material in closed trucks and trains to avoid wind entrainment;
- To clean regularly the paved and unpaved roads to reduce resuspension of dust;
- To promote the increase of green spaces all around the port area to reduce air as well as noise pollution.

Regular monitoring of air quality should be further strengthened to evaluate the effectiveness of air quality management plans. Simultaneously meteorological monitoring in the same sites could help to better understand local dispersion patterns and impacts in the surrounding areas. To reduce the impact of petcoke dust emissions on local air quality, even before the implementation of the selected barrier configuration, the APA implemented several good practices such as: i) spray of water over the bulk material unloading from ships and loading heavy trucks; ii) reduce the time of petcoke storage outdoors to decrease the rate of fugitive dust emissions; iii) clean ground surfaces after temporary storage material has been removed and transported to final destination; iv) reduced the size of the pile to reduce exposure to high wind speeds, and v) alter the way petcoke is offloaded, releasing the petcoke closer to the ground.

5.4. Air quality monitoring

Over the last years, the residents of this urban community have reported high quantities of black dust in and around their residences, during handling and storage of petcoke in the SBT. These episodes lead to the necessity of evaluating the impacts of the different port activities in the surrounding environment. To assess the contribution of petcoke unloading and handling operations to the air pollution problems in the residential area near the port of Aveiro, a 90-days monitoring campaign was set up. The campaign performed by the Institute of Development and Environment included three air quality monitoring periods (30 days each): one during the winter (December 19th 2014 to January 20th 2015), one in the summer (July 2nd to August 1st 2014) and one in the spring (May 5th to June 4th 2015). During the three measurement periods, several arrivals of cargo ships carrying petcoke were registered, meaning that petcoke was handled in the SBT during about 70 % of the measurement time. Each cargo ship transported between 6000 and 10000 tonnes of petcoke, which were unloaded and stored in stockpiles at the SBT. PM10 measurements showed that the daily concentration limit of 50 $\mu g/m^3$, established by the European Air Quality Directive 2008/50/EC, was surpassed in 21 of the 90 monitoring days (Sorte et al. 2018). According to the legislation, concentration of PM10 may exceed the legal limit a maximum of 35 times per year. Considering the sampling period, the number of exceedances of the daily limit to the human health protection that

would be critical was computed to be 8. This means that the established daily limit value of 50 µg/m^3 should not be surpassed more than 8 times to ensure proper human health protection. This limit was exceeded in all measurement periods. Moreover, the average concentration of PM10 was 45 µg/m^3, indicating potential influence of Port activities. Exceedances of the limit value for PM10 concentration matched the petcoke movements in the SBT; this campaign was conducted under the influence of dominant Northwest winds. This was an important feature since the most common wind directions that affect the residential area blow from the West-North quadrant. Thus, the monitoring campaign was performed under the most critical situation in terms of dispersion of particles reaching the port's neighbour urban area (IDAD 2015).

During the measurement campaigns, the levels of V and Ni were also monitored. The highest average concentrations of 3.9 ng/m^3 (V) and 10.4 ng/m^3 (Ni) were reported at the closest location from the SBT during the winter measurement period (predominant winds from the SE direction).

Although there is a correlation between the PM10 concentrations and petcoke movement activities, monitoring of Ni and V was inconclusive in this matter. Mean concentrations registered for V were very close to levels reported in urban background areas of Spain (Moreno et al., 2007; Reche et al., 2011). Moreover, measured concentrations of V and Ni were below the respective limit and target values defined in the legislation.

5.5. Studying the type of barrier

To assess the implementation of a barrier upwind from the pile as a measure to block petcoke dispersion, qualitative and quantitative assessments of the petcoke emission for different types of barriers were performed. Qualitative observation was used as a pre-test, prior to the quantitative measurement of petcoke emission for different scenarios. Its main goal was to characterize the dispersion behaviour of the petcoke particles, namely in terms of the minimal dispersion velocity and size of the most easily transported particles. Speeds of 1 to 11 m/s were tested in the WT with increments of 1 m/s.

The open-circuit wind tunnel used for this study was 12 m long and had a model test section of 6.5 length x 1.5 m width x 1 m height. It has been previously used for physical modelling of urban flows and air quality in several studies (e.g., Borrego et al. 2007). When applicable, the guidelines proposed by the German Engineering Association for atmospheric boundary layer simulation in wind tunnels were followed for neutral stability conditions. The wind tunnel was designed to generate a uniform flow field through the test section. The atmospheric boundary layer was simulated using a specific setup of turbulence generators and floor roughness elements upstream of the test section. Vertical wind velocity profiles of the simulated atmospheric boundary layer were measured by means of a Pitot tube. The mean velocity profiles were measured at the petcoke pile's location (in the centre of the test section). The obtained wind velocity profiles agreed well with the power law profile when α=0.19, which corresponds to the velocity profile of a lagoon area (Avelar et al. 2012).

Petcoke stockpiles were created with the equivalent configuration to the ones in the SBT, simulating discharges of about 10,000 tonnes of petcoke. The dimension of each stockpile was scaled at 1/127 to represent a size of 7 m diameter x 10 m height (see Figure 22). To create the stockpiles for the tests, air-dried low granulometry petcoke samples were used, since these have the greatest dispersion potential.

(a) (b)

Figure 22. (a) Image of a real petcoke stockpile taken at the SBT, (b) Perspective of a typical petcock pile used in the wind tunnel experiments.

For this study, two types of obstacles were investigated: (i) a solid barrier, to simulate the effects of a windbreaker; and (ii) porous barriers, to simulate the effects of pierced fences/shrubs (barrier placed at the WT surface, Z=0 m) or trees (barrier placed 2 cm above WT surface, corresponding to Z=2 m in the field). Both types of barriers were placed 10 cm (10 m in the field) upwind from the petcoke pile, to reduce incident wind's intensity.

Figure 23 (a) shows the setup for the experiments performed with the solid barrier, while Figure 23b shows the setup of the experiments performed with the porous barrier (in this photograph, for Z=2 m). All tested barriers had a height of 10 cm (10 m in the field), which was the same of the petcoke stockpile

(a) (b)

Figure 23. (a) Solid and (b) Porous barriers used in the wind tunnel experiments (Z=2 m).

For the different barrier setups and the reference scenario, a set of experiments were carried out accounting for different wind directions (N, S and NW) and speeds (3, 7 and 11 m/s). As said, these variables represent typical wind conditions and the most critical conditions for the transport of

petcoke particles to the residential area. All the experiments had a duration of 10 minutes, which is common in wind tunnel experiments to capture the extremes of the wind speed record (Wainwright and Mulligan 2004). In these experiments, the petcoke pile weight and height were measured before and after the experiment. The emission of petcoke particles was computed from this difference (see Eq. 9), considering that the wind was the only agent responsible for dragging particles out of the pile and transport them downstream). The weight of the petcoke pile before and after the experiment was determined in an electronic balance (± 0.00001 g precision).

$$E = \frac{(w_i - w_f)}{w_i} \tag{9}$$

where E are the emissions (kg), w_i is the initial weight (kg) and w_f is the final weight (kg).

Emission reduction was determined comparing the emission obtained in the different scenarios with barrier with the one obtained for a reference scenario with no barrier.

Table 9 presents the results of the WT experiments for different types of barriers, in terms of reduction of pile height and mass emission. All types of barriers were placed 10 cm upstream of the pile (10 m in the field).

Table 9. Petcoke emission reduction and pile height reduction (%) for the different types of barriers tested, against the reference scenario with no barrier.

Obstacle	Wind velocity (m/s)	Pile height reduction (%)	Emission reduction (%)
Solid barrier	3	1.05 ± 0.28	98.78 ± 0.54
	7	3.16 ± 0.13	84.43 ±0.79
	11	4.00 ± 0.32	85.99 ± 0.63
Porous barrier Z=0 m	3	1.05 ± 0.44	93.00 ± 0.26
	7	2.11 ± 0.36	78.44 ± 0.69
	11	4.12 ± 0.49	80.61 ± 0.20
Porous barrier Z=2 m	3	1.05 ± 0.33	19.76 ± 0.86
	7	2.13 ± 0.12	18.96 ± 0.55
	11	2.13 ± 0.25	64.59 ± 0.45

All types of barriers tested reduced the transport of petcoke particles, with the solid barrier showing higher efficiency. The largest reduction (about 99 %) was registered for the lower wind velocity. The highest emission reduction obtained with porous barriers was recorded for 3 m/s winds, when the porous barrier was placed on the wind tunnel surface. For Z=2 m, emission reduction increased with higher wind velocity (maximum of approximately 65 % for 11 m/s wind), because the drag force exerted by the elevated porous barrier was greater in this case than when lower wind speeds were tested. Consequently, the number of edges increased around the barrier, with increasing vorticity, but tended to decrease around the stockpile. The turbulence was highly reduced around the pile for higher wind speeds, which helps explaining these results. The outcome of this set of experiments suggested that a solid barrier upwind from the pile should be chosen as method for reducing petcoke particles' dispersion. The results obtained were in accordance with APA's request, which sought an efficient, low-cost solution for the dispersion of particles. The main advantage of this solution was

the availability of containers within the harbour, which could be used to build the barrier. In addition, APA decided to alter the dimension of the stockpiles used in the SBT to decrease exposure to wind.

5.6. Studying the location and dimensions of the barrier

After assessing which type of barrier would be more effective, a new set of experiments was conducted in the WT aiming to optimize the position and dimensions of the barrier. Several simulations were performed to determine the main characteristics, i.e., dimensions and configuration, of the barrier that would lead to an effective minimization of petcoke emission to the atmosphere. Six different spatial configurations were tested in the wind tunnel for placement of the barrier. The position and dimension of the barrier for each configuration ("A" to "F") is represented in Figure 24.

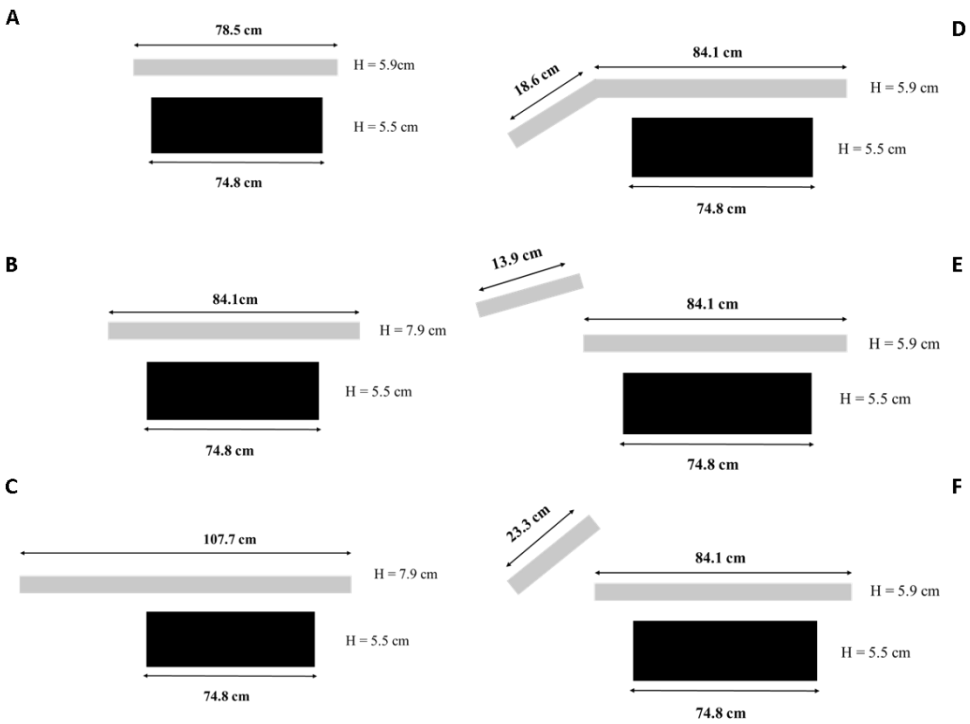


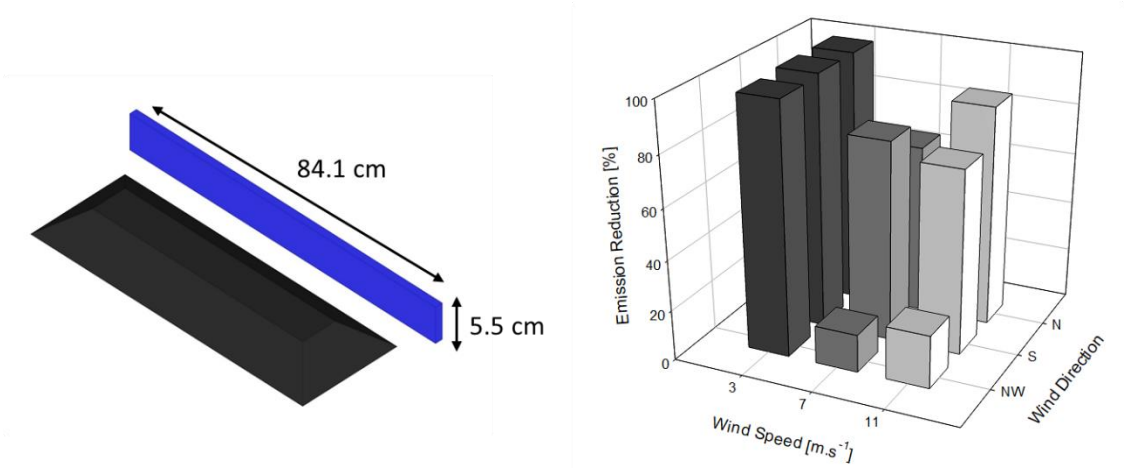
Figure 24. Schematic representation for the six different barrier configurations tested and their respective dimensions (black – stockpile and grey - barrier). The thickness of the barrier was 1.9 cm in all configurations.

For each of these configurations, a set of different simulations were carried on accounting for different wind directions (N, S and NW) and wind speeds (approx. 3, 7 and 11 m/s). These variables represent typical wind conditions and the most critical conditions for the transport of petcoke particles to the residential area.

Each experiment had the duration of 10 minutes and the petcoke pile was weighted before and after the wind tunnel simulation. The petcoke fugitive emissions were obtained by the weight difference, considering the wind the only responsible agent for dragging particles out of the pile and for its transport downstream. The emission reduction was determined from the loss of pile weight

registered. The mass emission rates used were those calculated for the worst-case scenario in terms of potential emissions, i.e., the moisture content below 1 % (0.59 % ± 0.066).

In a first step, the behaviour of the pile was evaluated in the presence of a single barrier, with: (i) different barrier heights (with 3, 4 and 5 containers in height, corresponding to 5.9 cm, 7.9 cm, and 9.8 cm, respectively); (ii) different barrier extensions (74.8 cm, 78.5 cm, 84.1 cm, 107.7 cm); (iii) different positioning of the barrier (upstream and downstream of the pile). From these experiences, the schematic configuration shown in Figure 25(a) was the most efficient configuration. From now on this configuration will be named “configuration A”.



(a) (b)

Figure 25. (a) Schematic representation for the barrier configuration A tested and its dimensions at 1/127 scale (petcoke pile in black, barrier in blue); (b) Petcoke emission reduction (%), considering different wind conditions.

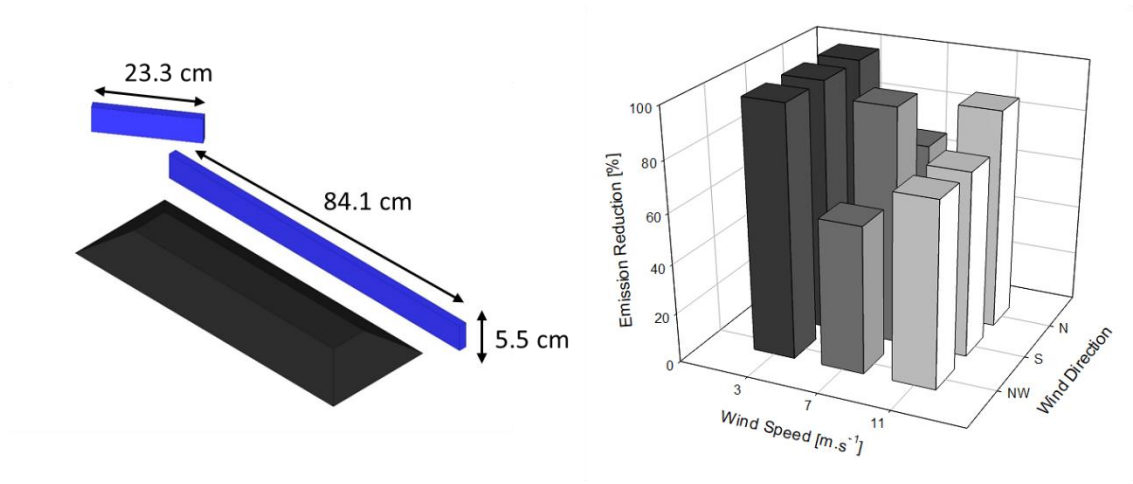
Results obtained with configuration A showed that using an upstream barrier causes a positive impact on petcoke dust emissions, comparing with placing a barrier downstream. The best results were obtained for an 84.1 cm length x 5.9 cm height barrier (1:127 scale).

Placement of the barrier with this configuration proved to be particularly efficient for N and S winds, achieving emission reductions of 62 % and 66 % for 7 m/s wind speed and 79 % and 59 % for 11 m/s wind speed, respectively. For the NW wind speed scenarios, results were somehow lower than for N direction: only about 25 % reduction of petcoke emission for wind speed higher than 3 m/s.

In a second step, the focus was on the optimization of a solution that included a secondary barrier, towards NW direction. With this goal, a new set of experiments was performed to test: (i) different positioning of this secondary barrier in relation to the main one (different angles between barriers); (ii) different barrier extensions (23.3 cm, 13.9 cm, 18.6 cm, 30.0 cm) and (iii) different distances between both barriers. In this set of experiments the distance between the petcoke stockpile and the tested barriers varied as function of the stockpile’s dimension: for 6000 tonnes stockpiles, the main barrier was placed 8.4 cm from the pile (10.7 m in the field); for 10,000 ton stockpiles, the main barrier was distanced from the pile 4.5 cm (5.7 m in the field). These dimensions corresponded to the

maximum distance allowed by the APA, to maintain the operational space needed for the movement of trucks and trains.

Results showed that the use of an upwind solid barrier led to a very positive impact on petcoke dust emissions, with considerable reduction for both tested configurations (up to 100 % in some cases). The most efficient configuration for the double barrier consisted of a main barrier of 84 cm length (109 m in the field) plus a secondary barrier of 23 cm length (30 m in the field). This configuration was always at least as efficient as configuration A and will be named from now on as “Configuration B” (see Figure 26(b)).



(a) (b)

Figure 26. (a) Schematic representation for the barrier configuration B and its respective dimensions at 1/127 scale (petcoke stockpile in black, barrier in blue); (b) Petcoke emission reduction (%) obtained with this configuration of barrier

For the highest speed and N wind direction, which represents one of the most registered conditions in this region, 88 % emission reduction was achieved for the double barrier scenario (B). Moreover, considering this higher wind speed, the best results were achieved exactly for the N direction wind, which is a very promising result in terms of reducing fugitive particles endangering the residential neighbourhood (only 5 % of the pile was dragged by the wind).

Considering that N and NW are the most endangering directions for the population, it is especially worth noting that, for the higher speeds tested, the configuration B produced always higher emission reductions then configuration A, proving the importance of adding the secondary barrier. For 11 m/s NW winds, the difference between efficiencies was the greatest: configuration B caused a reduction more than three times higher than configuration A. For S wind, the double barrier also had a positive effect in reducing dispersion of dust particles.

Figure 9 present some images taken after the wind tunnel experiments, for configurations A and B, for the 3, 7 and 11 m/s (left to right, respectively), and NW wind conditions.

(a)

(b)

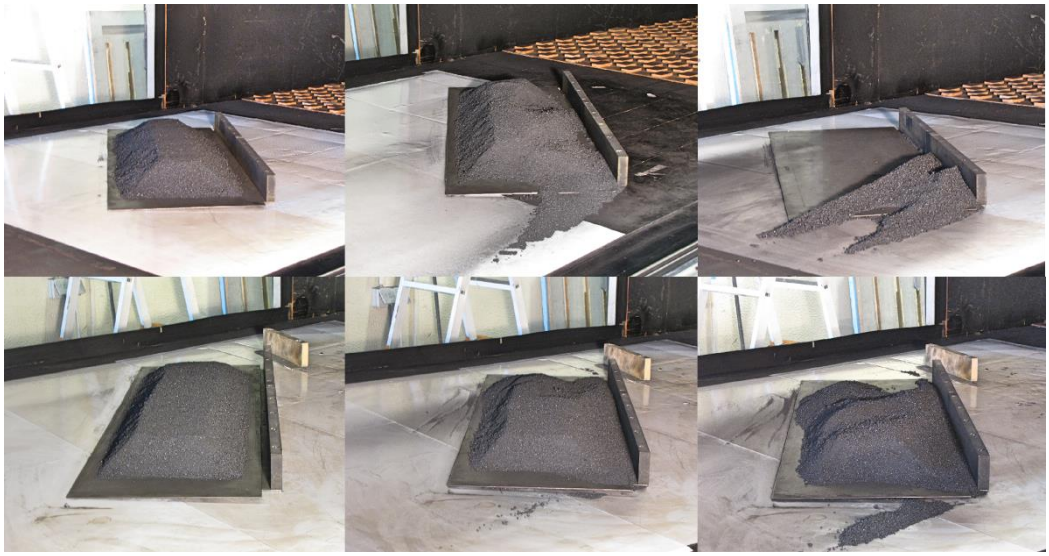


Figure 27. Images of the two barrier configurations tested for NW winds of 3, 7 and 11 m/s speed (left to right): (a) Southeast perspective of the petcoke pile for barrier configuration A placement experiments; (b) Southeast perspective of the petcoke pile for barrier configuration B placement experiments.

It is worth noting that the described experimental study was performed using very low petcoke moisture contents (always below 1 % weight after air-drying), which leads to higher emissions.

5.7. CFD numerical modelling

In this study, the commercial Ansys Fluent CFD software was applied to a set of baseline numerical simulations. These baseline simulations were performed to assess the impacts of storage and transport of petcoke on local air quality, allowing a spatial extended characterization of the air quality problem, previously identified by the population. Furthermore, the CFD model was used to perform a set of simulations – mitigation scenarios – to assess the effectiveness of the implementation of an upstream barrier from the petcoke stockpile.

The Ansys Fluent is a multi-purpose commercial CFD software tool available, including well-validated physical models able to provide accurate results across a wide range of multidisciplinary physics application. Ansys Fluent has been applied in the simulation of flow and dispersion of fluids and particles within confined and open complex geometries in different scientific and technical domains. The Ansys Fluent suitability for urban air quality modelling was previously evaluated (Borrego et al., 2003, Martins et al., 2009). In this study, the Ansys Fluent was applied to the numerical simulation of the turbulent flow dynamics and the dispersion of petcoke fugitive dust emissions, using the Reynolds-Averaged Navier-Stokes (RANS) approach with a first order k-ε turbulence closure scheme. The numerical simulations were performed with an hourly time basis assuming steady-state flow conditions. The inflow and boundary conditions were defined based on the typical meteorological data discussed in Section 5.2.2. The inflow conditions were defined using the theoretical log-wind profile.

5.7.1. Baseline simulations

The computational domain was designed using the Ansys Fluent processor applying an unstructured mesh. In the baseline simulations, the computational domain encompasses the SGT terminal, as well as a first row of buildings of the residential area, located nearest the terminal. The dimensions of the computational domain are 3 km x 2.5 km, and the grid resolution is 1 m x 1 m. The main goal of the baseline CFD numerical simulations was to assess the impacts of particulate matter dispersion from the petcoke stockpile on local air quality. For that purpose, a set of numerical simulations of the turbulent flow dynamics were carried out considering two distinct wind directions, North and Northwest, and several wind speeds ranging from very low wind speed conditions (1.3 m/s) to high wind speed conditions (11 m/s). After the flow dynamics simulation, the transport of particles from the petcoke stockpile was evaluated applying the Lagrangian particle tracking approach available on Ansys Fluent. The baseline simulation results show that wind blowing from North directions lead to highest concentrations downstream near the petcoke stockpile, while Northwest directions conduct to transport of particles to the industrial area. Table 10 presents the maximum concentrations obtained within the computational domain, the residential area, and the industrial area.

Table 10 Maximum concentrations obtained within the computational domain, the residential and the industrial area.

Wind speed (m/s)	Wind direction	Maximum concentration (µg/m^3)		
		Domain	Residential	Industrial
1.3	NW	100	<1	12
11	NW	500	<1	19
1.3	N	300	9	<1
11	N	600	9	<1

The petcoke fugitive emissions are driven by the turbulent flow downstream the pile along the wake over the recirculation areas. The very low wind speeds found in these areas promote the accumulation of particles. Thus, the area within the domain recording the maximum concentration is located downstream near the petcoke pile. The contribution to the local air quality in the surroundings of the seaport ranges from 9 µg/m^3 in the residential areas up to 19 µg/m^3 in the industrial areas. Additionally, the numerical results indicate that North winds drive the particulate matter to the residential area, while Northwest winds conduct the particles to the industrial area. Noteworthy, North wind direction is substantially unfavourable to particles dispersion around the piles, and this may be critical for the exposure of harbour staff.

5.7.2. Mitigation scenarios simulations

To evaluate the effectiveness of the implementation of an upstream barrier from the petcoke stockpile, the CFD model FLUENT was used to perform a new set of numerical simulations. A new computational domain of 500 m x 500 m includes the petcoke stockpile and the upstream barrier with

the configuration F of Figure 27 – the optimal configuration identified by the wind tunnel simulations. Figure 5 shows, as an example, the top view of particles' trajectories speed within the computational domain for the wind blowing from NW with a wind speed of 11 m/s.

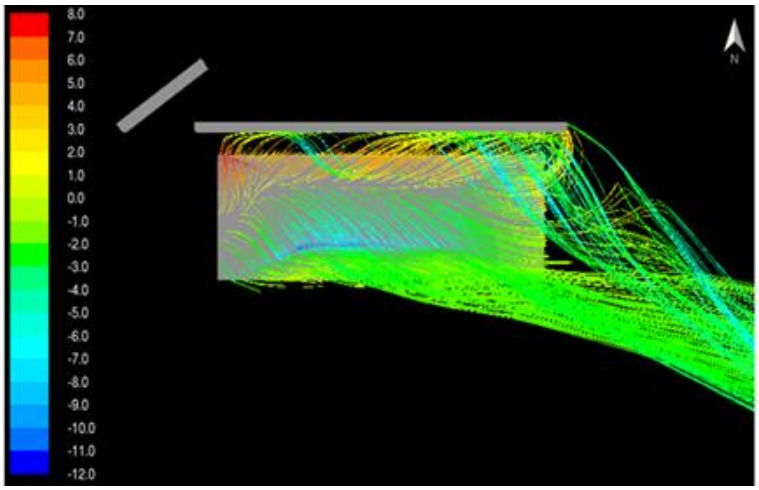


Figure 28. Top view of particles' trajectories for the simulation with the upstream barrier from the pile with Northwest direction and wind speed of 11 m/s as inflow conditions. The colour scale represents the trajectories' speed in m/s.

The effectiveness of the barrier implementation is quantified by comparing the reduction of the petcoke dust transported from the stockpile in the mitigation scenarios simulations compared with the baseline results. The implementation of an upstream barrier from the petcoke stockpile reduces the wind intensity over and around the pile, leading to a reduction of particle matter transported of approximately 68 %, in the case of high wind speed (11 m/s), either from North or Northwest directions. The numerical results are in good agreement with the physical results, since in Section 5.2.5. is presented a reduction of 88 % for North and 74 % for Northwest directions. The implementation of the upstream barrier led to an effective reduction of the numerical particles transported up to the residential area, for both wind direction situations. Following this, the numerical results confirm the implementation of the upstream barrier as an optimum mitigation measure to protect the urban community located south from the SGT.

5.8. Conclusions

This paper focused on the development of a feasible solution to reduce petcoke emissions from a Portuguese harbor (Port of Aveiro), which could be endangering the population and dwellings of the surrounding urban area.

The experimental air quality field campaigns show that under meteorological conditions the atmospheric concentration of PM10 could be significantly high, particularly in the second campaign. However, during the last period of measurements, the pollution levels decreased, and this could be

related with a change in procedure when handling and storing the petcoke, following the suggestions of keeping the piles more humid and protected from wind, alter the way petcoke is offloaded and releasing the petcoke closer to the ground, to reduce dust emissions.

The use of different barriers, positioned downstream and upstream of the petcock pile, resulted in reductions in particulate emissions compared to the reference scenario. Upstream placement was the most efficient solution. The barrier typology that led to the largest reductions in particle emissions was the solid barrier, which blocked more efficiently the passage of wind to the pile. Results showed that placing a double barrier with the position and configuration named as B would be the most efficient solution to reduce petcoke dust emissions for the most critical wind directions concerning the residential areas (N and NW).

Results from the numerical modelling for the optimal configuration found with WT simulations showed good agreement with the trends observed with the physical modelling. Modelling results also indicate a significant reduction of fugitive particles with the placement of the barrier, confirming the wind tunnel conclusions. The optimized barrier has been implemented in the field and monitoring campaigns are currently being carried out to assess its effectiveness.

The methodology discussed in this study allows quantifying the impacts of harbor activity in its surroundings, independently on its location. This way, the procedure described in this paper to assess the effectiveness of a barrier as a solution to mitigate fugitive dust emissions can be an effective methodology to implement in other harbor with similar air quality issues. The barrier has been implemented on a real scale and monitoring campaigns are currently being carried out to assess its effectiveness, in combination with good practices on (un)loading and storing petcoke in the port of Aveiro. The analyses of these experimental results will provide evidence on the actual improvement and the perception of it by the population.

Chapter 6

6. Conclusions, future challenges, and final remarks

6.1. Main findings

The literature review performed as a first step of this thesis produced a useful summary of the existing information and data on local air quality over harbour areas worldwide, giving some insights on the existing gaps in current knowledge. The scarcity of air quality monitoring data across harbours areas was confirmed, although all the cited studies agree on the relevance of harbour activities for the emission of some major pollutants, namely PM10, PM2.5, NO_2 and SO_2. Measured concentrations of the main air pollutants were compiled and intercompared, showing that, despite the large spatial variability of pollutants' concentration across the world, the highest NO_2 and PM concentrations were found in Europe and Asia, ranging between 12-107 µg NO_2/m^3 and 2-50 µg PM10/m^3, and 25 – 70 µg PM2.5/m^3 respectively. Results published on Asian ports showed that ships can contribute to 7 – 26 % of the total fine particulate matter concentration found in port vicinity. Regarding SO_2, a decrease in this pollutant emission is being registered across Europe since January 2010, due to the efforts made by EU and IMO to adopt mitigation measures that could effectively reduce ship emissions.

These values proved the need for thorough emission inventories covering all activities and sectors within ports, and a rigorous quantification of all the associated emissions. A recommendation is given that all European harbours should prepare their own detailed emissions inventory annually, based on their activities, and using up-to-date standards, following a harmonized/ standard approach.

The work presented in Chapter 3 highlighted the need for standardization of the methodologies used to estimate emissions from harbour activities. The two most used methodologies within the scientific community and port authorities, namely the methodologies applied by the European Monitoring and Evaluation Programme from the European Environment Agency and US Starcrest Consulting Group report were analysed and intercompared. This work suggested that EF used by EMEP/EEA method for cargo handling equipment emissions may raise less doubts and probably are the most recommendable, since they account for technology level and engine size class (according to the guidelines of emissions for non-road mobile machinery), rather than just ZH and DR, as defined by US/SCG method. On the other hand, regarding shipping, the US/SCG report may be more adequate considering the legislation in force since January 2010, given that it differentiates the emission factors according to the sulphur content of the fuel and the IMO level. Therefore, it was suggested the development of a new harmonized methodology combining both EMEP/EEA and US/SCG for the aspects where each one performs best. Besides emission factors, two other major issues need harmonization: (i) whether to consider the boiler as emission source or not, and (ii) to define if the main engine should be considered turned on or off, while hoteling.

Using one of the main Portuguese harbours as case-study – Port of Leixões – emissions were estimated and the community-scale webtool (C-PORT) was applied to assess the contribution of the activities to the air quality in surroundings. The concentrations measured in the air quality stations of

the Port of Leixões were close to the values obtained by C-PORT, proving the suitability of this tool to monitor and predict pollutants' concentration in the vicinity of the port. The highest PM10 concentrations simulated with the C-PORT tool were found within the Port of Leixões area, mainly in the South Container Terminal. The annual maximum value of PM10 concentrations of 263 µg/m^3 was recorded in the receptor located outside the port, in the first line of dwellings. The model was also able to capture the NOx magnitude values from one of the two urban stations studied. The maximum value of NO_X concentrations of 122 µg/m^3 was located near the entrance of the southern container terminal and highway. The C-PORT modelling estimated the relative contributions of various source sectors to air quality concentrations, including port (terminals, ships, and roads), roadway traffic, and industrial (refinery) sources that potentially affect the local urban community. The land-based emission sources, (including trucks, railways, cargo handling equipment and bulk material stored) at the Port of Leixões exhibit the highest contribution (approximately 80 %) to the levels of PM10 concentrations in the study area. Harbour activities and the refinery (located close to the port) have a negligible contribution. The docked ships are the main source with a contribution above 50 % for NOx concentration values, with ships in transit contributing below 1 %, justified by the extended hoteling time while the ships are docked.

Most mitigation measures proposed in the literature to improve air quality in harbours are preventive measures, failing to address structural mitigation measures. In this work, structural measure to be applied in port terminals were identified. The dispersion of petcoke around the Port of Aveiro was successfully minimised through the implementation of a barrier upwind from the petcoke piles. Optimal structure, size and position of that barrier were defined based on numerical and physical modelling. Results showed that the barrier can effectively reduce the petcoke dust emissions for the tested wind directions (N, NW, and S) when low wind speed regime applies (3 m/s). In the residential areas, for the most critical wind directions (N and NW), reductions on emission of 71 % and 60 % were obtained for the case of 7 m/s wind speed. For stronger winds (11 m/s), the reduction on petcoke dust emissions was even higher, reaching approximately 88 % for N winds and 74 % for NW winds. The studied barrier has been implemented on a real scale and monitoring campaigns are currently being carried out to assess its effectiveness, in combination with good practices on (un)loading and storing petcoke in the port of Aveiro.

6.2. Future challenges and final remarks

There is still an urgent need to improve our understanding in this research field, if countries aim at effectively control and reduce the impact of port-related activities on air quality, especially in the surroundings of ports, where thousands of people have their houses. This thesis can provide a useful starting point for the development of long-term emission estimates for future scenarios, based on the emission calculation procedure proposed in Chapter 3. Such estimates must be continuously updated and improved, considering the national policies and strategies foreseen. Moreover, emission inventories' methodology and results must be constantly updated based on scientific studies, as well as there must be more studies of measurements of port emissions, both on ships and on cargo

handling equipment. The shortage of real measurements on ship emissions strongly influences the emission factors adopted from each data source. Literature points to uncertainties in the order of 20 % - 50 % in emission factors for the different pollutants. To address this problem, there is a great need for more ship emission test studies, so that science can provide decision makers with thorough and updated data. These emission tests are usually an expensive and difficult task; therefore, the most recent emission factors were published back in 2010 (Entec (2010)). Regarding the Portuguese reality, it would be important to study emissions and air quality levels from all Portuguese ports, following a common methodology, as well as from all ships sailing up to 400 km off the Portuguese coast, which would allow an integrated assessment of emissions and air quality from port areas in Portuguese territory, and a continuous comparison of performance between ports. An integrated assessment of the emissions from harbour related activities, followed by the evaluation of the air quality levels in harbour areas, and their vicinity (e.g., through air quality monitoring campaigns, numerical and physical modelling studies), together with the quantification of the health-related impacts due to population exposure to those air pollution levels are crucial towards the mitigation of the environmental impacts of harbour activities.

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APPENDICES

Appendix A

Table S 1. List of case studies using source-apportionment methods to estimate other source contribution to PM values (like traffic, industry and natural).

	Reference	Location	Period	Method	PM	Others (%)
Europe	Saraga et al. (2019)	Thessaloniki harbour (Greece)	NA	PMF	PM2.5	Traffic - 45 Industry – NA Natural*- 1
	Scerri et al. 2018	Malta urban (Republic of Malta)	2016	PMF	PM2.5	Traffic - 27 Industry - NA Natural* - 15
	Manousakas et al (2017)	Patras harbour (Greece)	2011	PMF	PM2.5	Traffic - 34 Industry - NA Natural* - 11
	Manousakas et al (2017)	Patras harbour (Greece)	2011	PMF	PM10	Traffic - 33 Industry - NA Natural* -21
	Cesari et al. (2014)	Brindisi harbour (Italy)	2012.6 - 2012.10	PMF	PM2.5	Traffic - 16 Industry - 16 Natural* - 14
	Bove et al. (2014)	Genoa urban (Italy)	2011.5 - 2011.10	PMF	PM2.5	Traffic - 25 Industry - 11 Natural* - 7
	Pérez et al. (2016)	Barcelona harbour (Spain)	2011.02 - 2011.12	PMF	PM2.5	Traffic - 36 Industry - 14 Natural - 2
			2011.02 - 2011.12	PMF	PM10	Traffic - 40 Industry - 8 Natural* - 2
		Barcelona urban (Spain)	2011.02 - 2011.12	PMF	PM10	Traffic - NA Industry - 4 Natural* - 3
			2011.02 - 2011.12	PMF	PM2.5	Traffic - NA Industry - 6 Natural* - 4
	Amato et al. (2009)		2003 -2007	PMF	PM2.5	Traffic - 25 Industry - 3 Natural* - 17
			2003 -2007	PMF	PM10	Traffic - 18 Industry - 2 Natural* - 42
	Pandolfi et al. (2011)	Gibraltar Strait urban (Spain)	2003 -2007	PMF	PM2.5	Traffic - 28 Industry - 7 Natural* - 27
			2003 -2007	PMF	PM10	Traffic - 20 Industry - 5 Natural* - 10

Table S 1(cont.) List of case studies using source-apportionment methods to estimate other source contribution to PM values (like traffic, industry and natural).

	Reference	Location	Period	Method	PM	Others (%)
Europe	Healy et al. (2010)	Cork harbour (Ireland)	2008.5 - 2008.8	PMF	PM2.5	Traffic - 23 Industry - 11 Natural* - 14
	Hellebust et al. (2010)		2007.5 - 2008.4	MLR	PM10	Traffic - NA Industry - NA Natural* - 19
	Healy et al. (2009)		2008.8	PMF	PM2.5	Traffic - 23 Industry - NA Natural* -NA
Asia	Xu et al. (2018)	Xiamen harbour - industrial (China)	2015.4 - 2016.1	PMF	PM2.5	Traffic - 22 Industry - NA Natural* - 2
		Xiamen Urban (China)	2015.4 - 2016.1	PMF	PM2.5	Traffic - 10 Industry - 14 Natural* - 10
	Mamoudou et al. (2018)	Yangshan Island harbour (China)	2016	PMF	PM2.5	Traffic - 7 Industry - 88 Natural* - 4
	Tao et al. (2017)	Zhuhai urban (China)	2014 -2015	PMF	PM2.5	Traffic - 10 Industry - NA Natural* - NA
	Jeong et al. (2017)	Busan urban (Korea)	2013	PMF	PM2.5	Traffic - 22 Industry - 3 Natural* - 18
			2013	CMB	PM2.5	Traffic - 45 Industry - 5 Natural* - 1
			2013	PCA/ APCS	PM2.5	Traffic - 1 Industry - 2 Natural* - 38
	Yau et al. (2013)	Kwai Chung and Tsing Yi urban (Hong Kong)	2009.8 - 2010.3	PMF	PM2.5	Traffic - 11 Industry - 23 Natural* - 5
			2009.8 - 2010.3	PCA/ APCS	PM2.5	Traffic - 8 Industry - 2 Natural* - 7
North American	Gibson et al. (2013)	Halifax urban (Canada)	2011.7 - 2011.8	PMF	PM2.5	Traffic - 13 Industry - 2 Natural* -
	Minguillón et al. (2008)	Angeles Long Beach harbour (California)	2007	CMB	PM2.5	Traffic - NA Industry - NA Natural* - NA
	Kuwayama et al. (2013)	Oakland harbour (California)	2010.3	PMF	EC	Traffic - 56 Industry - NA Natural* -NA
	Kuwayama et al. (2013)	Oakland harbour (California)	2010.4 – 2010.5	PMF	EC	Traffic - 23 Industry - NA Natural* -NA

Appendix B

App. 1 - Cargo Handling Equipment

App. 1.1 – Load Factors

Table S 2. Cargo Handling Equipment Engine Load Factors (adapted from (SCG, 2019)).

CHE Type	Load Factor
Cone vehicle	0.51
RTG crane	0.20
Crane	0.43
Excavator	0.55
Forklift	0.30
Loader, backhoe	0.55
Top handler, side pick, reach stacker	0.59
Truck, other with off-road engine	0.51
Truck, other with on-road engine	0.51
Straddle carrier	0.20
Sweeper	0.68
Yard tractor with off-road engine	0.39
Yard tractor with on-road engine	0.39

Table S 3 Cargo Handling Equipment Engine Load Factors Adjustment (adapted from (EEA, 2019b)).

Technology Level	Load	Load factor	NO_x	VOC	CO	TSP	FC
Stage II and prior	High	>0.45	0.95	1.05	1.53	1.23	1.01
Stage IIIA	High	>0.45	1.04	1.05	1.53	1.47	1.01
Stage IIIB-IV	High	>0.45	1	1	1	1	1
Stage II and prior	Middle	0.25≤LF≤0.45	1.025	1.67	2.05	1.6	1.095
Stage IIIA	Middle	0.25≤ LF≤0.45	1.125	1.67	2.05	1.92	1.095
Stage IIIB-IV	Middle	0.25≤ LF≤0.45	1	1	1	1	1
Stage II and prior	Low	<0.25	1.1	2.29	2.57	1.97	1.18
Stage IIIA	Low	<0.25	1.21	2.29	2.57	2.37	1.18
Stage IIIB-IV	Low	<0.25	1	1	1	1	1

App. 1.2 – Fuel Correction Factors

Table S 4 Fuel Correction Factors for CHE (adapted from (SCG, 2019)).

CHE model year	PM	NO_X	SO_X	CO	HC	CO_2
1996-2010	1.336	1.087	2.495	1.000	1.470	1.000
2011 and newer	1.249	1.087	2.495	1.000	1.470	1.000

App. 1.3 – Deterioration factors adjustments

The deterioration factor adjustment for a given machinery type at a given time depends on the engine-size class (only for gasoline), the emission level and the average engine lifetime. For diesel and gasoline 2-stroke engines the deterioration factor adjustment is generally expressed as:

$$\mathrm{DF_{D,2ST}} = \frac{\mathrm{K}}{\mathrm{LT}} \mathrm{DF_{y,z}}$$

Where,
$DF_{D,2ST}$ is the deterioration factor adjustment for diesel and 2-stroke gasoline machinery;
K is the engine age (between 0 and average lifetime);
LT is the average lifetime;
y is the engine-size class, and z is the technology level.

Table S 5 Deterioration factors for diesel machinery relative to average engine lifetime (adapted from (EEA, 2019b)).

Emission Level	NO_X	VOC	CO	TSP
Before Stage I	0.024	0.047	0.185	0.473
Stage I	0.024	0.036	0.101	0.473
Stage II	0.009	0.034	0.101	0.473
Stage IIIA, IIIB, IV, V	0.008	0.027	0.151	0.473

Table S 6. Aggregation of diesel CHE type (adapted from (EEA, 2019b)).

CHE type	Lifetime (yrs)
Crane	10
Forklifts (diesel)	20
Shovel Excavator	10
Reach Stacker	10
Excavator	10
Container Handler	10
Terminal Truck	10
Sweeper	10
Loader	10

App. 1.4 – Emission Factors

Table S 7 Emission factors (g/kWh) for diesel Non-Road Mobile Machinery (EEA, 2019b)).

CHE	CHE Year	Power (kw)	CH (h)	Emission factors (g/(kWh))									
				PM10	PM2.5	DPM	NO_X	VOC	CO	BC	CO_2	N_2O	CH_4
Crane	2007/ 2008	670	4509	0.20	0.14	0.20	6.48	0.60	3.00	0.14	6320	0.070	0.014
	2001	400	1594	0.10	0.07	0.10	5.20	0.30	1.50	0.07	3160	0.035	0.007
	2014	765	2357	0.03	0.00	0.03	3.50	0.13	1.50	0.07	3160	0.035	0.003
	2016	750	333	0.03	0.00	0.03	3.50	0.13	1.50	0.07	3160	0.035	0.003
Forklift	1991	45	278	4.40	0.44	4.60	37.45	6.40	17.00	0.25	9480	0.105	0.154
	2008	60.3	1169	0.80	0.64	0.80	15.40	1.20	4.40	0.16	6320	0.070	0.028
	2009	147	1720	0.40	0.16	0.40	7.05	0.70	3.70	0.16	6320	0.070	0.017
	2000/ 2002	260	6433	0.13	0.13	0.13	5.60	0.43	3.00	0.09	6320	0.070	0.010
	2004	129	10630	0.05	0.05	0.05	0.80	0.26	3.00	0.04	6320	0.070	0.006
	1998	50	49	0.80	0.44	0.80	11.75	1.50	4.50	0.08	3160	0.035	0.036
	2000	60.3	557	2.50	1.43	2.60	19.45	3.90	10.50	0.16	6320	0.070	0.094
	1980	60	10	0.80	0.44	0.80	11.75	1.50	4.50	0.08	3160	0.035	0.036
	2007	68	451	0.20	0.16	0.20	3.81	0.40	2.20	0.08	3160	0.035	0.010
	1995	140	563	0.20	0.16	0.20	3.81	0.40	2.20	0.08	3160	0.035	0.010
	2005	145	965	0.40	0.32	0.40	7.70	0.60	2.20	0.08	3160	0.035	0.014
	2015	164	1045	0.80	0.20	0.90	11.20	0.50	2.50	0.08	3160	0.035	0.012
	2001	235	769	0.40	0.20	0.40	11.20	0.50	2.50	0.08	3160	0.035	0.012
	1966	180	4	0.10	0.07	0.10	3.24	0.30	1.50	0.08	3160	0.035	0.007
	2007	180	3557	0.10	0.10	0.10	5.20	0.30	1.50	0.07	3160	0.035	0.007
	2007	265	1907	0.03	0.03	0.03	0.40	0.13	1.50	0.02	3160	0.035	0.003
	1981	64	246	1.20	1.70	1.80	8.60	2.00	5.30	0.66	3160	0.035	0.048
Terminal Truck	2006/ 2007	205	5292	0.15	0.15	0.15	2.40	0.78	9.00	0.11	18960	0.210	0.018
	2010	205	1932	0.03	0.03	0.03	0.40	0.13	1.50	0.02	3160	0.035	0.003

Table S 7(cont). Emission factors (g/kWh) for diesel Non-Road Mobile Machinery (EEA, 2019b).

CHE	CHE Year	Power (kw)	CH (h)	Emission factors (g/(kWh))									
				PM10	PM2.5	DPM	NO_X	VOC	CO	BC	CO_2	N_2O	CH_4
Shovel excavators	2006	60.3	108	0.40	0.32	0.40	11.00	0.80	4.40	0.16	6320	0.070	0.020
	2006	61.5	392	0.40	0.32	0.40	11.00	0.80	4.40	0.16	6320	0.070	0.020
	2000	203	1249	0.40	0.28	0.40	15.20	0.60	3.00	0.16	6320	0.070	0.014
	2016	274	1270	0.06	0.04	0.06	0.80	0.26	3.00	0.14	6320	0.070	0.006
	2004	117	201	0.20	0.16	0.20	3.24	0.30	1.50	0.08	3160	0.035	0.007
	2005	127	667	0.20	0.16	0.20	5.20	0.30	1.50	0.08	3160	0.035	0.007
	2012	135	886	0.10	0.07	0.10	3.24	0.30	1.50	0.08	3160	0.035	0.007
	2015	129	1046	0.03	0.02	0.03	2.97	0.13	1.50	0.08	3160	0.035	0.003
	1992	60.3	13	0.40	0.32	0.40	7.70	0.60	2.20	0.08	3160	0.035	0.014
	1987	60	10	1.10	0.66	1.20	8.60	2.00	5.30	0.08	3160	0.035	0.048
	1986	147	223	0.80	0.40	0.80	12.40	1.00	2.50	0.08	3160	0.035	0.024
	1991	152	140	0.40	0.20	0.40	11.20	0.50	2.50	0.08	3160	0.035	0.012
	1998	137	174	0.40	0.20	0.40	11.20	0.50	2.50	0.08	3160	0.035	0.012
	1988	185	84	0.80	0.40	0.80	12.40	1.00	2.50	0.08	3160	0.035	0.024
	2002	258	786	0.10	0.07	0.10	5.20	0.30	1.50	0.07	3160	0.035	0.007
	2001	235	1439	0.40	0.28	0.40	15.20	0.60	3.00	0.14	6320	0.070	0.014
	2010	247	294	0.10	0.07	0.10	3.24	0.30	1.50	0.07	3160	0.035	0.007
	2001	247	276	0.10	0.10	0.10	5.20	0.30	1.50	0.07	3160	0.035	0.007
	2005/ 2007	256	9228	0.80	0.12	0.12	12.96	1.20	6.00	0.64	12640	0.140	0.028
	2011	256	5736	0.40	0.06	0.06	6.48	0.60	3.00	0.32	6320	0.070	0.014
Sweeper	2010	20	152	0.40	0.40	0.40	6.08	0.60	2.20	0.32	3160	0.035	0.014
Loader	1981	147	195	0.90	0.80	0.90	17.80	1.50	2.50	0.45	3160	0.035	0.036
	2007	114	657	0.20	0.40	0.40	3.24	0.30	1.50	0.16	3160	0.035	0.007
Container Handler	1994	190	96	0.10	0.10	0.10	3.24	0.30	1.50	0.07	3160	0.035	0.007
	2016	185	1037	0.10	0.10	0.10	3.24	0.30	1.50	0.07	3160	0.035	0.007

Table S 8 Emission Factors (g/kWh) for different CHE types (SCG, 2019).

CHE	CHE Year	Power (kw)	CH (h)	Emission factors (g/(kWh))									
				PM10	PM2.5	DPM	NO_X	SO_X	CO	HC	CO_2	N_2O	CH_4
Crane	2007/ 2008	670	4509	0.34	0.31	0.67	12.48	0.14	2.57	0.51	1524	0.042	0.096
	2001	400	1594	0.17	0.16	0.17	6.79	0.07	1.27	0.3	762	0.02	0.047
	2014	765	2357	0.17	0.16	0.34	6.25	0.07	1.29	0.26	762	0.021	0.048
	2016	750	333	0.15	0.14	0.18	6.08	0.07	1.24	0.2	762	0.021	0.048
Forklift	1991	45	278	2.81	2.59	4.66	35.26	0.24	14.07	4	2286	0.072	0.168
	2008	60.3	1169	0.66	0.62	0.66	13.55	0.16	8.41	0.56	1524	0.048	0.112
	2009	147	1720	0.32	0.28	0.32	6.64	0.16	2.52	0.32	1524	0.048	0.106
	2000/ 2002	260	6433	0.41	0.39	0.42	10.66	0.14	2.61	0.85	1524	0.04	0.094
	2004	129	10630	0.39	0.34	0.39	13.31	0.16	2.81	0.71	1524	0.04	0.094
	1998	50	49	0.93	0.86	1.26	11.74	0.08	4.69	1.33	762	0.024	0.056
	2000	60.3	557	1.89	1.75	1.89	18.62	0.16	9.43	2.69	1524	0.048	0.112
	1980	60	10	1.13	1.04	1.13	17.44	0.08	6.44	1.93	762	0.024	0.056
	2007	68	451	0.96	0.88	0.96	9.35	0.08	4.74	1.36	762	0.024	0.056
	1995	140	563	0.53	0.49	0.53	11.1	0.08	3.67	0.93	762	0.024	0.053
	2005	145	965	0.16	0.15	0.16	6.23	0.08	1.26	0.22	762	0.024	0.053
	2015	164	1045	0.01	0.01	0.01	0.37	0.08	1.26	0.08	762	0.024	0.053
	2001	235	769	0.17	0.15	0.17	6.71	0.07	1.25	0.27	762	0.02	0.047
	1966	180	4	1.03	0.95	1.03	18.78	0.08	5.9	1.77	762	0.024	0.053
	2007	180	3557	0.18	0.16	0.18	3.44	0.08	1.35	0.25	762	0.024	0.053
	2007	265	1907	0.21	0.15	0.16	3.37	0.07	1.28	0.19	762	0.02	0.047
	1981	64	246	1.15	1.06	1.15	17.53	0.08	6.48	1.95	762	0.024	0.056
Terminal Truck	2006/ 2007	205	5292	0.96	0.85	0.96	35.68	0.48	7.56	1.14	4572	0.144	0.318
	2010	205	1932	0.16	0.15	0.16	3.37	0.07	1.28	0.19	762	0.02	0.047

Table S 8 (cont.). Emission Factors (g/kWh) for different CHE types (SCG, 2019).

CHE	CHE Year	Power (kw)	CH (h)	Emission factors (g/(kWh))									
				PM10	PM2.5	DPM	NO_X	SO_X	CO	HC	CO_2	N_2O	CH_4
Shovel excavator	2006	60.3	108	0.64	0.59	0.64	13.45	0.16	8.29	0.52	1524	0.048	0.112
	2006	61.5	392	0.65	0.6	0.65	13.48	0.16	8.32	0.54	1524	0.048	0.112
	2000	203	1249	0.31	0.28	0.31	6.63	0.16	2.5	0.3	1524	0.048	0.106
	2016	274	1270	0	0.02	0.02	0.73	0.14	2.49	0.16	1524	0.04	0.094
	2004	117	201	0.26	0.24	0.26	6.35	0.08	3.64	0.31	762	0.024	0.053
	2005	127	667	0.27	0.25	0.27	6.4	0.08	3.68	0.32	762	0.024	0.053
	2012	135	886	0.2	0.19	0.2	3.32	0.08	3.7	0.16	762	0.024	0.053
	2015	129	1046	0.01	0.01	0.01	0.37	0.08	3.72	0.08	762	0.024	0.053
	1992	60.3	13	0.93	0.86	0.93	11.73	0.01	4.68	1.33	762	0.024	0.056
	1987	60	10	1.13	1.04	1.13	17.44	0.08	6.44	1.93	762	0.024	0.056
	1986	147	223	0.52	0.48	0.52	11.01	0.08	3.64	0.92	762	0.024	0.053
	1991	152	140	0.52	0.47	0.52	10.99	0.08	3.63	0.92	762	0.024	0.053
	1998	137	174	0.2	0.19	0.2	8.41	0.08	1.24	0.43	762	0.024	0.053
	1988	185	84	0.51	0.47	0.51	10.98	0.08	3.63	0.91	762	0.024	0.053
	2002	258	786	0.17	0.15	0.17	6.72	0.07	1.25	0.28	762	0.02	0.047
	2001	235	1439	0.34	0.3	0.34	13.42	0.14	2.5	0.55	1524	0.04	0.094
	2010	247	294	0.15	0.14	0.15	3.3	0.07	1.24	0.14	762	0.02	0.047
	2001	247	276	0.16	0.15	0.16	6.67	0.07	1.24	0.26	762	0.02	0.047
	2005/ 2007	256	9228	0.78	0.61	0.68	17.82	0.28	5.15	0.83	3048	0.08	0.188
	2011	256	5736	0.44	0.32	0.34	6.81	0.14	2.6	0.46	1524	0.04	0.094
Sweeper	2010	20	152	0.21	0.2	0.21	6.46	0.09	3.7	0.14	762	0.027	0.061
Loader	1981	147	195	1.15	1.05	1.15	17.51	0.08	6.47	1.95	762	0.024	0.056
	2007	114	657	0.2	0.18	0.2	3.31	0.08	3.68	0.15	762	0.024	0.053
Container Handler	1994	190	96	0.51	0.47	0.51	10.98	0.08	3.63	0.91	762	0.02	0.047
	2016	185	1037	0.01	0.01	0.01	0.37	0.08	1.26	0.08	762	0.02	0.047

App. 2 – Ship methodology

App. 2.1 - Engine/Fuel type profiles

Table S 9. Engine speed by gross tonnage of each ship type (adapted from ENTEC, 2010).

Ship Type	≤5 000 GT [a]	5 000 – 25 000 GT	>25 000 GT
Bulk Carrier	MSD [b]	SSD [c]	SSD
Container Ship	MSD	MSD	SSD
General Cargo	MSD	SSD	SSD
Passenger	HSD [d]	MSD	MSD
Ro-Ro cargo	MSD	MSD	SSD
Tanker	MSD	SSD	MSD
Others	MSD	MSD	SSD

[a] GT – Gross Tonnage; [b] MSD – Medium Speed Diesel; [c] SSD – Slow Speed Diesel; [d] HSD – High Speed Diesel

Table S 10 Fuel Type used per ship and engine type (adapted from ENTEC, 2010).

Ship Type	ME [a] Fuel Type	AE [b] Fuel Type
Bulk Carrier	RO [c]	MGO [d]
Container Ship	RO	RO
General Cargo	RO	MGO
Passenger	MDO [e]	MDO
Ro-Ro cargo	RO	RO
Tanker	RO	MGO
Others (see note h)	MGO	MGO

[a] ME – Main Engine; [b] AE – Auxiliary Engine; [c] RO – Residual Oil; [d] MGO – Marine Gas Oil; [e] MDO – Marine Diesel Oil;

App. 2.2 –Load Factors

Table S 11 Load factors of main and auxiliary engines according to operational modes (adapted from (EEA, 2019a)).

Operational Mode	ME[a] load (%)	AE[b] load (%)
Manoeuvring	20	50
Hotelling	0	40; 60[c]

[a] ME – Main Engine; [b] AE – Auxiliary Engine; [c] For Tankers.

Table S 12 Load factors of main and auxiliary engines according to ship type (adapted from (SCG, 2019)).

	ME[a] load (%)		AE[b] load (%)	
Ship Type	**Hotel.**	**Manoeuv.**	**Hotel.**	**Manoeuv.**
Auto carrier	0.00	0.15	0.26	0.45
Bulk carrier	0.00	0.27	0.10	0.45
Container ship	0.00	0.15	0.19	0.48
Cruise ship	0.00	0.15	0.64	0.80
General cargo	0.00	0.15	0.22	0.45
OG Tug	0.00	0.27	0.22	0.45
RORO	0.00	0.27	0.26	0.45
Reefer	0.00	0.27	0.32	0.67
Tanker	0.00	0.27	0.26	0.33
Miscellaneous	0.00	0.27	0.22	0.45

[a] ME – Main Engine; [b] AE – Auxiliary Engine.

Table S 13 Assumptions for the average cruise speed (adapted from (SCG, 2019)).

Ship Type	**Default cruising speed (km/h)**
Auto carrier	11
Bulk carrier	9
Container ship	11
Cruise ship	11
General cargo	11
OG Tug	9
RORO	9
Reefer	9
Tanker	9
Miscellaneous	9

App. 2.3 – Fuel Correction Factors

Table S 14 Fuel correction factors for ship (adapted from (SCG, 2019)).

Baseline Fuel and % S	**Used fuel and % S**	**SO_X**	**NO_X**	**HC**	**CO_2**	**CO**	**PM2.5**	**PM10**	**CH_4**	**N_2O**
HFO/RO (2.7%)	MGO (0.1%)	0.037	0.94	1	0.95	1	0.20	0.17	1	0.9
	RO (2.7%)	1.000	1.00	1	1.00	1	1.00	1.00	1	1.0
	MGO (0.5%)	0.185	0.94	1	0.95	1	0.29	0.25	1	0.9

App. 2.4 – Emission Factors

Table S 15 Emission Factors (g/kWh) for different engine types/fuel combinations and operational modes (manoeuvring and hotelling) (EEA, 2019a; IMO, 2010; Merk, 2014; Nunes et al., 2017).

Engine	Engine Type	Fuel Type	Sulfur (%)	SO_X	NO_X	VOC	HC	CO_2	CO	PM2.5	PM10	CH_4	N_2O
Main	SSD	RO	2.7	11.6	13.6	1.8	0.6	682	0.54	1.32	2.4	0.012	0.031
		MDO	1.0	6.2	12.8	1.8	0.6	647	0.54	0.44	1.2	0.012	0.031
		MGO	0.5	0.8	12.8	1.8	0.6	647	0.54	0.29	0.9	0.012	0.031
		MDO/MGO	0.1	0.4	17.0	1.8	0.6	588	1.40	0.17	0.2	0.006	0.031
	MSD	RO	2.7	12.7	10.5	1.5	0.5	745	0.54	1.32	2.4	0.010	0.034
		MDO	1.0	6.8	9.9	1.5	0.5	710	0.54	0.46	1.2	0.010	0.034
		MGO	0.5	0.9	9.9	1.5	0.5	710	0.54	0.30	0.9	0.010	0.034
		MDO/MGO	0.1	0.4	13.2	1.5	0.5	646	1.10	0.17	0.2	0.004	0.031
Auxiliary	MSD/SSD	RO	2.7	12.3	13.8	0.4	0.4	722	0.54	1.32	0.8	0.008	0.036
		MDO	1.0	6.5	13.0	0.4	0.4	690	0.54	0.45	0.4	0.008	0.036
		MGO	0.5	0.9	13.0	0.4	0.4	690	0.54	0.29	0.3	0.008	0.036
		MDO/MGO	0.1	0.4	13.9	0.4	0.4	690	1.10	0.17	0.2	0.004	0.031
Boiler	NA	HFO/RO	2.7	16.5	2.1	0.3	0.1	970	0.20	0.64	0.8	0.002	0.080
	NA	MDO/MGO	0.1	0.43	2.6	0.3	0.5	649	1.1	0.24	0.26	0.01	0.029

Table S 16 Emission Factors (g/kWh) for different engine types/fuel combinations and operational modes (manoeuvring and hotelling) (adapted from (EPA, 2009; SCG, 2019)).

Engine	Engine Type	Fuel Type	IMO Tier	Sulfur (%)	SO_X	NO_X	VOC	HC	CO_2	CO	PM2.5	PM10	CH_4	N_2O
Main	SSD	RO	NA	2.7	10.3	18.1	1.8	0.6	620	1.4	1.31	1.42	0.006	0.031
		MDO	NA	1.0	3.6	17.0	1.8	0.6	588	1.4	0.42	0.45	0.006	0.031
		MGO	NA	0.5	1.8	17.0	1.8	0.6	588	1.4	0.28	0.31	0.006	0.031
		MDO/ MGO	0	0.1	0.4	17.0	1.8	0.6	588	1.4	0.24	0.26	0.012	0.029
			1	0.1	0.4	15.9	1.8	0.6	588	1.4	0.24	0.26	0.012	0.029
			2	0.1	0.4	14.4	1.8	0.6	588	1.4	0.24	0.26	0.012	0.029
			3	0.1	0.4	3.4	1.8	0.6	588	1.4	0.24	0.26	0.012	0.029
	MSD	RO	NA	2.7	11.2	14.0	1.5	0.5	677	1.1	1.32	1.43	0.004	0.031
		MDO	NA	1.0	3.9	13.2	1.5	0.5	646	1.1	0.43	0.47	0.004	0.031
		MGO	NA	0.5	1.9	13.2	1.5	0.5	646	1.1	0.29	0.31	0.004	0.031
		MDO/ MGO	0	0.1	0.4	13.2	1.5	0.5	649	1.1	0.24	0.26	0.010	0.029
			1	0.1	0.4	12.2	1.5	0.5	649	1.1	0.24	0.26	0.010	0.029
			2	0.1	0.4	10.5	1.5	0.5	649	1.1	0.24	0.26	0.010	0.029
			3	0.1	0.4	2.6	1.5	0.5	649	1.1	0.24	0.26	0.010	0.029
Auxiliary	MSD/ SSD	RO	NA	2.7	11.9	14.7	0.4	0.4	722	1.1	1.32	1.44	0.004	0.031
		MDO	NA	1.0	4.2	13.9	0.4	0.4	690	1.1	0.45	0.49	0.004	0.031
		MGO	NA	0.5	2.1	13.9	0.4	0.4	690	1.1	0.29	0.32	0.004	0.031
		MDO/ MGO	0	0.1	0.5	13.8	0.4	0.6	686	1.4	0.24	0.26	0.012	0.029
			1	0.1	0.5	12.2	0.4	0.6	686	1.4	0.24	0.26	0.012	0.029
			2	0.1	0.5	10.5	0.4	0.6	686	1.4	0.24	0.26	0.012	0.029
			3	0.1	0.5	2.6	0.4	0.6	686	1.4	0.24	0.26	0.012	0.029
Boiler		HFO/ RO	NA	2.7	16.5	2.1	0.3	0.5	970	0.2	0.64	0.80	0.002	0.080
		MDO/ MGO	NA	0.1	0.6	1.9	0.3	0.5	922	0.2	0.13	0.14	0.002	0.075

App. 3 – Results

App. 3.1 – Cargo Handling Equipment

Table S 17 In-port emissions of pollutants by CHE at Port of Leixões during 2016 (in tonne/year) with EMEP/EEA methodology.

CHE	Pop	NO_X	CO	N_2O	PM	PM10	PM2.5	VOC	CH_4	SO_X	CO_2	NH_3	BC
Crane	5	52.86	16.51	0.17	1.13	1.49	0.75	2.17	0.03	0.56	15521	0.01	0.35
Forklift	28	11.69	12.50	0.11	0.99	1.17	0.69	1.53	0.02	0.22	9830	0.01	0.12
Shovel excavator	21	6.15	4.06	0.05	0.55	0.57	0.35	0.56	0.01	0.17	4378	0.00	0.10
Reach stackers	10	16.40	11.95	0.18	0.49	1.98	0.42	1.48	0.04	1.20	16119	0.01	0.77
Empty handler	2	0.71	0.52	0.01	0.04	0.04	0.04	0.07	0.00	0.01	783	0.00	0.02
Loader	2	0.78	0.39	0.00	0.15	0.12	0.14	0.08	0.00	0.01	359	0.00	0.03

Table S 18 In-port emissions of pollutants by CHE at Port of Leixões during 2016 (in tonne/year) with US/SCG methodology.

	Pop	NO_X	CO	N_2O	PM10	PM2.5	HC	CH_4	SO_X	CO_2
Crane	5	16.81	3.15	0.05	1.02	0.51	0.94	0.08	0.44	1871
Forklift	28	10.36	2.47	0.03	0.48	0.42	0.87	0.05	0.30	1201
Shovel excavator	21	2.72	1.45	0.01	0.13	0.12	0.21	0.02	0.13	527
Reach stackers	10	11.95	3.28	0.05	0.66	0.52	0.82	0.09	0.45	1943
Empty handler	2	0.17	0.18	0.00	0.01	0.01	0.03	0.00	0.02	94
Loader	2	7.73	7.38	0.04	0.54	0.50	0.55	0.08	0.39	1504

App. 3.2 – Ship

Table S 19 In-port emissions of pollutants by ship type at Port of Leixões during 2016 (in tonne/year) with EMEP/EEA methodology.

	Container	Passenger	General Cargo	Ro-Ro cargo	Fishing	Liquid bulk ships	Dry bulk carries	Tugs	Others
SO_X	749	29	15	67	4	14	10	25	1
NO_X	866	148	174	53	91	203	136	502	28
VOC	43	7	7	2	3	8	5	21	1
HC	28	6	5	1	3	7	3	16	1
CO	66	14	13	3	5	16	8	37	2
$PM_{2.5}$	22	3	3	1	1	3	2	7	0
PM_{10}	28	4	4	2	1	4	2	9	0
CH_4	0	0	0	0	0	0	0	0	0
N_2O	2	0	0	0	0	0	0	1	0
CO_2	46713	8720	8539	2319	4053	10000	5233	24299	7154

Table S 20 In-port emissions of pollutants by ship type at Port of Leixões during 2016 (in tonne/year) with US/SCG methodology.

	Container	**Passenger**	**General Cargo**	**Ro-Ro cargo**	**Fishing**	**Liquid bulk ships**	**Dry bulk carries**	**Tugs**	**Others**
SO_X	128	12	9	18	2	10	6	21	1
NO_X	347	152	79	32	39	99	34	259	13
VOC	20	7	4	2	2	5	2	14	1
HC	25	9	5	1	2	7	2	14	1
CO	35	15	8	3	4	10	3	25	1
$PM_{2.5}$	11	3	2	1	0	3	1	6	0
PM_{10}	12	4	2	1	0	3	1	6	0
CH_4	0	0	0	0	0	0	0	0	0
N_2O	2	1	0	0	0	1	0	1	0
CO_2	36714	12670	6856	1759	3009	9443	2291	19967	9328

App. 4 – Overview of shipping emissions from harbour-related activities

Table S 21 Total annual emissions of ships for different harbours (in tonne/year) with US/SCG methodology, number of ship calls and operational modes considered in different studies.

Reference	Year	Total of Ship	Report	Mode / Type	Emissions (tonne/year)							
					NO_X	SO_X	PM10	PM2.5	HC	VOC	CO	CO_2
Port of Leixões, This Study	2016	2717	EMEP/ EEA, 2019	M	190	193	25	16	8	20	10	15172
				H-a	566	662	40	43	21	25	34	55674
				H-b	1445	59	29	27	62	52	155	96046
				ME	116	85	19	10	6	17	6	7371
				AE	1877	318	38	45	55	55	127	94597
				B	208	511	38	32	31	26	66	64925
Port of Izmir, Saraçoğlu et al., 2013	2007	2803	EMEP/ EEA, 2013	M	178	161	23	-	15	-	-	9501
				H	227	207	15	-	9	-	-	12165
Port of Samsun, Alver et al., 2018	2015	2504	EMEP/ EEA, 2013	M	109	101	14		9	-	-	-
				H	196	181	16	-	9	-	-	-
				ME	80	74	7	-	7	-	-	-
				AE	225	208	19	-	12	-	-	-
Port of Leixões, Nunes et al., 2017	2014	2612	EMEP/ EEA, 2013	M	418	142	16	15	11	4	15	7236
				H	7745	3132	348	318	259	104	301	192200
Port of Leixões, This Study	2016	2717	US/SCG, 2019	M	226	174	16	15	10	20	18	14462
				H-a	560	667	48	41	31	24	45	56251
				H-b	829	32	13	13	56	36	86	79317
				ME	152	67	9	8	5	16	12	6561
				AE	1293	298	40	37	48	37	119	40007
				B	170	509	29	24	43	26	17	47211
Port of Busan, Song and Shon, 2014	2009	-	US/SCG, 2009	M	944	922	-	-	-	91	-	54395
				H	6322	6255	-	-	-	208	-	369457
Port of Nanjing, Zhang et al., 2017	2014	901	US/EPA, 2009	ME	66	1	1	1	3	-	7	4051
				AE	130	2	2	2	5	-	14	8503
Port of Yangshan, Song, 2014	2009	6518	US/SCG, 2010	M	1178	634	104	83	34	-	91	63466
				H	3020	2051	290	229	86	-	229	207533
				ME	5335	2166	571	457	367	-	725	229751
				AE	5190	2432	434	347	141	-	388	2411450
				B	233	1025	73	55	11	-	22	107547
Port of Ambarlı, Deniz and Kilic, 2010	2005	5432	US/SCG, 2005	M	305	47	-	-	-	20	110	15368
				H	496	190	-	-	-	483	2013	61385

Mode operation: M - Manoeuvring; H-a – Hotelling at anchorage; H-b – Hotelling at berth
Engine Type: ME – Main Engine; AE – Auxiliary Engine; B - Boiler

Table S 22 Total annual emissions from CHE for different harbours (in tonne/year) estimated using the US/SCG methodology, considered in different studies.

Reference	Year	Report	CHE	Emissions (tonne/year)							
				Nº	NO_X	SO_X	PM_{10}	$PM_{2.5}$	HC	CO	CO_2
Port of Leixões, This Study	2016	US/ SCG, 2019	Crane	5	16.81	0.44	1.02	0.51	0.94	3.15	1871
			Forklift	28	10.36	0.30	0.48	0.42	0.87	2.47	1201
			Shovel excavator	21	2.72	0.13	0.13	0.12	0.21	1.45	527
			Reach stacker	10	11.95	0.45	0.66	0.52	0.82	3.28	1943
			Container handler	2	0.17	0.02	0.01	0.01	0.03	0.18	94
			Loader	2	7.73	0.39	0.54	0.50	0.55	7.38	1504
Port of Nanjing, Zhang et al., 2017	2014	US/ EPA, 2009	Crane	22	1.31	0.03	0.11	0.10	0.40	0.44	146
			Reach stacker	6	10.82	0.33	0.82	0.75	2.24	3.55	1401
			Container Handler	14	26.72	0.83	1.37	1.26	4.06	7.69	3173
			Container Trailer	30	40.38	1.04	1.57	1.44	9.14	9.62	6232
			Forklift	22	3.76	0.10	0.40	0.37	0.55	2.54	399