Impact of harbour activities on local air quality: A review

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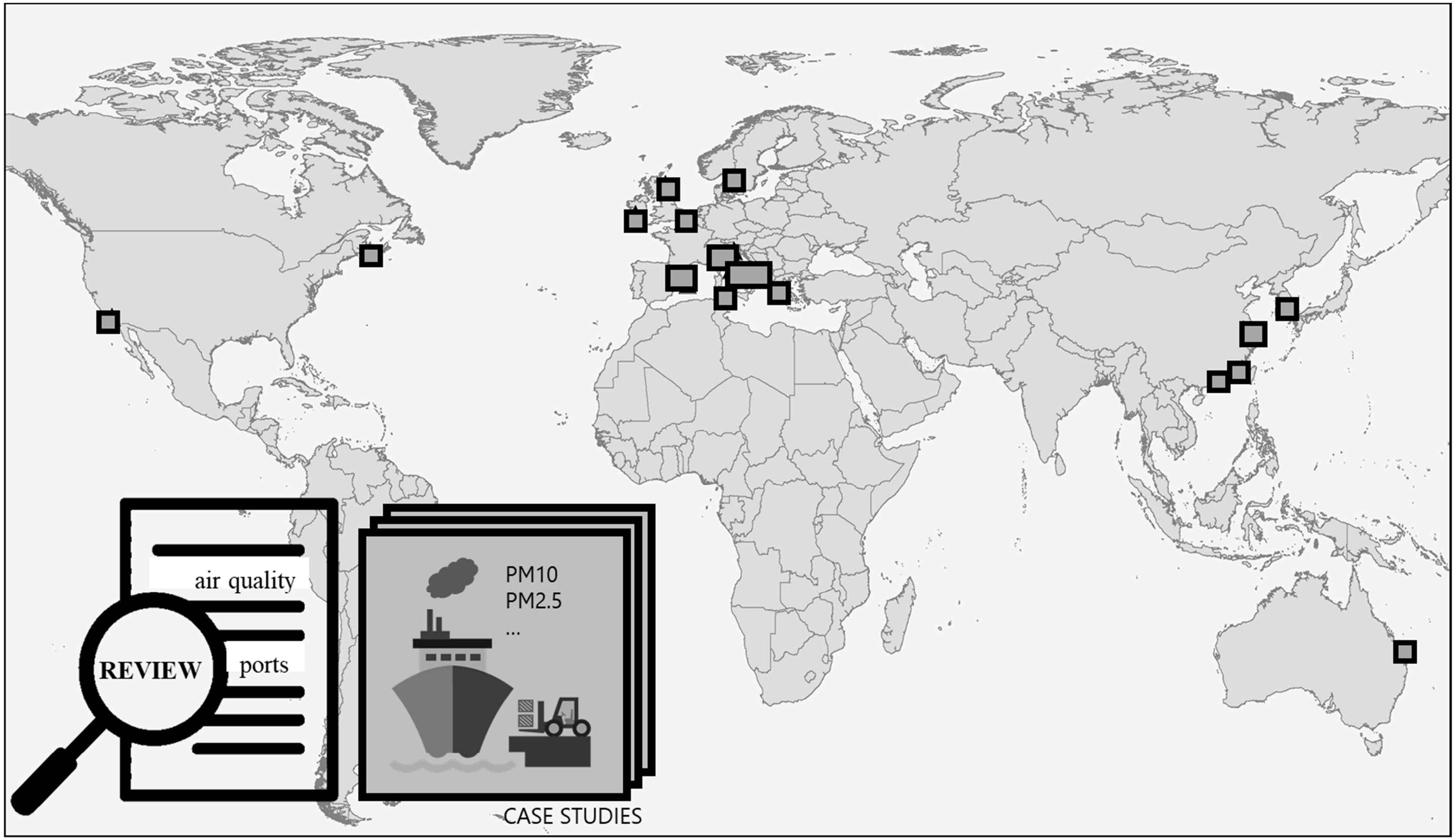
air quality
REVIEW
ports
PM10
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CASE STUDIES

Impact of harbour activities on local air quality: a review

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Abstract

Several harbour activities cause negative environmental impacts in the harbours' surrounding areas, namely the degradation of air quality. This paper intends to comprehensively review the status of the air quality measured in harbour areas. The published studies show a limited number of available air quality monitoring data in harbours areas, mostly located in Europe (71 %). Measured concentrations of the main air pollutants were compiled and intercompared, for different countries worldwilde allowinga large spatial representativeness. The higher NO_2 and PM_{10} concentrations were found in Europe - ranging between 12-107 µg/m^3 and 2-50 µg/m^3, respectively, while the higher concentrations of $PM_{2.5}$ were found in Asia (25 - 70 µg/m^3). In addition, the lower levels of SO_2 monitored in recent years suggest that current mitigation strategies adopted across Europe were very efficient in promoting the reduction of SO_2 concentrations.

Part of the reviewed studies also estimated the contributions from ship emissions to PM concentration through the application of source apportionment methods, with an average of 5-15 %. In some specific harbour areas in Asia, ships can contribute up to 7 to 26 % to the local fine particulate matter concentrations. This review confirms that emissions from the maritime transport sector should be considered as a significant source of particulate matter in harbour areas, since this pollutant concentrations are frequently exceeding the established standard legal limit values. Therefore, the results from this review boost the implementation of mitigation measures, aiming to reduce, in particular, particulate matter emissions.

Capsule

A review was performed focused on air quality over harbour areas, concluding that shipping activities have a significant contribution to PM and NO_2 emissions: the highest concentrations of NO_2 and PM_{10} are found in European harbours, ranging between 12 - 107 µg/m^3 and 2-50 µg/m^3, respectively, while the highest $PM_{2.5}$ concentrations were found in Asia, ranging from 25 to 70 µg/m^3.

Keywords: ports; ships; air pollution; SO_2, NO_2, PM_{10}, $PM_{2.5}$; source apportionment; PMF

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 et al., 2008; Eyring et al., 2010). 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 (e.g. Isakson et al., 2001; Sorte et al. 2018). The growth of ship movements and consequent release of air pollutants called also the attention to this emission source. 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. 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 (Viana et al., 2014; Monteiro et al., 2018, Ramacher et al., 2019).

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 European Union (EU) limits and World Health Organization (WHO) guidelines. In particular, the latest official air quality data released by the European Environment Agency (EEA) in 2018 indicate 19 % of PM_{10} 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, 2018).

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 levels for PM_{10} and $PM_{2.5}$ (WHO, 2016). The summary report of this database discusses the PM_{10} 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 PM_{10} above 200 µg/m^3; the cities of Cairo and

Dhaka reported PM_{10} concentrations above 150 µg/m^3, while the cities of Mumbai, Beijing and Kolkata reported PM_{10} concentrations above 100 µg/m^3.

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 Sulfur 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 sulfur 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; Karl et al., 2019; Maragkogianni et al., 2016). Moreover, the European Union has established a legal requirement limiting at 0.1 % the sulfur 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 (Ledoux et al., 2018; Schembari et al., 2012; Xu et al., 2018; Contini et al., 2015). 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 (see for example Rao et al., 2013; Chuwah 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 et al., 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; Jonsson et al., 2011) or using receptor models based on the identification of chemical tracers associated with ship emissions (Viana et al., 2009; Pandolfi et al., 2011; Cesari et al., 2014; Contini et al., 2015).

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 on 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 (Pandolfi et al., 2011; Contini et al., 2011; Cesari et al., 2014; Viana et al., 2014; Ramacher et al., 2019).

Exposure to air pollution has been associated with severe health pathologies, including asthma, lung cancer, cardiovascular diseases, and heart attacks. Ship emissions, in particular, have been associated with those pathologies (Quaranta et al., 2012; Tian et al., 2013; Corbett et al., 2007; Sofiev et al., 2018). For instance, PM emissions from marine vessels activities have been related to an increase of hospitalizations due to cardiovascular episodes (Tian et al 2013; 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 NO_2 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 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 vessels 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 current 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. In order 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 describes the selected case studies; Section 3 presents the impact of harbour areas activities on ambient SO_2, NO_2, PM_{10}, $PM_{2.5}$ concentrations; Section 4 compiles the published results of ship emissions' contributions based on receptor modelling tools and Section 5 presents the summary and conclusions.

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.

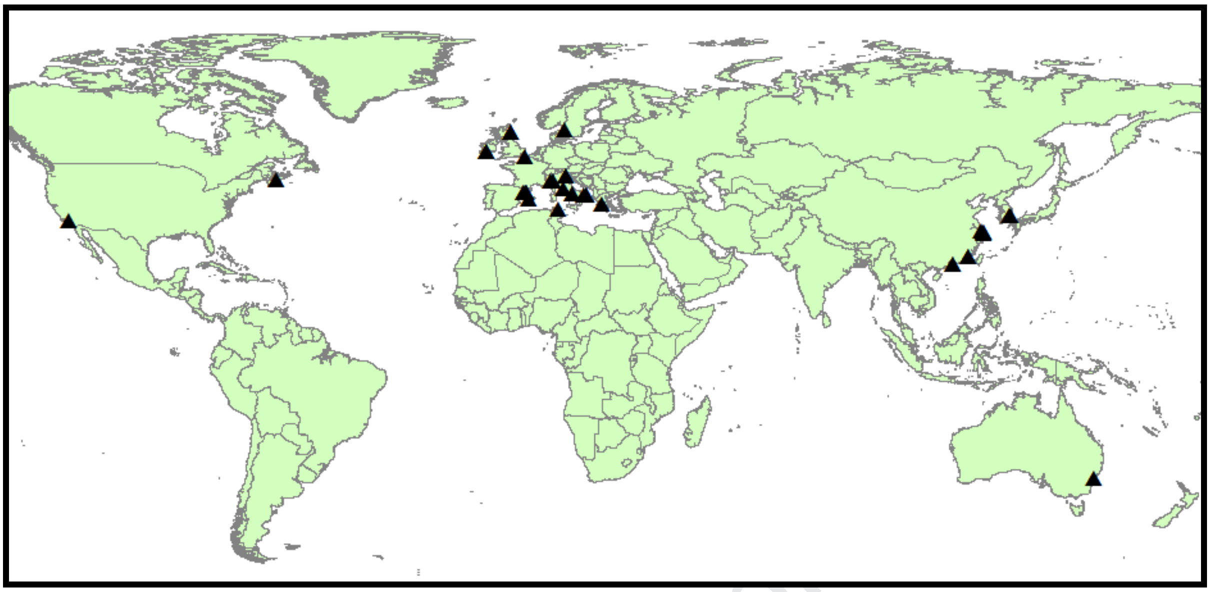


Figure 1. Location of the analysed published studies focusing on air quality over harbours.

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 located 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.

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, PM_{10} and $PM_{2.5}$ – 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, PM_{10} and $PM_{2.5}$ recorded for each case study, during a specific period.

	Reference	Case study	Period	NO_2 (µg/m^3)	SO_2 (µg/m^3)	$PM_{2.5}$ (µg/m^3)	PM_{10} (µg/m^3)
Europe	Ledoux et al. (2018)	Calais harbour (France)	2014.1 - 2014.4	22.0	3.1	NA	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
Prati et al. (2015)	Naples urban (Italy)	2012.4	44.1	5.7	NA	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
Oceania	Broome et al. (2016)	Sydney urban (Australia)	2010 - 2011	NA	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

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 nitrogen dioxide (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 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 vessels. 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, during summer time, 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 ug/m^3 above the urban background levels, while for SO_2 this value was 4.5, for background levels of 11.3 and 1.6 ug/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 (Merico et al., 2016; Donateo et al., 2014) 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 $PM_{2.5}$ concentration in harbour areas (Contini et al., 2015; Liang et al., 2016), but with limited effects on metals and PAHs concentrations

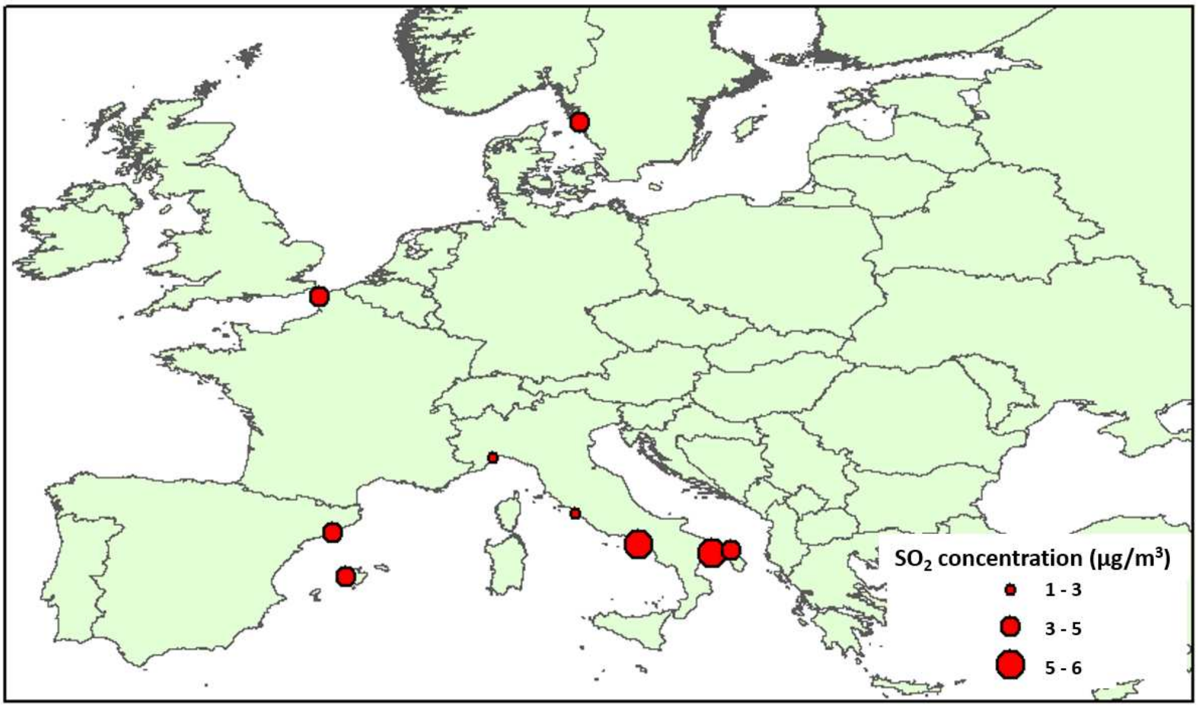
(Gregoris et al., 2016). Hong Kong harbour is subject to IMO regulation and a voluntary low sulphur program. The study focus on Brindisi (Donateo et al., 2014) indicates the impact on SO_2 concentrations of manoeuvring during the ship's arrival and/or departure. On the other hand, the hoteling phase 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 of Tunis and Shanghai (Schembari et al., 2012, Zhao et al., 2013).

Some of these studies have also revealed that ship emissions contribute more to fine particles, and especially to ultrafine particles, than to coarse aerosols (Viana et al., 2009 and Saxe and Larsen, 2004). 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 $PM_{2.5}$ 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.

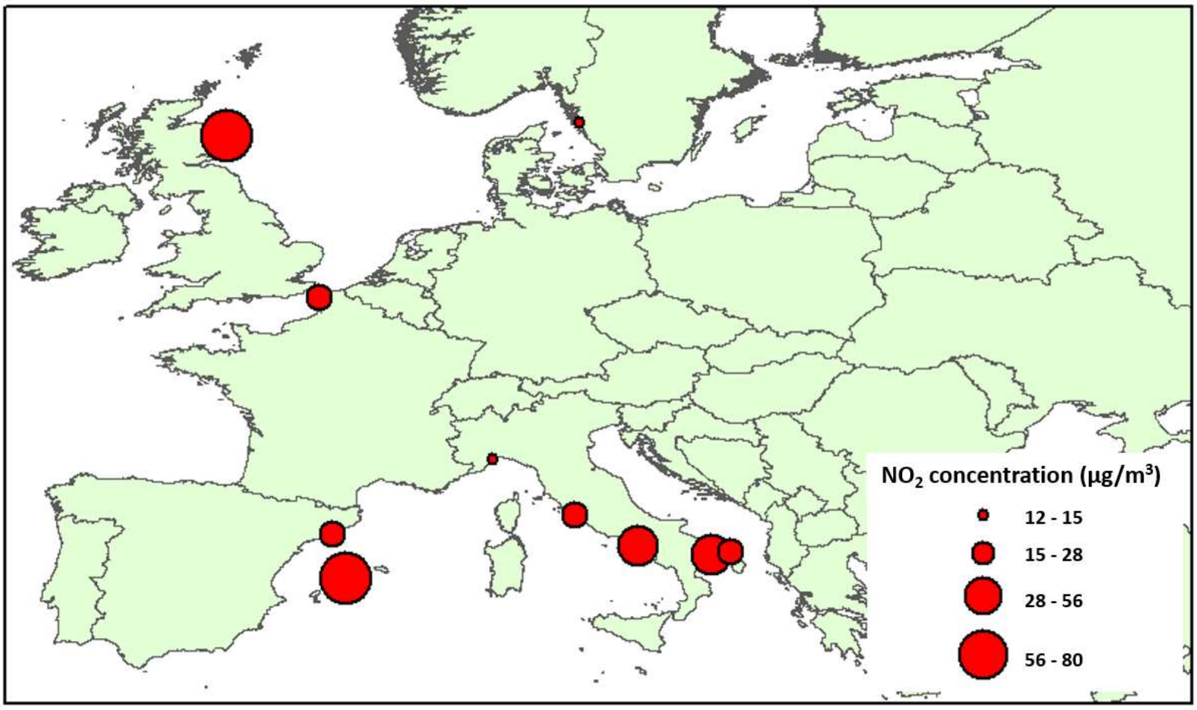
Despite being the most studied pollutants in the literature, PM, NO_2 and SO_2 are not the only pollutants affecting air quality over harbour areas. An array of other compounds can be found at significant concentrations around harbours. For instance, Vanadium (V) and nickel (Ni), as well as BC (black 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).

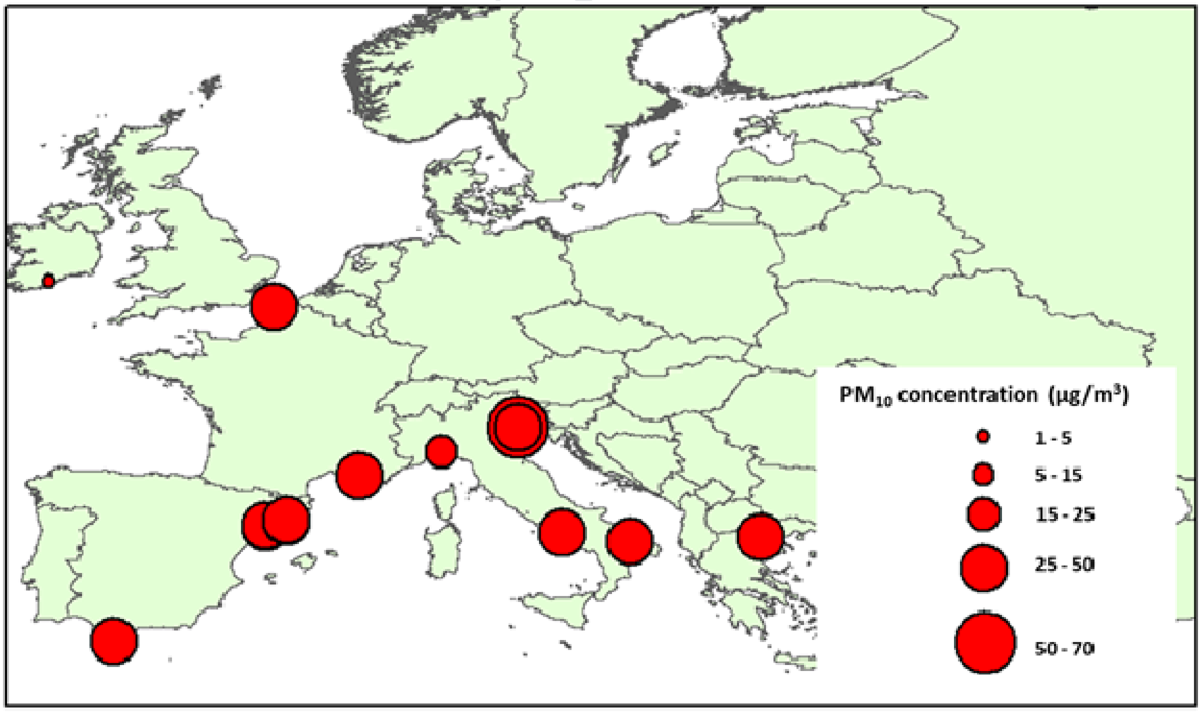
In the particular case of Europe, the high number of available studies addressing marine transport emissions and its related air quality denotes the relevance of this issue in Europe. Figure 2 shows the geographical distribution of the case studies located in Europe and the respective magnitude of the pollutant's concentration measured, regarding the pollutants considered in this review: SO_2, NO_2, PM_{10} and $PM_{2.5}$. Palma de Mallorca and Aberdeen show the highest NO_2 levels, while for SO_2 the highest values are recorded in Civitavecchia and Naples. In the case of Palma de Mallorca, data comes from a monitoring station located in the city area; however, it will be influenced by the harbour zone. Regarding PM, Figure 2c) and 2d) show a higher concentration measured in Mediterranean cities.



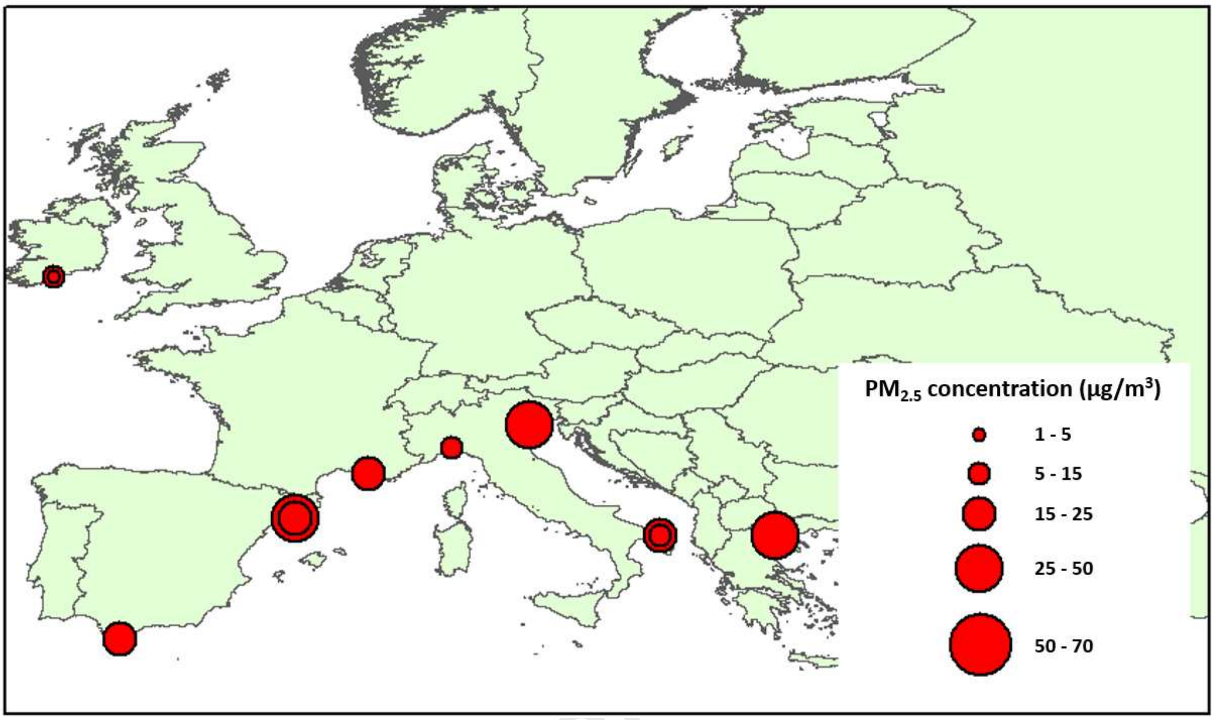
a)


NO_2 concentration (µg/m^3)
12 - 15
15 - 28
28 - 56
56 - 80

b)


PM_{10} concentration (µg/m^3)
1 - 5
5 - 15
15 - 25
25 - 50
50 - 70

c)



d)

Figure 2. Average concentration values measured for each European case study, during the corresponding sampling period, for the distinct pollutants: a) SO_2, b) NO_2, c) PM_{10} and d) $PM_{2.5}$.

4. Contribution of ship emissions to PM

A set of studies addressed the contribution of shipping 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 V 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 S1 of the supplementary material).

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	$PM_{2.5}$	13
		Thessaloniki harbour (Greece)	NA	PMF	$PM_{2.5}$	9
	Scerri et al. 2018	Malta urban (Republic of Malta)	2016	PMF	$PM_{2.5}$	5
	Manousakas et al (2017)	Patras harbour (Greece)	2011	PMF	$PM_{2.5}$	10
	Manousakas et al (2017)	Patras harbour (Greece)	2011	PMF	PM_{10}	15
	Cesari et al. (2014)	Brindisi harbour (Italy)	2012.6 - 2012.10	PMF	$PM_{2.5}$	15.3*
	Bove et al. (2014)	Genoa urban (Italy)	2011.5 - 2011.10	PMF	$PM_{2.5}$	11
	Merico et al. (2017)	Brindisi urban (Italy)	2012	V	$PM_{2.5}$	2.8
	Gregoris et al. (2016)	Venice harbour (Italy)	2007 - 2013	PMF	PM_{10}	2.5
	Gregoris et al. (2016)	Venice harbour (Italy)	2007 - 2013	PMF	$PM_{2.5}$	3.3
	Merico et al (2017)	Rijeka harbour (Croatia)	2014	PMF	$PM_{2.5}$	10.5
	Merico et al. (2017)	Rijeka urban (Croatia)	2012	V	$PM_{2.5}$	1.1
	Merico et al. (2017)	Rijeka harbour (Croatia)	2013-2014	V	$PM_{2.5}$	0.5
	Pérez et al. (2016)	Barcelona harbour (Spain)	2011.02 - 2011.12	PMF	$PM_{2.5}$	14
			2011.02 - 2011.12	PMF	PM_{10}	8
		Barcelona urban (Spain)	2011.02 - 2011.12	PMF	PM_{10}	4
			2011.02 - 2011.12	PMF	$PM_{2.5}$	6
	Amato et al. (2009)		2003 -2007	PMF	$PM_{2.5}$	6-7
			2003 -2007	PMF	PM_{10}	5
	Pandolfi et al. (2011)	Gibraltar Strait urban (Spain)	2003 -2007	PMF	$PM_{2.5}$	5-10
			2003 -2007	PMF	PM_{10}	3-7
	Viana et al. (2009)	Melilla urban (Spain)	2007	PMF	$PM_{2.5}$	14
			2007	PMF	PM_{10}	2-4
	Alastuey et al. (2007)	Tarragona harbour (Spain)	2004.09 - 2005.09	FA	PM_{10}	16

	Healy et al. (2010)	Cork harbour (Ireland)	2008.5 - 2008.8	PMF	$PM_{2.5}$	2
	Hellebust et al. (2010)		2007.5 - 2008.4	MLR	PM_{10}	< 5*
	Healy et al. (2009)		2008.8	PMF	$PM_{2.5}$	2
Asia	Xu et al. (2018)	Xiamen harbour - industrial (China)	2015.4 - 2016.1	PMF	$PM_{2.5}$	26*
		Xiamen urban (China)	2015.4 - 2016.1	PMF	$PM_{2.5}$	13*
	Mamoudou et al. (2018)	Yangshan Island harbour (China)	2016	PMF	$PM_{2.5}$	2.4
	Mamoudou et al. (2018)	Yangshan Island harbour (China)	2016	V	$PM_{2.5}$	0.23
	Tao et al. (2017)	Zhuhai urban (China)	2014 -2015	PMF	$PM_{2.5}$	18
	Zhao et al. (2013)	Shanghai harbour (China)	2011	V	$PM_{2.5}$	4.23
	Jeong et al. (2017)	Busan urban (Korea)	2013	PMF	$PM_{2.5}$	7
			2013	CMB	$PM_{2.5}$	1
			2013	PCA/ APCS	$PM_{2.5}$	5
	Yau et al. (2013)	Kwai Chung and Tsing Yi urban (Hong Kong)	2009.8 - 2010.3	PMF	$PM_{2.5}$	19
			2009.8 - 2010.3	PCA/ APCS	$PM_{2.5}$	25
North America	Gibson et al. (2013)	Halifax urban (Canada)	2011.7 - 2011.8	PMF	$PM_{2.5}$	3
	Minguillón et al. (2008)	Angeles Long Beach harbour (California)	2007	CMB	$PM_{2.5}$	< 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	$PM_{2.5}$	8.8

* Mixed industrial source and shipping

PMF is the most commonly 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 (Cesari et al., 2016; Belis et al., 2014; Viana et al., 2008; Liang et al., 2016). 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 $PM_{2.5}$ concentrations at Yangshan Harbor, when compared to the PMF method.

The maximum $PM_{2.5}$ contribution estimated by PMF for ship emissions was 26.1 % in Xiamen, followed by Kwai Chung urban (19 %) and Zhuhai (18 %). For PM_{10}, the major contributors were the harbours of Tarragona (16 %), followed by Patras (15 %) and Barcelona (8 %). The ship emission contributions to $PM_{2.5}$ differed significantly between European, Asian and North American harbour cities, ranging from 2-14 %, 1–25 % and 3-29 % of $PM_{2.5}$, respectively. On the other hand, the ship emission contributions to PM_{10} in Europe ranged between 3-16 %. In overall, a greater ship contribution was seen in East Asian harbour cities compared to Europe, as a result 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 $PM_{2.5}$ 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 $PM_{2.5}$ (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.

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 $PM_{2.5}$ emissions was estimated by PMF to be 45 % in Thessaloniki, followed by Barcelona and Patras harbours (36 and 34 % respectively). For PM_{10}, 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 PM_{10} emissions (Saraga et al., 2019; Manousakas et al., 2017; Pérez et al., 2016; Alastuey et al., 2007). 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 $PM_{2.5}$ 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 (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 $PM_{2.5}$ emissions in Brindisi, followed by 14 and 12 % in Barcelona and Tarragona harbours, respectively. Regarding PM_{10}, 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 $PM_{2.5}$ contribution estimated by PMF from natural sources was 27 % of $PM_{2.5}$ in the Gibraltar harbour, followed by Busan (18 %) and Barcelona urban (17 %) (Pérez et al. 2016; Pandolfi et al. 2011; Jeong et al. 2017). 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 $PM_{2.5}$, respectively.

5. Summary and 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 PM_{10}, $PM_{2.5}$, 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 PM_{10} concentrations varied between 2 and 62 µg/m^3. The maximum concentrations for all the considered pollutants where found in European harbour areas (e.g. maximum concentrations of 107, 47, 37 and 62 µg/m^3 for NO_2, SO_2, $PM_{2.5}$ and PM_{10}, 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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, in particular 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 in the near future 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.

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- Maritime sector emissions are a main source of air pollution over harbour areas
- The highest concentrations of NO_2 and PM_{10} are found in European harbours
- Recent mitigation strategies in the European territory have proved their efficiency
- PMF is the most common method for source-apportionment among all case studies