Atmospheric Environment 90 (2014) 96–105

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Review

Impact of maritime transport emissions on coastal air quality in Europe

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HIGHLIGHTS

- Shipping contributions to European coastal air quality degradation are reviewed.
- Maritime transport is a significant and increasing source of air pollutants.
- Chemical tracers are available for use as markers in receptor models.
- Mitigation strategies are effective and should be implemented on EU-scale.
- Research gaps are identified.

GRAPHICAL ABSTRACT

ARTICLE INFO

Article history:
Received 2 October 2013
Received in revised form
19 March 2014
Accepted 24 March 2014
Available online 24 March 2014

Keywords:
Source apportionment
Vessels
Mitigation strategies
Harbour operations
Shore power

ABSTRACT

Shipping emissions are currently increasing and will most likely continue to do so in the future due to the increase of global-scale trade. Ship emissions have the potential to contribute to air quality degradation in coastal areas, in addition to contributing to global air pollution. With the aim to quantify the impacts of shipping emissions on urban air quality in coastal areas in Europe, an in depth literature review was carried out focussing on particulate matter and gaseous pollutants but also reviewing the main chemical tracers of shipping emissions, the particle size distribution of ship-derived particulates and their contributions to population exposure and atmospheric deposition. Mitigation strategies were also addressed. In European coastal areas, shipping emissions contribute with 1–7% of ambient air PM_{10} levels, 1–14% of $PM_{2.5}$, and at least 11% of PM_1. Contributions from shipping to ambient NO_2 levels range between 7 and 24%, with the highest values being recorded in the Netherlands and Denmark. Impacts from shipping emissions on SO_2 concentrations were reported for Sweden and Spain. Shipping emissions impact not only the levels and composition of particulate and gaseous pollutants, but may also enhance new particle formation processes in urban areas.

1. Introduction

Maritime transport of goods is a relatively clean form of transportation per kilogram of material, and it is therefore currently gaining relative weight with respect to air and road transport (Micco and Pérez, 2001; Grewal and Haugstetter, 2007). This form of

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http://dx.doi.org/10.1016/j.atmosenv.2014.03.046

transportation has also been increasing (and will most likely continue to do so in the future) due to the globalization of manufacturing processes and the increase of global-scale trade (Corbett and Fishbeck, 2000; Marmer et al., 2009; US-EPA, 2009). However, emissions from the marine transport sector contribute significantly to air pollution globally (Wang et al., 2008; EEA, 2012). Around 15% of global anthropogenic NO_x and 5–8% of global SO_x emissions are attributable to oceangoing ships (Eyring et al., 2005; Corbett et al., 2007). Because nearly 70% of ship emissions are estimated to occur within 400 km of land (Endresen et al., 2003), ships have the potential to contribute significantly to air quality degradation in coastal areas. In addition, emissions are also generated while vessels are at berth, given that the main engines are not always switched off by all types of vessels (De Meyer et al., 2008). Large efforts have been made in Europe to reduce other types of emission sources (industrial, power generation, etc.), and this results in an increase of the relative weight of shipping emissions to the total of anthropogenic emissions. Only under new and strong climate (and air pollution) policies, energy intensity improvements could offset the growth in ship emissions (IEA et al., 2009). Ship emissions affect not only major ports, but also medium and small-scale ones (Viana et al., 2009). Despite this, even though shipping contributes significantly to the international transportation sector, its emissions are not well quantified and are one of the least regulated anthropogenic sources (IMO, 2008). Several studies point towards the need of international regulations on ship emissions, as those active in Europe, where the land based emissions of sulphur have been successfully reduced since 1980's, and where over the first decade of the 21st century total SOx emission have fallen by 54% in the EU (EEA, 2011). In this context, it is necessary to investigate the current impact of the ship emissions on the ambient air levels of primary and secondary aerosols, and how the predicted future growth of ship traffic and the geographical expansion of waterways and ports, possibly combined with international regulations, are going to affect the atmospheric composition (Becagli et al., 2012). In order to design and implement effective regulation to minimise environmental impacts of these emissions, detailed knowledge is necessary of their effects on climate and of their contribution to atmospheric pollution (Marmer et al., 2009).

The assessment of shipping emissions on the global and regional scales is of interest due to their various impacts on human health, climate and ecosystems. A detailed description of these impacts may be found in EEA (2013). Based on this assessment, it seems evident that urgent efforts should be made to reduce emissions from the maritime transport sector. Different approaches are used in different countries to reduce shipping emissions; however, actions to address these emissions have not yet achieved the goals for protecting human health (US-EPA, 2009).

Thus, the aim of the present work is to review existing studies dealing with the impact of shipping emissions on air quality in European coastal areas, in order to obtain a quantitative picture of these impacts.

2. Results and discussion

An in depth literature review was carried out focussing on the assessment of the impacts of shipping emissions and activities on urban air quality in European coastal areas. The summary of the main findings from each of the works reviewed, as a function of the specific topics addressed, may be found below. The details of the studies reviewed may be found in EEA (2013).

2.1. Chemical tracers of shipping emissions

Numerous studies in the literature have succeeded in identifying specific tracers of shipping emissions. As tracers of combustion processes based on crude oil as the main fuel, vanadium (V) and nickel (Ni) are generally identified as markers of shipping emissions. In addition, other markers identified are thorium (Th) (Querol et al., 1997), lead (Pb) (Isakson et al., 2001; Hellebust et al., 2010), zinc (Zn) (Isakson et al., 2001) and sulphate (SO_4^{2-}) (Viana et al., 2008; Becagli et al., 2012). However, the direct identification of shipping emissions by means of these tracers is complex, given that they are also markers for other types of combustion processes such as energy generation, petroleum refinery and other types of industrial processes, which are located on land and very frequently in the vicinity of harbour areas. Consequently, more detailed analyses have been carried out focussing not only on tracer species, but on tracer ratios (in terms of airborne concentration), which might aid in the more exhaustive identification of shipping emissions by means of modelling approaches such as, e.g., the multi-linear engine (Reinikainen et al., 2001).

In Genoa (Italy) (Mazzei et al., 2008), V/Ni concentration ratios were calculated by means of receptor modelling tools (PMF), which were found to be fairly constant for the three size fractions analysed (PM_{10}, $PM_{2.5}$, PM_1). It was concluded that heavy oil combustion may be identified by the concentration ratio V/Ni = 3.2 ± 0.8 in all PM fractions. From an emission point of view, a wide V/Ni ratio (2.3–4.5) was measured by direct sampling at the exhausts of different auxiliary ship engines fed by different fuels (Nigam et al., 2006), and from the main propulsor ship engine at different speed mode (Agrawal et al., 2008a,b). Similar results were obtained for ambient air concentrations in Spain across the Gibraltar Strait (Viana et al., 2009), where valid tracers of commercial shipping emissions in ambient PM_{10} and $PM_{2.5}$ were ratios of V/Ni = 4 ± 1. The ratio V/EC < 2 was also suggested as a tracer in this study. Characteristic ratios obtained from land sources (non vessel-derived) for their study area were V/Ni = 12 and V/EC > 8, excluding the influence of shipping emissions (by means of wind rose analysis). Other ratios (V/S (Viana et al., 2007), La/Ce (Moreno et al., 2008), Zn/Ni and Pb/Zn (Isakson et al., 2001), OC/EC (Fridell et al., 2008)) and tracers (Pb, Zn) were also tested, but did not always correlate with this source (Viana et al., 2009). Also in the South of Spain, shipping emissions were characterised by La/Ce concentration ratios between 0.6 and 0.8 and V/Ni ratios around 3 for both PM_{10} and $PM_{2.5}$ (Pandolfi et al., 2011). In contrast, elevated La/Ce values (1–5) were attributable to emissions from refinery zeolitic fluid catalytic converter plant, and low average V/Ni values (around 1) resulted mainly from contamination from stainless steel plant emissions. Finally, on the island of Lampedusa (Italy) (Becagli et al., 2012), PM_{10} samples influenced by ships were characterized by elevated Ni and V soluble fraction (80% for aerosol from ships, versus about 40% for crustal particles), high V and Ni to Si ratios, and values of soluble V > 6 ng/m^3. Data suggested a characteristic non sea-salt SO_4^{2-}/V concentration ratio in the range 200–400 for ship emission aerosols in summer at Lampedusa. The Ni/Si ratio was one order of magnitude higher than expected for crustal particles in 79% of the measured PM_{10} samples. As expected, V and Ni concentrations in a ship aerosol event characterised in this study displayed a maximum in the finest mode (diameter < 0.4 μm). Conversely, their concentrations peaked at larger size (1.1–2.1 μm for Ni, and 0.4–0.7 μm for V) during a Saharan dust event monitored during the same period.

These particulate tracers were found to correlate with gaseous tracers in a number of studies. In Gothenburg, Ni, Pb, V and Zn were shown to have positive correlation with NO emissions from ships (Isakson et al., 2001). In addition, results show that there is a considerable local impact of shipping-related emissions on air quality in the vicinity of major harbours, in particular, from NO_x, SO_2, PM, and VOC emissions (Saxe and Larsen, 2004; Eyring et al., 2010). In addition, shipping emissions also impact particle

number concentration as a contributor to increased levels of ultrafine particles by causing nucleation episodes at midday in Southern European environments (Reche et al., 2011).

2.2. Particle size distribution

The grain size distribution of ship-derived aerosols in coastal areas has not been the subject of a large number of studies, according to the results of this review, probably due to the difficulty related with uniquely identifying these contributions. Direct measurements of sub-micrometer particles in ship plumes reveal a bimodal number size distribution (with modes at 40 nm and 70 nm), which are strongly enhanced as compared to background air (Isakson et al., 2001). In ambient air, shipping emissions contribute more strongly to fine than to coarse aerosols (Viana et al., 2009), and especially to ultrafine particles (Saxe and Larsen, 2004), although in some regions the contributions of ship emissions to PM_{10} and to $PM_{2.5}$ have been seen to be basically the same within experimental uncertainty (Contini et al., 2011). Indeed, primary particles emitted by ships are predominantly in the submicron size fraction (Petzold et al., 2008; Healy et al., 2009). Elements arising from heavy oil combustion (V, Ni, Al, Fe) are distributed in the sub-micrometric fraction of the aerosol, and the metals are present as free metals, carbonates, oxides hydrates or labile complex with organic ligands (Becagli et al., 2012). Shipping emissions have been seen to contribute to increased particle number concentrations, and are thus dominated by ultrafine particles (Reche et al., 2011).

2.3. Impact of shipping emissions on ambient PM_x concentrations

The impact of shipping emissions on PM_x concentrations is mainly calculated by applying source apportionment tools, either by means of dispersion and receptor models or by using chemical tracer methods. When using receptor models, shipping emissions generally appear mixed with other combustion sources (Viana et al., 2008), most likely due to the modelling challenge of separating sources with common tracers (Jalkanen et al., 2009) and/or to the absence of specific marker species for shipping emissions in the input data sets. Single particle analysis techniques have shown that identification of specific particle types may provide information which can improve receptor model results; however, results are not always quantitative. The separation of sources with common tracers was identified as a research gap.

The number of studies addressing the impact of shipping emissions on airborne PM differs across European coastal regions. In Gothenburg (Isakson et al., 2001), multivariate analysis was applied to sub-micron particle data and increased concentrations due to ship emissions were quantified for ships entering the inner part of the harbour. It was observed that exposure to transient particles (less than 0.1 μm in diameter) in this part of the harbour increased by a factor of 3 in number concentration when a ship plume was recorded. In Cork (Ireland) (Hellebust et al., 2010), fresh ship plumes were not found to make a significant contribution to primary $PM_{2.5-0.1}$ concentrations adjacent to the shipping channel (less than 5% of ambient $PM_{2.5-0.1}$ mass, indeed more likely less than 1%, <0.5 μg/m^3), but this was partially attributed to the ultrafine nature of ship emissions (Isakson et al., 2001; Petzold et al., 2008; Healy et al., 2009). The authors concluded that the low contribution of this component to total $PM_{2.5-0.1}$ mass does not indicate that the local air quality is not influenced by frequent ship emissions, and that this contribution should be measured in terms of particle number or toxicity, instead of mass. In the Copenhagen harbour, ships were estimated to contribute with insignificant mass concentrations of PM in the populated areas near the harbour, with a contribution to the annual PM_{10} mean of 0.08–0.15 μg/m^3 (Saxe and Larsen, 2004). Also along the Atlantic coast of Europe, evidence of shipping emissions was found in The Netherlands (Mooibroek et al., 2011) by means of source apportionment with positive matrix factorization (PMF (Paatero, 1997)), in the form of a mixed oil combustion source considered by the authors to include shipping emissions along with other potential combustion sources (e.g., municipal district heating power plants and industrial power plants using heavy oil). The maximal contribution registered for this source was 2% of $PM_{2.5}$ as an annual mean (0.4 μg/m^3). By means of a dispersion modelling approach, one study (Hammingh et al., 2012) focussing on North Sea shipping evidenced that sea shipping contributions to $PM_{2.5}$ concentrations depend on the proximity of a country to the North Sea and the busy shipping lanes, and can be as high as 7% in certain coastal areas. According to this work, the contribution to country averages is the highest in the Netherlands and the United Kingdom with 5% (0.11 μg/m^3) and 4% (0.24 μg/m^3), respectively. This contribution is decreasing to 3% (0.17 μg/m^3) for Belgium, 2% (0.06 μg/m^3) for France, and 2% (0.08 μg/m^3) Germany. The lowest contributions were modelled for Sweden and Norway with about 1% (around 0.01–0.02 μg/m^3). The contribution in Luxembourg and Switzerland is about 2% and 1% of $PM_{2.5}$, respectively.

In the Mediterranean basin, on the other hand, a particularly high contribution of heavy oil combustion to PM_1 (about 5 μg/m^3, over 25 μg/m^3, 20%) was found in the summer at one coastal station in Genoa (Northern Italy) (Mazzei et al., 2008). In Venice, the contribution from shipping emissions to $PM_{2.5}$ and to PM_{10} was estimated to range between 1% and 8% (Contini et al., 2011). Also in the Mediterranean region, the contribution of shipping emissions to urban background PM levels was quantified by PMF in Melilla, located in the vicinity of the Gibraltar Strait (Viana et al., 2009). Results evidenced that shipping emissions contributed with 2% and 4% of mean annual PM_{10} levels (0.8 μg/m^3 primary particles and 1.7 μg/m^3 secondary particles, with 20% uncertainty) and 14% of mean annual $PM_{2.5}$ levels (2.6 μg/m^3). These results were of the same order of magnitude as others from a nearby location (Southern Spain) (Pandolfi et al., 2011), where the direct contribution from shipping in the Bay of Algeciras was estimated at 1.4–2.6 μgPM_{10}/m^3 (3–7%) and 1.2–2.3 μg$PM_{2.5}$/m^3 (5–10%). The total contribution from shipping (primary emissions + secondary sulphate aerosol formation) reached 4.7 μgPM_{10}/m^3 (13%) and 4.1 μg$PM_{2.5}$/m^3 (17%). Also along the Mediterranean coast of Spain, shipping emissions were identified in Barcelona (Amato et al., 2009) as part of an oil combustion source identified by receptor modelling tools. The oil combustion source, including shipping emissions, accounted on the annual mean for 5% of ambient PM_{10} levels, 6% of $PM_{2.5}$ and 8% of PM_1. Finally, on the isle of Lampedusa (Becagli et al., 2012) (South of the Sicily channel), ship emissions accounted as a summer average for at least 30% of the total non-sea-salt SO_4^{2-} (1.2 μg/m^3, with a maximum of 47%), 3.9% of PM_{10} (with a maximum of 15%), 8% of $PM_{2.5}$, and 11% of PM_1.

In addition to contributions in terms of particle mass, current research evidences that shipping emissions may also enhance new particle formation in urban areas and thus contribute to other forms of air quality degradation. As such, in Barcelona (Spain) hourly SO_2 maxima (originating from the harbour area) coincided with the noon peak of particle number concentration (N), suggesting that SO_2 from shipping could be a major contributor to nucleation episodes at midday (Reche et al., 2011), although research is ongoing in this field. Finally, the contribution of shipping emissions to PAH concentrations was also assessed (Contini et al., 2011), with results showing that the direct contribution of ships traffic to PAHs in the gas phase is 10%.

Fig. 1 and Table 1 summarise the results from the literature review, showing mean annual contributions from shipping emissions to air quality (PM_{10}, $PM_{2.5}$, PM_1) across Europe. Based on the studies reviewed, shipping emissions contribute to ambient PM levels (annual means) in European coastal areas with 1–7% of PM_{10}, 1–14% of $PM_{2.5}$ (reaching a summer maximum of 20% in Genoa, Italy) and at least 11% of PM_1 (this size fraction was only monitored at 1 location). Thus, it is evident that the impact of shipping activities increases with decreasing particle size. For the sake of comparison, contributions reported for non-European harbours (USA) were <5% of $PM_{2.5}$ in Los Angeles (Minguillón et al., 2008) and 4–6% of $PM_{2.5}$ in Seattle (Kim and Hopke, 2008). Spatially, Fig. 1 suggests that shipping contributions to urban air quality degradation (regarding PM) are higher in Mediterranean cities than in Atlantic coastal areas, although this result could be influenced by the more limited number of studies available for the Northern (7 studies) than the Southern regions of Europe (11 studies). In addition, it must be highlighted that results were obtained using different approaches and methodologies in the different regions, given that no common EU-wide methodology is currently available. It should also be highlighted that the North Se and the Baltic Sea are SECA areas, and thus SO_2 emissions are lower and this would be expected to result in lower secondary inorganic particle formation.

As a result, this review evidences the need for a novel approach to tackle the issue of comparability between studies and regions in a consistent way across Europe. Measurement and modelling studies should aim to be applied in a homogeneous manner across Europe, if data are to be compared across regions. Currently, North Sea studies focus mainly on shipping emissions of gaseous pollutants, while Southern studies deal with the chemical composition of particulates. A more standardised application of modelling studies could possibly be the best way to move towards comparable data on the impact of shipping emissions on air quality. Also, the application of similar methodologies across different regions would improve comparability between results.

2.4. Impact of harbour loading and unloading operations on ambient PM_x concentrations

The impact on air quality of harbour operations (loading and unloading of vessels, fuelling, etc.) was assessed in the literature by a much more limited number of studies. However, all studies agree on the relevance of this impact. In 2003, evidence was found (Viana, 2003) of the impact from handling operations (loading/unloading) on ambient PM_{10} and $PM_{2.5}$ concentrations in the form of re-suspension of mineral dust (road dust), even though no significant differences in PM levels or chemical composition were found between the harbour and city background under study. In the Los Angeles harbour (US), vehicular sources together with road dust explained up to 54% of the PM mass, whereas ship contribution was lower than 5% of total fine PM mass (Minguillón et al., 2008). Results clearly indicated that, although ship emissions may be significant, PM emissions in the area of the largest US harbour were dominated by road transport. Finally, loading and unloading of tankers and cargo vessels was found to contribute substantially to harbour emissions (Alastuey et al., 2007; Moreno et al., 2007; Eyring et al., 2010).

2.5. Impact of shipping emissions on gaseous pollutant concentrations

The impact of shipping emissions on gaseous pollutant concentrations is mostly carried out by dispersion modelling tools, given that observational source apportionment tools are less

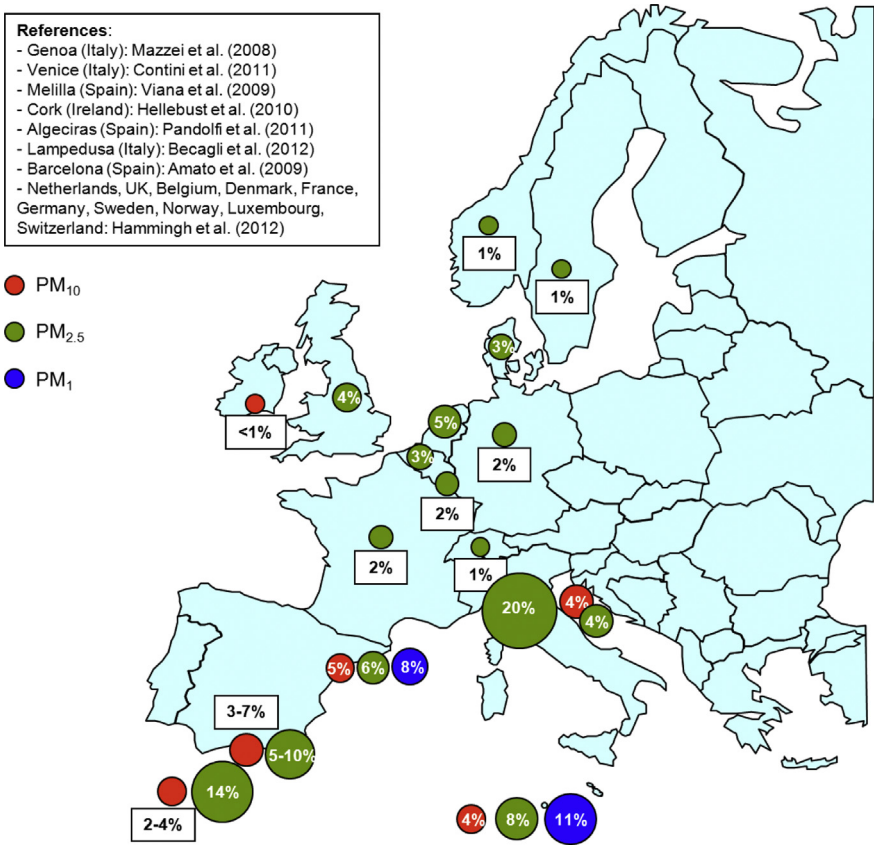

Fig. 1. Contribution from shipping emissions to air quality (PM_{10}, $PM_{2.5}$ and PM_1) across Europe.

Table 1
Summary of shipping contributions to air quality across Europe, focussing on airborne particles. OC: organic carbon. Contributions are expressed as % to the annual mean.

Airborne particles				
Reference	Source	Contribution	Size fraction/PM component	Location
Kim & Hopke	Oil combustion	4–6%	$PM_{2.5}$	Seattle (US)
Mazzei et al. (2008)	Oil combustion	20%	PM_1	Genoa (Italy)
Minguillón et al. (2008)	Shipping	<5%	OC	Los Angeles (US)
		<5%	$PM_{2.5}$	Los Angeles (US)
Viana et al. (2008)	Oil combustion	10–30%	PM_{10} and $PM_{2.5}$	Europe
Amato et al. (2009)	Oil combustion	5%	PM_{10}	Barcelona (Spain)
		6%	$PM_{2.5}$	Barcelona (Spain)
		8%	PM_1	Barcelona (Spain)
Viana et al. (2009)	Shipping	2–4%	PM_{10}	Melilla (Spain)
		14%	$PM_{2.5}$	Melilla (Spain)
Hellebust et al. (2010)	Shipping	<1%	$PM_{2.5\text{-}10}$ and $PM_{0.1\text{-}2.5}$	Cork (Ireland)
Contini et al. (2011)	Shipping	1–8%	PM_{10}	Venice (Italy)
		1–8%	$PM_{2.5}$	Venice (Italy)
Pandolfi et al. (2011)	Shipping	3–7%	PM_{10}	Algeciras (Spain)
		5–10%	$PM_{2.5}$	Algeciras (Spain)
Becagli et al. (2012)	Shipping	30%	nss SO_4^{2-}	Lampedusa (Italy)
		3.9%	PM_{10}	Lampedusa (Italy)
		8%	$PM_{2.5}$	Lampedusa (Italy)
		11%	PM_1	Lampedusa (Italy)
Hammingh et al. (2012)	Shipping	1–5%	$PM_{2.5}$	North Sea coastal countries
		5%	$PM_{2.5}$	The Netherlands
		4%	$PM_{2.5}$	UK
		3%	$PM_{2.5}$	Belgium
		3%	$PM_{2.5}$	Denmark
		2%	$PM_{2.5}$	France
		2%	$PM_{2.5}$	Germany
		2%	$PM_{2.5}$	Luxembourg
		1%	$PM_{2.5}$	Norway
		1%	$PM_{2.5}$	Sweden
		1%	$PM_{2.5}$	Switzerland

useful when dealing with gaseous pollutants. Nitrogen oxide emissions from North Sea shipping were expected to be responsible for 7–24% of country-average nitrogen dioxide concentrations in North Sea coastal countries, in an estimation for the year 2030 (Hammingh et al., 2012). Nitrogen oxide emissions from ships are also responsible for 1–5% of the fine particulate matter concentrations ($PM_{2.5}$) in the North Sea countries. Compared with the contribution made by North Sea shipping to nitrogen dioxide concentrations, the contribution to particulate matter concentrations is relatively lower due the relatively higher contributions from various land-based sources. The estimated contribution to country averages in 2030 is the highest in the Netherlands and Denmark with 24% and 19%, respectively, and the lowest in Germany and France with 7% and 8%, respectively. The contribution to the country average concentrations in Ireland is around 7%, but absolute concentrations are relatively low (Hammingh et al., 2012). In Rotterdam (The Netherlands), harbour activities did not result in elevated ammonium sulphate concentrations in the city (due to the low sulphur concentrations, or simply because these contributions were not distinctly identified) (Keuken et al., 2013), but near the waterways of the harbour shipping was estimated to cause an enhancement of the surface NO_2 mixing ratio of 5–7 ppb (Keuken et al., 2005). In Gothenburg, increases in SO_2 concentrations were only reported as relative increases due to ship emissions with respect to background concentrations and not in absolute terms (Isakson et al., 2001). Also along the Northern coast of Europe, in the Baltic Sea, NO_x emitted by ships in the port of Copenhagen contributed substantially to the overall NO_x pollution in central Copenhagen, exceeding 200 $\mu g/m^3$ of NO_x and causing values of 50–200 $\mu g/m^3$ over several square kilometres of central Copenhagen (Saxe and Larsen, 2004). In Barcelona (Reche et al., 2011), midday increases in SO_2 concentrations were detected coinciding with the inland transport of shipping emissions by the sea breeze. Mean hourly SO_2 levels reached during midday peaks were a factor of 3–4 higher than the mean SO_2 concentrations measured during the rest of the day. Finally, SO_2 concentrations were also measured in Mediterranean harbours after 2010 (Schembari et al., 2012), with the aim to test the efficiency of mitigation strategies linked to sulphur fuel content. These results are presented in Section 6 below.

As a reference, in the US, it was estimated that ship manoeuvring in harbours contributes about 6% of NO_x and 10% of SO_2 to total shipping emissions (Corbett and Fischbeck, 1997).

Shipping contributions to air quality in the central Mediterranean basin near the surface and at higher atmospheric levels (as opposed to at coastal areas) was also assessed for the summer months (Marmer and Langmann, 2005). The authors concluded that locally released NO_x is mainly responsible for the production of ozone, and that switching off the release of NO_x by ships in the model reduced surface ozone concentration by 15% in this area. The formation of HNO_3 and HCHO in the experiment was then reduced by 66% and 24%, respectively. OH concentration was simultaneously reduced by 42% contributing to decreased formation of H_2SO_4 and sulphate aerosol. The resulting mean sulphate aerosol concentration over the Mediterranean Sea was reduced by 46% in the lowest model level. The reduction of SO_x emissions did not result in a linear reduction of sulphate aerosol load, because of nonlinear chemical reactions. Ship emissions are released only in the lowest model level and their contribution to sulphate concentration dominated in the lowest 300 m. In conclusion, ship emissions of NO_x contribute to the formation of secondary aerosols hence considerably decreasing Mediterranean air quality in summer. Based on their results, the most significant issue was the formation of nitric acid, which was reduced by 66% without ship emissions according to the model.

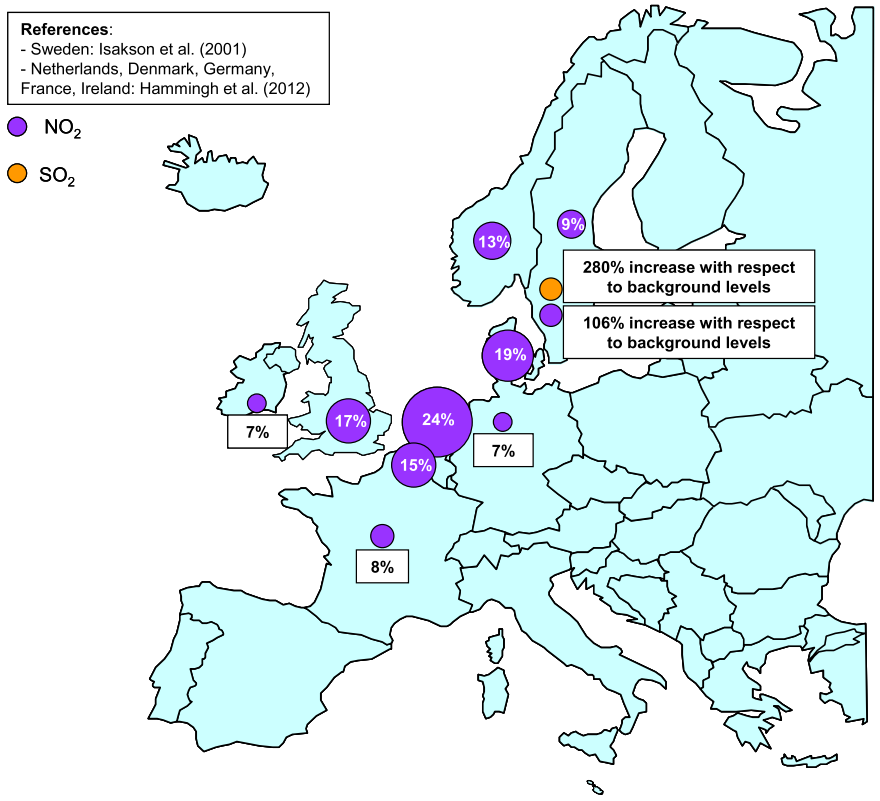

Fig. 2. Contribution from shipping emissions to air quality (NO_2 and SO_2) across Europe. In the case of Sweden, shipping contributions were not reported as absolute concentrations, only as relative increases with respect to background levels.

The results of the literature review evidence that the data available on gaseous pollutants is scarcer than for PM_x (Fig. 2 and Table 2). Contributions from shipping to ambient NO_2 levels range between 7 and 24%, with the highest values being recorded in the Netherlands and Denmark. As stated above, hourly increases in SO_2 concentrations due to shipping emissions were reported in Sweden, but not in absolute terms, and in Spain. The chemical processes leading to reductions in ozone and increases in nitric acid formation in the Mediterranean basin are described, but further quantification of direct impacts on air quality should be pursued.

Table 2
Summary of shipping contributions to air quality across Europe, focussing gaseous pollutants (SO_2 and NO_2). Contributions are expressed as % to the annual mean. In the case of Sweden, shipping contributions were nor reported as absolute concentrations, only as relative increases with respect to background levels.

Gaseous pollutants			
Reference	Shipping contribution	Species	Location
Isakson et al. (2001)	106%[a]	NO_2	Gothenburg (Sweden)
	281%[a]	SO_2	Gothenburg (Sweden)
Keuken et al. (2005)	5–7 ppb	NO_2	Rotterdam (The Netherlands)
Hammingh et al. (2012)	7–24%	NO_2	North Sea coastal countries
	24%	NO_2	The Netherlands
	19%	NO_2	Denmark
	17%	NO_2	UK
	15%	NO_2	Belgium
	13%	NO_2	Norway
	9%	NO_2	Sweden
	8%	NO_2	France
	7%	NO_2	Germany
	7%	NO_2	Ireland

[a] Increase with respect to background concentrations.

2.6. Modelling contribution to population exposure

Only two European studies were found which dealt with the quantification of shipping contributions to population exposure (Andersson et al., 2009; Hammingh et al., 2012), as opposed to contributions to air quality. Other studies have dealt with this issue globally (Corbett et al., 2007). Seven year average concentrations from different European regions and including international shipping as an emission source were analysed using an Eulerian three-dimensional chemistry transport model (Andersson et al., 2009). Results evidenced that the population weighted exposure to airborne pollutants derived from shipping emissions was lower compared to the average concentration contributions, since the emissions occur in less populated areas. The relative contribution to population weighted concentration (PWC) of sea traffic emissions was larger for secondary inorganic aerosols (80% of total PWC) than for primary $PM_{2.5}$. However, differences were found between countries, e.g. for Iceland and the Netherlands the primary shipping contribution to population exposure was more important. On average across Europe, shipping emissions contributed with 8% of population exposure to primary $PM_{2.5}$, 16.5% of population exposure to NO_x, and 11% of population exposure to SO_x. The contribution from shipping emissions to population exposure to CO, NMVOC and NH_3 was lower than 1%. These values are higher than the observational estimates given that they refer to population weighted exposure, as opposed to impact on air pollutant concentrations.

The impacts of shipping emissions on population weighted exposure to $PM_{2.5}$ (for EU27 + CH + NO), and the impacts on the years of life lost (YOLL) were also assessed (Hammingh et al., 2012). The total YOLL in 2030, by international shipping from the North Sea only, may be calculated as 24×10^3 kYOLLS, which is 2.4% of all

YOLLs in the EU in 2030. The contribution of North Sea ships to the YOLLs in only the North Sea coastal countries was somewhat higher (4.4%).

2.7. Modelling of atmospheric deposition

Atmospheric deposition was targeted by a number of studies in this review. A statistical Lagrangian atmospheric transport model (FRAME) was used to generate annual maps of deposition of sulphur and oxidised and reduced nitrogen for the UK for the year 2002 (Dore et al., 2007). A future emissions scenario for the year 2020 was used, and results showed that, if shipping emissions were assumed to increase at a rate of 2.5% per year, their relative contribution to sulphur deposition was expected to increase from 9% to 28% between 2002 and 2020. Enforcement of the MARPOL convention to reduce the sulphur content in marine fuel to 1% was estimated to result in a 6% reduction in total sulphur deposition to the UK for the year 2020.

Nitrogen oxide emissions from North Sea shipping were estimated to be responsible for 2–5% of nitrogen dioxide deposition in North Sea coastal countries in 2030. Compared with the contribution made by North Sea shipping to nitrogen dioxide concentrations, the contribution to nitrogen deposition is relatively lower due the relatively higher contributions from various land-based sources (Hammingh et al., 2012).

Finally, model calculations (OSPAR, 2009) largely suggest that atmospheric deposition of selected heavy metals and organic contaminants (lindane and polychlorinated biphenyls) and of nitrogen substantially declined in the period 1990–2006 in the OSPAR maritime area (www.ospar.org). Model calculations suggested that atmospheric deposition of nitrogen decreased in the Greater North Sea over the period 1995–2006 but stagnated or increased in the other OSPAR Regions.

In addition to these studies, numerous works may be found in the literature dealing with shipping emissions estimations, based on dispersion and chemical transport models and on emission inventories, as well as onboard emissions (EEA, 2013). They conclude that oceangoing ships emitted around 600–900 Tg CO_2 in 2000 (Eyring et al., 2010). In general, most of these works focus on climate impacts of shipping emissions (Agrawal et al., 2008a,b; Petzold et al., 2008; Lack et al., 2009; Moldanová et al., 2009), whereas studies dealing with impacts on air quality are scarce. The emissions studies provide emission factors for various gases (CO, NO_x, SO_2, CO_2), chemical compounds in particulates (S, metals), and aerosol mass emitted from marine engines fed with heavy fuel oil (Becagli et al., 2012). A review of works on shipping emissions, their estimation and climatic impact may be found elsewhere (EEA, 2013), and are not addressed in the current review.

2.8. Assessing and testing the efficiency of mitigation strategies

The United Nations International Maritime Organisation (IMO) emission limits adopted in 2010 apply to the sulphur content of fuels and nitrogen oxides. Particulate matter emissions are mostly co-reduced through the use of cleaner fuels with less sulphur. The European Union focuses in addition on extra requirements for fuel quality and engine usage as vessels approach land (12-mile zone only). The EU Directive 2005/33/EC on sulphur emissions from ships incorporates the IMO sulphur regulation, but additionally requires that all ships in ports use fuels with a sulphur content of less than 0.1% by weight from January 2010 onwards. There are also national regulations or initiatives that aim to reduce air pollution from ships, such as specific charges for operators of land and sea-based activities for NO_x emissions (Norway), the introduction of shore side electricity (Sweden, Netherlands, Germany) or the development of an Environmental Ship Index (ESI) to identify and reward seagoing ships that perform better in reducing air emissions.

Globally, mitigation strategies for the maritime sector have been proposed and tested (Wismann and Oxbol, 2005; Corbett et al., 2009; Campling et al., 2010a; Lack et al., 2011; Lai et al., 2011). The efficiency, cost-effectiveness and cost-and-benefits of air pollution mitigation measures in shipping and ports (e.g., low sulphur fuels, sulphur scrubbers, NO_x mitigation measures, use of liquid natural gas, slow steaming, soot particle filters) have been assessed in various types of studies (Wang and Corbett, 2007; Corbett et al., 2009).

The direct impact of mitigation strategies on ambient PM_x concentrations has been addressed by ex-post monitoring studies in Dutch (Velders et al., 2011) and Mediterranean harbours (Schembari et al., 2012). In Rotterdam (The Netherlands), SO_2 concentration levels were mostly constant between 2000 and 2006 and decreased rapidly between 2007 and 2010. According to the authors, this results from the changes in emissions from refineries, sea shipping in the North Sea, nearby inland shipping and within port emissions, and SO_2 emissions from abroad. The SO_2 emissions from sea shipping decreased after 2006 as a result of the use of fuel with a lower sulphur content due to the SECA (sulphur emission control area) regulations in the North Sea.

The concentrations of SO_2 in Mediterranean harbours (Schembari et al., 2012) were found to decrease significantly (at the 5% significance level) from 2009 to 2010 in three out of the four EU harbours, the average decrease of the daily mean concentrations in the different harbours being 66%. No decrease was observed in the non-EU harbour of Tunis. Neither NOx nor BC concentrations showed significant changes in any of the harbours.

Mitigation strategies for harbour operations have also been tested, although data were only found for the US (Port of Oakland) (Dallmann et al., 2011). A diesel particle filter retrofit and truck replacement program was implemented. A comparison of emissions measured before and after the implementation of the program showed a $54 \pm 11\%$ reduction in the fleet-average BC emission factor. A significant reduction in the fleet-average NO_x emission factor ($41 \pm 5\%$) was also observed, most likely due to the replacement of older trucks with new ones.

There are also quite a number of ex-ante evaluations for specific air pollution mitigation measures in shipping in the European region. One study (Cofala et al., 2007) concluded that shipping measures like low sulphur fuels (0.5% sulphur content) and NO_x reductions (50% reduction in new built) can substantially contribute to a more cost-effective air quality policy in the European region. Another (Bosch et al., 2009) found that the new IMO regulations by 2008 offers an overall estimated €15 to €34 billion in benefits to the EU in improved health and reduced mortality by the year 2020. The inclusion of the European maritime sector into a newly developed land-based Emission Trading Scheme (ETS) for NO_x and SO_2 has also been subject for a study (Campling et al., 2010b). Finally, the impact of potential extensions of emission control areas around the EU on shipping emissions were recently studied (Campling et al., 2012), including the costs and impacts from slow steaming options and soot particle filters.

3. Conclusions and knowledge gaps

A literature review was carried out with the aim to characterise and quantify the contribution from the maritime transport sector to air quality degradation along European coastal areas. Results evidenced that the number of studies assessing the impact of shipping emissions on air quality is not large, although it has been increasing in the past years. All of the studies reviewed agree on

the relevance of this emission source (both stack emissions and related harbour operations), whose impact on air quality was estimated in a qualitative or quantitative manner for PM_x, NO_x, SO_2, PAHs and particle number (N). Because of the fine grain size distribution of these emissions, it was suggested by certain authors that other metrics such as ultrafine particle number concentration or particle toxicity might be better tracers of this emission source. Specific tracers are available for shipping emissions, and the most commonly used marker is the ratio V/Ni, which ranges mostly between 3 and 4 in PM_{10} and $PM_{2.5}$, at different study regions and probably depending on fuel type and composition. Shipping contributions to ambient PM_x in harbour areas are mainly detected in the form of secondary particles (only few studies are available, concluding that shipping contributions are 60–70% secondary particles and 40–30% primary in PM_{10} and $PM_{2.5}$ mass concentrations). However, exposure studies suggest that it might be more efficient from a health perspective to reduce primary emissions from ships (e.g., EC, metals) than precursors of secondary components (e.g., SO_2), which are the target of current mitigation strategies. Quantitatively, the contributions from shipping emissions to PM_x and gaseous pollutant concentrations show a large spatial variability, with maximal contributions in the Mediterranean basin and the North Sea: on average, shipping emissions contribute with 1–7% to annual mean PM_{10} levels, with 1–20% to $PM_{2.5}$, and with 8–11% to PM_1, and with 7–24% to NO_2 concentrations. Consequently, the emissions from the maritime transport sector cannot be considered a negligible source of atmospheric pollutants in European coastal areas. Current mitigation strategies have proved their efficiency, with decreases in SO_2 levels ranging between 50 and 66% (subsequent decreases in secondary PM are not fully quantified). Therefore, the results from this review encourage the continuation of existing measures, as well as the implementation of new ones with a special focus on primary particle emissions from ships.

A number of knowledge gaps were identified in the course of this work:

- Impact on PM_x of harbour operations: only few works were found in the literature dealing with this topic, but they suggest that the contribution to air quality degradation of harbour operations may exceed that of stack emissions.
- Mitigation strategies for harbour operations: the efficiency of strategies such as retrofitting of loading and unloading trucks has only been tested in a few studies in the US.
- Inland shipping: even though the EU has a waterway network of more than 35,000 km covering large to small rivers and canals (INE; Inland Navigation Europe, http://www.inlandnavigation.org/en/factsandfigures.html) and shipping and transport on inland waterways accounted for 465.3 million tonnes of cargo in Europe in 2005 (De La Fuente Layous, L. A.; Eurostat, Ed.; Office for Official Publications of the European Communities, 2006), data on emissions and their impact on air quality is scarce. Research on this topic would be highly relevant to the scientific and policy-maker communities.
- Homogenisation of monitoring strategies: studies carried out in the North Sea focus traditionally on shipping emissions of gaseous pollutants (NO_2, SO_2), whereas those performed in the Mediterranean Sea focus more preferentially on PM_x levels and chemical composition. The establishment of potential collaborations between research teams from the different European regions would favour obtaining data on gaseous and particulate pollutants emitted by ships, which would be comparable across European coastal areas and which would allow for a more integral assessment of the impact on air quality of air pollution from the maritime transport sector.
- Separation of sources with common emission tracers: building on the availability of chemical tracers, further work should be carried out to introduce such tracers in receptor modelling tools in order to achieve a more precise discrimination of fuel-oil derived emission sources.
- Particle size distribution: further work should be carried out to characterise the particle size distribution of particles (primary and secondary) derived from ship emissions, as these reach coastal areas.

Acknowledgements

This work was funded by the Spanish Ministry of Agriculture, Food and the Environment (UCA2009020083), and by national projects IMPACT (CGL2011-26574) and VAMOS (CLG2010-19464-CLI). The literature review was carried out in the framework of the European Topic Centre for Air Quality and Climate Change Mitigation (ETC/ACM) consortium and for the European Environmental Agency (EEA). Support is acknowledged to Generalitat de Catalunya 2009 SGR8 and to project GRACCIE. Abstract art image kindly provided by www.sxc.hu (Image ID: 1388506). The EEA was involved in the study design, and approved the submission of the article for publication. The rest of the funding sources were involved in covering personnel costs.

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