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Particulate matter in marine diesel engines exhausts: Emissions and control strategies

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ABSTRACT

Marine diesel engines emit particles that have a complex nature, being composed by carbonaceous particles, with size spanning from few nanometres to less than one micron, and inorganic particles of micron size mainly made by ashes and sulphates.

On a global scale, international shipping is responsible for few percentages of the particulate matter emissions, which also affect climate, but the regional distribution of naval traffic suggests the insurgence of significant exposure risk for population living along the coastal areas, due to chronic exposure effects. Specific strategies should be implemented to reduce the emissions of all the components of particulate matter. This paper aims to present a survey on the current and innovative strategies to remove particles from marine diesel engine exhausts, along with a critical review of the most recent findings on ships emitted particles. Evidences on physical–chemical properties, toxicology and emission factors of the particles were reported. This survey indicates that several strategies can provide a significant reduction of particulate matter emissions from ships and integration between innovative after-treatment systems, ships design and operation procedures can potentially lead to overall reduction of more than 99% even with parallel fuel savings.

Introduction

Shipping is the most energy efficient and environmental friendly conventional (fossil fuel-fired) transport modality, being the emission rate per ton-mile far lower than that required for aviation, rail and car transport (European Environment Agency, 2014; Natural Resources Defence Council, 2014). For this reason, around 85% of world trading shipments follow maritime routes. Almost 70% of these routes are concentrated within 400 km from the coastline (Corbett et al., 1999; Endresen et al., 2003; Eyring et al., 2010; Eyring et al., 2005b). Shipping emissions influence air quality at long distances from the emitting source (Attica Project, 2009; Eyring et al., 2010; Eyring et al., 2007) and some pollutants have a worldwide dispersion (Bond et al., 2013; Seinfeld and Pandis, 1998) also affecting climate.

The most widely adopted ship propulsion systems are the slow-speed two strokes (60–300 rpm) and medium-speed four strokes (300–1000 rpm) diesel engines fuelled with the relatively inexpensive intermediate (or "heavy") fuel oil (IFO) that, unfortunately, leads to massive emissions of pollutants. Statistical results on the emissions from existing ships based on data of the Lloyd's Register and of the US Coast Guard (EPA, 2000) reported a typical range of concentrations as follows: O_2 10–12% $v/v_{dry\ basis}$; CO_2 3–10% $v/v_{dry\ basis}$; NO 600–1500 ppm $v_{dry\ basis}$; SO_2 80–1000 ppm $v_{dry\ basis}$; CO 50–500 ppm

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$v_{dry\ basis}$; VOC 50–400 $ppm_{wet\ basis}$ (volatile organic compound). The 6th European Framework Programme project QUANTIFY (2010) provided further details on the VOC compounds, which are mainly formed by benzene, toluene, butyl-acetate and xylene (Moldanová et al., 2009). Cooper et al. (1996) reported tentative emission factors for selected hydrocarbons, Polycyclic aromatic hydrocarbon (PAH) and Polychlorobiphenils (PCB), emitted by two passenger ferries under normal operating conditions (four-stroke medium-speed main engines). Ethene, propene, isobutene, benzene and C9–C12 alkanes dominated the hydrocarbon compositions, although their relative proportions differed considerably between the two ferries. The PCB emissions were negligible while the PAH accounted for around 1% of the total VOC.

After recognizing the severity of health effects related to PM, NO_x and SO_2 exposure and the relevance of shipping on the worldwide and regional atmospheric pollution, specific guidelines were introduced in the Regulations 13 and 14 of the Annex VI of the International Convention for the Prevention of Pollution from Ships (MARPOL) of the International Maritime Organization (IMO) that entered in force on the 19th of May 2005. According to the MARPOL VI Article 14, SO_2 and sulphates particles emissions should be reduced either by using proper scrubbers as after-treatment system or by lowering the sulphur content in the fuels. In specific Environmental Control Areas (ECA), the admitted sulphur weight content in the fuel had to be lower than 1% from the 1st of January 2010 to the 31st of December 2014. Nowadays the maximum admitted sulphur content is 0.1% that will be extended to all ships in the world by 2020. The former limit allowed the use of low sulphur fuels as Marine Diesel (Gas) Oil, known by the acronyms MDO or MGO. Ultra low sulphur fuels (ULSF) are required to comply with current limits. Unfortunately, the unit costs of these fuels is far higher than that of conventional IFO and cost benefit analyses are now driving part of the Maritime Sector towards the adoption of after-treatment systems to comply with regulations. Cullinane and Bergqvist (2014) recently highlighted that the introduction of ECA regions did not produce a modal shift towards other transport means. The same authors also envisaged that the large socio-economic benefits and the global challenges of containing pollution in densely populated areas, such as the Mediterranean and Asia, emphasize the importance of designating more regions as ECAs.

Diesel particulate matter (PM) emitted by ship engines is a mixture of different kinds of particles with size spanning from few nanometres to several microns. Among them, the soot particles are related to severe pathologies and classified as carcinogenic of Class I by the World Health Organization (International Agency for Research on Cancer, 2012). Marine diesel engines also contribute to the emission of black carbon, which is recognized as an important climate-forcing agent.

Ships are responsible for a small fraction of the worldwide particles emissions, but the regional distribution must be carefully considered (Eyring et al., 2007). For example, biomass combustion is dominant and diffused in the South hemisphere, while around 70% of the emissions in Europe, North America and Asia derives from industry and transportation (Bond et al., 2013). The impact of shipping on environmental pollution levels is the highest along the west and east coasts of the United States, North Europe, North Pacific and Mediterranean Sea and close to Indian coasts (Eyring et al., 2005a,b; Eyring et al., 2007). The impact on population is still largely unrecognized but widely diffused. In fact, census data show that in the USA and in the European Union about 53% (Crosset, 2004) and 40% (Collet and Engelbert, 2013) of the resident population lives in coastal areas. In South America and Asia (with the exception of India) from 60% to 75% of the population lives within 400 km from the sea (Hinrichsen, 1998). On a global scale, 23% of the world population density live within 100 km from the shoreline (Nicholls and Small, 2002), and 23 up to 28 of the largest megalopolis, with more than 10 million inhabitants are in coastal areas.

In spite of these findings, to date there are no specific regulations pertaining to the emission of particulate matter, apart from the side effect related to the use of low sulphur fuel in the Marpol VI. Nevertheless, in the last years, the IMO started a panel to investigate the black carbon emission, measurements and possible mitigation strategies with reference to its climatic effects.

The evaluation of exposure risk associated with ships' engine emitted particles and the assessment of strategies to control particulate emissions are important research topics echoing a concrete societal need. The mitigation strategies must take into account the different constituents of particulate matter. The coarse particles in the ship's engine exhausts are related to the presence of sulphur and ashes in the fuel and can be effectively removed by using distillate fuels or by adopting scrubbers, as indicated by the MARPOL Annex VI. Finer particles with submicron size are related to both fuel properties and combustion processes. Strategies to reduce their emissions include optimization of ship and engine designs, use of cleaner fuels and adoption of proper exhaust gas cleaning systems. The implementation of ship operation practices to reduce energy consumption is also of interest.

Unfortunately, the absence of specific regulations does not allow to collect significant amount of quantitative data on the effective removal of particulate matter associated with these different strategies. However, reliable indications on the reduction of particles emissions can be derived from experimental studies at laboratory and pilot scale, from the experience on the removal of PM associated pollutants (e.g., CO_2, SO_2) or from data on composition and usage of fuels.

This paper presents a survey on the state of the art of the current knowledge on the physical–chemical properties, the toxicology and the emissions of particles from ships, together with an analysis of the mitigation strategies commercially available, as well as of those concepts and emerging technologies under development and proved either at pilot or laboratory-scales.

For the sake of simplicity, this paper is divided into two main parts. The first one provides an overview of the current knowledge on particulate matter emissions and characteristics. To simplify this description, this part is divided into paragraphs discussing characteristics, toxicology, climatic effects, emissions and regulations. The second part reports a critical review on the consolidated and innovative mitigation strategies to reduce particulate emissions from the exhaust gases.

Particulate matter emitted from marine diesel engines

Characteristics

Particulate matter emitted by IFO fuelled diesel engines is a complex ensemble of particles of different kinds whose dimensions spans from few nanometres to over 20 μm. A comprehensive study on chemical specification of marine diesel exhausts from ocean-going vessels during real operation was reported in the 6th European Framework Project Quantify (2010).

Particulate emissions are composed by three main fractions (Fridell et al., 2008; Lack et al., 2009; Moldanová et al., 2009):

- Mineral ashes, derived from fuel impurities, usually having size between 200 nm and 10 μm.
- Sulphates and (in minor fraction) nitrates together with associated water, usually in the micrometre size range.
- Soot particles, largely in the submicron (<1 μm) and ultrafine (<200 nm) range and commonly classified into two main components: elemental carbon (EC) and organic matter (OM), mostly made up of organic carbon (OC).

A brief summary of definitions and metrics concerning particles is reported in Table 1, which reported, in its columns, some major information on physical–chemical properties, size, metric concepts, techniques to measure particle diameter and definitions of mean diameters.

The International Maritime Organization (2009) and Petzold et al. (2011) indicated that weight fractions of sulphates and sulphur derived particles were by far dominant, accounting for 80 and 78% in weight of the emitted particles, respectively. This percentage was found to increase to 85% including ashes. In the same studies, the EC and OM fractions were between 2% and 3% and 10–13.5% respectively.

Table 1
Summary of particles definitions and metrics in terms of their different properties, size, metric concepts, techniques to measure particle diameter and definitions of mean diameters.

Typology	Sizes		Metrics	Diameter measurement	Mean diameter
Sulphates (and linked water): Inorganic particles derived from sulphur in the fuel	Ultrafine particles < 100 nm (Nanoparticles <50 nm)	Nucleation particles d< 10 nm	Total suspended particles (TSP), mg/Nm^3	Microscopy SEM/TEM (d= 1 nm- 5 μm) Optical m. (d> 1000 nm) Shape factors can be addressed	Mass based: Each fraction is weighted with the third power of particle diameter – enhanced contributions of coarse particles
Ashes: Mineral particles		Aitken mode d<100 nm	PM_x: the mass concentration of particles passing, through a filter with cut diameter x. E.G. PM_{10}, $PM_{2.5}$, PM_1	Laser diffraction methods	Number based More representative of actual particle fractions
Elemental carbon (EC): Carbon particles stable above 350°C	Fine particles < 1000 nm (sometimes referred to < 2500 nm, $PM_{2.5}$)	Accumulation mode d<1000 nm		Dynamic light scattering	
Black carbon (BC): Fraction of the EC able to absorb solar radiation across all visible wavelengths	Coarse mode: d > 1000 nm		Number concentration: #/Nm^3	Sieving classifications: (mesh diameters)	
Organic Matter (OM): Condensed vapor that evaporate above 350°C			Particle Surface area m^2/Nm^3	Aerodynamic diameter	
			Composition specific metrics	Electrical mobility classification diameter	
			Source specific metrics		

The properties of mineral ashes and of sulphates and nitrates particles are largely known in the pertinent literature. References to their features and toxicity are discussed in several textbooks, e.g., Seinfeld and Pandis (1998), and are not discussed thereinafter.

One important point of discussion is that, while sulphate and ash particles are dominant in terms of weight emissions, they become negligible with reference to particles' numerical concentration, which is dominated by soot particles that form the most toxic particulate fraction. Soot particles are by-products of every combustion, gasification or pyrolysis processes and result from undesired reactions pathways (Commodo et al., 2013; Cormier et al., 2006; D'Anna, 2009). The two fractions of diesel soot, elemental carbon (EC) and organic matter (OM) have different characteristics. The EC, is composed by nanometric size spherules made by clusters of stacks formed by 3–4 planes of planar polycyclic aromatic hydrocarbons (PAH) structures, as shown in Commodo et al. (2013) and in D'Anna (2009). These spherules can aggregate to former larger chain-like structure with size in the range of some hundreds of nanometres. A fraction of EC is able to absorb light across the visible wavelength (Bond et al., 2013). This fraction is called Black Carbon (BC) and is sometime coincident with the whole EC emitted.

The simpler measurable difference between of EC and OM is the particles stability at high temperatures: EC is stable also at high temperature, while the organic matter is made by condensed vapours that volatilize above 350 °C. A fraction of the organic matter, called OC is composed by PAHs organic compounds with high molecular weights condensed on metallic nuclei (as Sodium, Calcium, Potassium) and transition metals (Vanadium, Nickel, Iron, Lead, Zinc, Aluminium) that are generated in the post flame region. Although the basic qualitative structure of soot particles is mainly independent from the combustion process, the concentration of heavy metals in the fuel influences the properties of the soot, especially the OM fraction, giving rise to a certain degree of correlation between soot particles properties and parent fuel characteristics, as, for example, reported by Petzold et al. (2011).

Fig. 1 shows a sample number and weight particle size distribution for IFO fuelled diesel engines based on the data of Fridell et al. (2008) along with pictures representing the different fractions of particulate matter. In terms of number concentration, the submicron soot components are by far dominant in the IFO fuelled diesel engine emissions. Differently, in terms of weight fraction, it appears that particulate matter emission has a tri-modal size distribution with two peaks in the submicron range and a third one above several microns. The same tri-modal particle size distribution was observed from several other studies (Petzold et al., 2011; Ushakov et al., 2013b; Winnes and Fridell, 2009). Soot aggregates in the nucleation and accumulation modes dominate the sub-micron fraction, while mineral ash particles and sulphur related aerosol compose the coarse fraction.

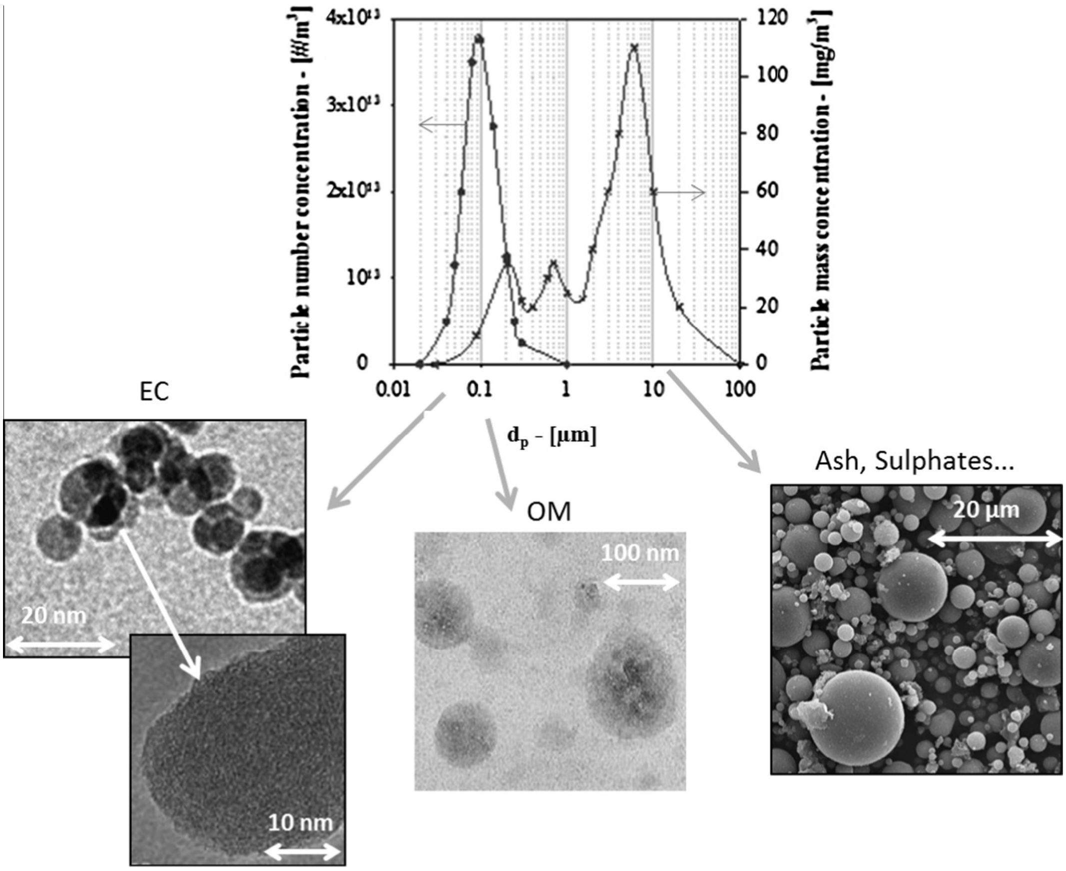

Fig. 1. Mass and number distribution of particles from an IFO fuelled marine diesel engine and TEM/SEM pictures of relevant fractions.

Fig. 1 also shows pictures of the typical morphology of the particles classes present in the diesel engines. It is worth noticing that while the coarse particles mainly show a globular shape, the EC particles are highly irregular while OM particles appear as irregularly shaped condensing droplets. One important consequence of this evidence is that, while the concept of mean aerodynamic diameter is a good approximation for ashes and sulphates, this may not be appropriate for EC particles, whose shape is very different from a sphere. Mobility or optical equivalent diameters are a more appropriate measure of soot particle diameter. Methods for nanoparticles diagnostics are reported in D'Anna (2009).

When atmospheric measurements were carried out, primary emitted particles are accompanied by secondary particles whose amount and characteristics depend on the occurrence of secondary reactions between pollutant in the plume and the air and strongly depend on plume ageing (Viana et al., 2014). Petzold et al. (2008) measured the transformations of aerosols in a plume of a large container ship. The authors found that the particle size distribution of the raw exhaust has a count median diameter of 52 nm (in the nucleation mode) that became about 70 nm in the young plume and 100 nm in 30 min aged plume (in the accumulation mode). The mass of BC and the associated number of non-volatile combustion particles were correlated with the excess CO_2 in the plume and a maximum plume lifetime of about 24 h was estimated for a well-mixed marine boundary level. Further discussion on plume ageing effects is reported in the 'Regional scale impact of ships emissions' section.

Toxicological and epidemiological evidences

Even in low doses, chronic exposure to soot particles was associated with several respiratory pathologies, such as bronchitis, pneumonia, tracheitis, and asthma (Lighty et al., 2000), as well as to the occurrence of disruption of vascular functions (Rundell et al., 2007), ischaemic heart diseases (Slezakova et al., 2013) and neurological effects at some brain regions (Oberdörster et al., 2004). Recently Pedata et al. (2013) showed that the exposition of endothelial cells to flame generated nanoparticles for 24 and 48 h, led to apoptotic cell death.

For these reasons, diesel soot is a highly hazardous material classified by the International Agency for Research on Cancer (IARC) as human mutagen and carcinogenic agent (International Agency for Research on Cancer, 1985, 2012).

The toxicology of diesel particles relies on three main parameters: particles morphology, chemical–physical structure and size (Harrison and Yin, 2000; Lighty et al., 2000). Kumar and co-workers (Kumar et al., 2011; Kumar et al., 2010), Tetley (2007), Oberdörster et al. (2005) and Wang et al. (2005) indicated that soot chemical composition gives rise to high surface reactivity and ability to cross cellular membranes. Particles surface area is a key toxicological parameter since it determines the amount of available functional groups and of toxic gaseous species that can be adsorbed on particulate matter.

Particle size is directly involved in this phenomenon: at the same concentration, the finer the particle, the higher the surface area and the toxicity. Moreover, once inhaled, finer particles penetrate deeper in the lungs and those smaller than 300 nm can cross cellular membranes (Oberdörster et al., 2004) extending their action to other organs (Oberdörster et al., 2005, 2004; Tetley, 2007; Wang et al., 2005). Besides, particles with irregular morphology, as those composing the EC, also have larger surface areas than spherical particles of equivalent diameter and are potentially more harmful. On the overall, these considerations make finer particles potentially more toxic than larger ones.

The characteristics of particles generated by marine diesel engines and of the exhaust gas make them particularly harmful since, in addition to the PAH based structure common to all EC and OM particles, and they can contain vaporized heavy metals that are derived from the parent fuel and the lube oils. The presence of several transition metals, such as Vanadium, Lead and Nickel in IFO fuels, makes the marine diesel particles potentially more harmful than those produced by other sources, such as gasoline or natural gas (Cormier et al., 2006; Harrison and Yin, 2000; Laden et al., 2000; Lighty et al., 2000; Sippula et al., 2014; Zimmermann et al., 2013).

Zimmermann et al. (2013) exposed human alveolar basal epithelial cells to fresh, diluted exhaust gases produced by either IFO or Marine Diesel Oil (MDO) fuelled ship engines. The authors found that the chemical profile of particles generated with IFO and MDO was very different: although the EC content of MDO exhaust was higher, the IFO particles resulted considerably more toxic. This higher toxicity was correlated with the larger aromatic structures and transition metal content in the IFO fuelled engine particles. The same finding was reported by Sippula et al. (2014) who showed that IFO fuels produced larger amounts of hazardous species (PAHs, Oxy-PAHs, NPAHs, transition metals) but less amount of EC compared with diesel fuels. However, since the toxicity of EC particles is not well established yet, the authors concluded that it is difficult to rank the toxicity of IFO and diesel fuels at this stage. This was further confirmed by the same research group (Oeder et al., 2015), which recently showed that human lung cell exposed to IFO particles generated higher responses in terms of pro-inflammatory signal, oxidative stress and xenobiotic metabolism, but diesel fuel particles generated broader biological response that may lead to higher risk of disturbance of normal cell functions.

The role of additives (Gualtieri et al., 2014) and of lubricant oils (Sippula et al., 2014; Zhang and Balasubramanian, 2014) must be accounted for to properly address particle toxicity. In particular, Gualtieri et al. (2014), found that particles produced by diesel fuels doped with 10 wt.% of additives of commercial interest (i.e., mono-ring aromatics, multi-ring aromatics, naphthenic compounds and oxygenated components as fatty acid methyl esters) have toxic potential enhanced by 10 times. The addition of fatty acid methyl esters provided the highest cytotoxicity.

Zhang and Balasubramanian (2014) compared the characteristics and the toxicology of particles emitted by non-road engine fuelled with biodiesel–diesel and butanol–diesel blends. They did not highlight significant differences in the cytotoxicity, measured in terms of cell viability, of the particles emitted by the different blends. However, through correlation between the cytotoxicity and OM content in the engine exhausts, they argued that the role of lubricating oils in generating OM particles should be properly addressed to understand the actual toxicological effects showed by the diesel blends.

At the best of our knowledge, large-scale epidemiological studies on ships emitted particles are not available to date. Similarly, we have not found any reference to epidemiological studies on diesel soot exposure risk so far. Differently, thanks to the large grid of data concerning air quality standards in USA and EU, there are several studies concerning the exposure risks related to fine particles in ambient air. These studies correlated exposure risk to measurements of the mass concentration of particulate matter finer than 2.5 μm ($PM_{2.5}$). It was indicated that about 800.000 deaths/year are associated with $PM_{2.5}$ exposure (Corbett et al., 2007) and several toxicological studies in Netherlands (Brunekreef et al., 2009) and United States of America (Krewski et al., 2009; Pope III et al., 2002) highlighted a clear correlation between chronic exposure to $PM_{2.5}$ particles and mortality risk. The highest values of the relative risk of mortality are related to ischaemic heart diseases and lung cancer, with stronger correlations for nonsmokers. Considerations on particles size and toxicity lead to presume that stronger correlations may be found between soot particle exposure and occurrence of associated pathologies.

With reference to ships emissions, by considering exposure risk as the product between specific risk and exposure frequency, the highest risk is expected to be related to submicron particles and to occur for seafarers, harbour personnel and citizens living along the coastlines of intense naval traffic areas.

Climatic effects

The particles emitted from ships also affect climate. One important effect is that related to cloud formation, since they act as water condensation nuclei (Booth and Bellouin, 2015; Hobbs et al., 2000; Hodnebrog et al., 2014; Seinfeld and Pandis, 1998). This role is actually more pronounced for sulphate particles and is thus stronger for IFO fuels. On the overall, the sulphate particles are well known to be associated with a cooling effect (Eyring et al., 2010). The climate effects related to sulphates and nitrates aerosols are the subject of several studies and are reported in reference textbooks on atmospheric chemistry and physics (Seinfeld and Pandis, 1998).

A fraction of the elemental carbon, known as *black carbon* (BC), is able to absorb solar radiation across all visible wavelengths and freshly emitted BC has a mass absorption efficiency of 5 m^2/g at the mid-visible wavelength of 550 nm (Institute of Marine Engineering, 2011). BC is the second most important climate warming agent after CO_2, having a radiative index of 1.1 W/m^2, although new evidence is recently reconsidering its role in climate forcing (Booth and Bellouin, 2015; Hodnebrog et al., 2014). The intensity of this light absorption varies with the composition, shape, size distribution and mixing state of the particles. The measure of black carbon requires the adoption of new optical methods, mainly available for laboratory scale applications, as those based on Transmittance/Reflectance, Thermal speciation, Photo-acoustics, Refractive index measure, Laser incandescence or Extinction/Scattering (Institute of Marine Engineering, 2011). A recent and comprehensive review on the emissions and the characteristics of Black Carbon was published by more than thirty co-authors coming from different research centres (Bond et al., 2013). To date, this is the most important and authoritative reference in this field. Corbett, Lack and co-workers, reported specific review studies on the Black carbon emissions from ships (Corbett et al., 2010; Lack and Corbett, 2012; Litehouz et al., 2012).

A recent study by Petzold et al. (2011) on an IFO fuelled externally charged 1L32/44 single-cylinder test engine with 400 kW power, showed that black carbon emissions and elemental carbon emissions are almost coincident when the engine load was kept below 75%. For higher load, BC is less than 75% of the corresponding EC concentration.

In recent years a growing attention is on the role of particulate matter, and in particular black carbon, on the Arctic climate arose. This eventually drove the beginning of IMO discussions (BLG 15 & MEPC 62) on the introduction of measures to determine and reduce black carbon emissions from ships (Lack and Corbett, 2012; Litehouz et al., 2012). During the 68th Meeting of the Marine Environmental Pollution Committee of the IMO, in May 2015, the definition of Black carbon provided by (Bond et al., 2013) was officially adopted. No control measure for black carbon was however discussed. In a recent paper, Winther et al. (2014) highlighted that in 2012 the largest fraction (about 45%) of emitted BC in the Arctic region (above 58.95 N latitude) was related to fishing ships with total emissions of 1.585 kT/year.

Ships emissions

Emission factors

Statistical data reported by the EPA on the basis of Lloyd's register and the US Coast Guard show that the emissions of particulate matter depend on engine load and span over a wide range from 0.3 up to 10 g/kW h (EPA, 2000; Winnes and Fridell, 2010). Early tests on particulate matter from ships at berth fuelled with MDO and IFO fuels were reported by Cooper (2003), who measured total particulate matter emissions factors from 0.48 to 0.67 g/kW h from IFO fuelled engines and from 0.14 to 0.48 g/kW h from MDO fuelled engines. More detailed estimation of each component of the particulate was reported recently by Moldanová et al. (2009), Petzold et al. (2008), Agrawal et al. (2008), Moldanova et al. (2013) and by the formerly cited works of the International Maritime Organization (2009) and Petzold et al. (2011).

Experimental data were gathered either from atmospheric measurements or from engine exhaust pipe either on board or on a test rig.

Lack et al. (2009) reported an average particle composition as 46% sulphates, 39% OM, and 15% BC starting from observations gathered during the Texas Air Quality Study/Gulf of Mexico Atmospheric Composition and Climate Study 2006 field campaign. Diestch et al. (2013) investigated the emissions of 139 vessels on the banks of the Elbe river in Germany findings on average, an emission of PM_1 of 2.4 ± 1.8 g/kg$_{fuel}$ of which 0.15 ± 0.17 g/kg$_{fuel}$ made by BC and a total number of emitted particle of $2.55 \pm 1.91 \cdot 10^{16}$ #/kg$_{fuel}$.

Finally, in a very recent paper, Westerlund et al. (2015) reported a comprehensive study on the particle emissions associated with 154 different ships. The emission factors from these ships ranged between 0.14 and $8.63 \cdot 10^{16}$ #/kg$_{fuel,}$ corresponding to about 340–5600 mg/kg$_{fuel.}$ The corresponding values of the non-volatile particle emissions were $0.11–4.11 \cdot 10^{16}$ #/kg$_{fuel}$ and 120–1550 mg/kg$_{fuel}$. The average particle diameter was in the range 20–40 nm.

During tests in a test rig, Petzold et al. (2008) reported data on the emission of particles emitted from a serial MAN B&W four-stroke marine diesel engine operating on heavy fuel oil with 2.21% sulphur and 0.03% ash. The authors showed that EC emissions are 0.038 g/kW h while the total number of emitted particles was $7.27 \pm 2.71 \cdot 10^{15}$ particles per kW h for an engine load spanning from 85% to 110%.

Tests on board ships were carried out by Agrawal et al. (2008), Moldanová et al. (2009), Jonsson et al. (2011) and Moldanová et al. (2013). Agrawal et al. (2008) reported emission factors of the main engine of an ocean going PanaMax class container vessel, at certification cycle and several engine loads, during actual operation at sea. In its experimental campaign the emissions factor for engine load from 27% to 70% were: EC = 0.017 g/kW h; OM = 0.270 g/kW h; Ashes (based on fuel ash content) = 0.123 g/kW h and sulphates = 1.227 g/kW h. The EC and OM emission almost doubled for 8% engine load. Moldanová et al. (2009) found similar emission factors for a cargo ship operated in the Celtic Sea, the English Channel and the North Sea. In particular, they reported emission factors of: EC = 0.02 g/kW h; OM = 0.30 g/kW h; Ashes = 0.19 g/kW h and sulphates = 0.15 g/kW h. Jonsson et al. (2011) estimated the particle emission factors from six different ships (passenger ships, cargo ship and tugs) in terms of numerical emission of total and non-volatile particles. On average, the total emitted particles were between 0.8 and $4 \cdot 10^{16}$ particles per kg of burned fuel while the non-volatile particles were between 0.3 and $2.6 \cdot 10^{16}$ particles per kg of burned fuel. Moldanová et al. (2013) reported the emission factors of a cargo ship travelling in the Baltic Sea in 2010 and on a cargo/passenger ferry on line traffic between Sweden and Germany in March–April 2010. The measures referred to four-strokes diesel engines fuelled with different kinds of MDO and IFO with a sulphur content from 0.1% to 1%. Experimental results indicated a correlation among sulphur content and particulate emissions. Increasing the sulphur content from 0.1% to 1% led to an increase of PM_{10} from 0.34 to 2 g/kW h. Similar reductions were shown for $PM_{2.5}$ and PM_1 emissions, which varied in the range 0.23–1.41 g/kW h and 0.27–1.9 g/kW h, respectively.

Contribution to worldwide particulate emissions

Different papers and statistical data can be used to provide an evaluation of the global and regional relevance of ship's emissions of particulate matter. A first indication of ship's emission compared with other industrial applications derives from an assessment of worldwide crude oil consumption. A synopsis of the energy consumption as derived from the data of International Energy Agency (2011), the United Nations Conference on Trade and Development (2011, 2012) and from the World Energy Council (2011) is shown in Fig. 2. Maritime transport results responsible of about 5.4% of crude oil consumption and of about 2.3% of the total fossil fuel combustion. Including the contribution of biomass for domestic heating and cooking, this percentage lowers to about 1% (Bond et al., 2013). However, perspectives on trade market indicate that the amount of goods transport by shipping may triplicate within 2050, leading to a corresponding increase of fuel consumption. This still represents a minor fraction of fossil fuel consumption but a high increase of crude oil demand and of emissions in high traffic density areas.

Assessment of the global emissions of soot may derive from recent calculation on black carbon (BC) inventories. In the comprehensive review of Bond et al. (2013), the worldwide emission inventory of BC was estimated to span within 4 and 29 MT/year, with an average value of 7 MT/year. Marine diesel engines were considered responsible for about 0.07 MT/year of black carbon (the value spans between 0.04 and 0.29 MT/year) corresponding with 1% of total emissions. Eyring et al. (2010) formerly estimated an almost double mean value of 0.16 MT/year, with lower and upper bounds of estimations at 0.06 and 0.24 MT/year, respectively. This last estimate weights global ships emissions of BC at about 2.25%.

World Energy Demand (2010), EJ Total = 532.8 EJ		
Nuclear	26.40	(5%)
Crude oil	184.6	(35%)
Renowables	52.75	(10%)
Natural Gas	121.3	(23%)
Coal	147.7	(28%)

Crude oil Demand (2010), MBPD Total = 87.4 MBPD		
Electric power	4.37	(5%)
Building	8.74	(10%)
Industrial	26.95	(25%)
Transportation	47.34	(54%)

Transportation demand (2010), MBPD		
Low duty vehicles (LDV)	24.62	(52%)
Trucks	8.05	(17%)
Buses	1.89	(4%)
Marine	4.73	(10%)
Aviation	4.73	(10%)
Rail	1.42	(3%)
Other	1.42	(3%)

Fig. 2. Synopsis of average values of energy consumption and crude oil use by category. Data from International Energy Agency (2011), United Nations Conference on Trade And Development (2011, 2012), World Energy Council (2011). MBPD is the acronym of Million Barrels Per Day.

Assuming that BC is between 75% and 100% of the EC emissions (Petzold et al., 2011) and using the emission factors data as a source of average particulate matter composition, it also may be estimated that total particulate matter emissions from ships should weight for about 1.4–1.8 MT/year. Eyring et al. (2010) confirmed a value close to 1.8 MT/year. Neef (2009) estimated, on historical bases, a total PM emissions in 2009 of about 1.6 MT/year, with a linear increasing trend that was expected to reach 2 MT/year in 2014. In the same year, Lack et al. (2009) estimated ship emissions as 0.9 MT/year being consistent with the historical data of Eyring et al. (2005b).

Regional scale impact of ships emissions

Although the contribution of particulate matter (and BC) from maritime sector appears as a low percentage of the worldwide emissions, their regional distribution showed that the largest portion of ships emissions is located along coastal shipping routes within 400 km from coastlines (Eyring et al., 2010; Eyring et al., 2007) with high population density levels. Eyring et al. (2010) estimated that ships emissions contribute up to 5–20% of the non-sea-salt sulphate concentrations in coastal regions over land of the Northern Hemisphere. Lauer et al. (2007) found that ship emissions contribute to 10–40% to sulphate concentrations over most of the Northern Hemisphere oceans. Modelling of atmospheric dispersion pathways, in the 6th European Framework Specific Support Action Project ATTICA (2009), indicated that SO_2 pollution related to ship emissions affects air quality in more than 70% of the European territory: a similar result is expected for the finer fractions of emitted PM.

To better understand the risk associated with particles emitted by ships, it is worth presenting data on the residence time, the distances travelled by particles in ship plumes and the data on the emission factors of ships during manoeuvring and in idle. However, the first relevant issue to face when trying to assess PM emissions from ships is the individuation of a robust and reliable tracer to mark this particulate from other emitted sources. Agrawal et al. (2009) indicated the concentration of Vanadium and Nickel as robust markers for heavy fuel oil burning on board ships. In particular, ships emissions of PM_{10} and $PM_{2.5}$ can be referred to a V/Ni mass ratio of about 3–4, mainly due to the fuel properties. In the same period, Healy et al. (2009) performed tests at Tivoli Docks in the Port of Cork (Ireland) finding that ships emissions are accompanied by specific temporal trends of OC/EC/Na/Ca/V/Ni/Fe/sulphate concentration which are associated peaks in ultrafine particle number up to two orders of magnitude than background particulate emission. Similarly, Sippula et al. (2014) recently indicated that Ca, P, and Zn were suitable markers for the lubricant oils and their emissions increased with engine load. These markers can be used to assess the impact of shipping at regional scale. Viana et al. (2014) confirmed the use of V/Ni ratio as a marker for PM_{10} and $PM_{2.5}$ and explained how shipping contributions to ambient aerosols in harbour areas can be mainly detected in the form of secondary particles: a few studies available for primary and secondary particle contributions indicated that shipping PM_{10} and $PM_{2.5}$ emissions are in general made by 60–70% secondary particles and 40–30% primary particles. The V/Ni ratio was then recognized as a robust IFO fuels engine emissions tracer.

As regards particle residence time, Balkanski et al. (2010) indicated that BC and OM residence time in the atmosphere is around 7.3 and 7.6 days respectively, while that of sulphate is 4.6 days. Similarly, in a recent paper, Beecken et al. (2014) measured concentrations and particle size distribution (in the range 15–560 nm) particulate matter in ship plumes, at distance up to 8 km from the source of emission, analysing a set of 158 ships travelling on the Baltic and North Sea during 2011 and 2012. The authors found that the average emission factor of particles was of $1.8 \pm 1.3 \cdot 10^{16}$ #/kg_{fuel} and that particle size distribution was mono-modal with size between 45 and 54 nm. Besides, the author found that particle concentration reduced only for about 2/3 by measuring concentration at distance within 0.5 km or within 5–8 km from the source. Finally, in a recent paper, Anderson et al. (2015) clearly indicated that the particles emission factors may increase by one order of magnitude by reducing engine load from 35% to idle. These results indicated that the ship plumes influence the quality of air at significant distance from the source and for a long time.

In intense naval traffic areas, the role of ships' emissions to air quality is relevant and, for their location, usually in or close to city centres, harbours represent a massive source of exposure to harmful concentrated pollutants for the citizens. In this sense, a number of studies on the air quality close to harbours confirmed the significant contribution of shipping on the actual PM concentration in neighbouring areas, even in the presence of intense urban traffic. This topic was recently reviewed in the pertinent literature (Matthias et al., 2010; Mueller et al., 2011) and discussed thereinafter.

Using V/Ni ratio as a tracer, Agrawal et al. (2009) indicated that ships emissions are responsible for about 8.8% $PM_{2.5}$ atmospheric concentration close to Los Angeles harbour and 1.4% of $PM_{2.5}$ at 80 km inland. Lack of specific data to correlate PM_1 particle air concentrations did not allow evaluating the effect of shipping on the EC and OC concentration.

Minguillón et al. (2008) described the results of an experimental campaign carried out in 2007 in the area of Los Angeles harbour. This is the busiest harbour in the US and the fifth in the world, but the area is also affected by the emission sources associated with harbour activities (marine vessels, heavy-duty trucks, locomotives, cargo handling equipment and harbour crafts), intense road traffic from nearby freeways and local streets, multiple petroleum refineries and other industrial facilities. The authors found that the high vehicular traffic dominated the emission profile in the area of Los Angeles, being ship emissions responsible for about 5% of the total emissions of PM_1 and "quasi-ultrafine" particles. At larger distance from the Los Angeles harbour areas (Pasadena – at about 50 km distance – and Riverside, at about 70 km distance), the concentration of organic aerosols was only related to vehicular traffic (Hayes et al., 2013). Lu et al. (2006) analysed ship plumes over the Vancouver (Canada) area, showing that ship emissions could generate a significant contribution to population exposure risk. The detailed measurements collected during this study showed that ship emissions led to an order of magnitude increase in SO_2, NO and VOCs (particularly methylated compounds) above background. Particulate matter mass and count levels were 3–5 times higher in the ship plume compared to the urban background. The largest increase was shown for particles with diameters in the range 100–200 nm while fine particle mass ($PM_{2.5}$) increased threefold above the urban background. This last effect was related to nucleation and accumulation (Aitken) modes organics, but also to sulphate. Compared to the urban background distribution, the ship plume particles had smaller sizes with the main mode in the EC/OM distribution at 70 nm and the sulphate mode at 150 nm. While ultrafine organic particles also dominate the emissions from other fossil fuel combustion sources, small sulphate particles were derived uniquely from ships exhausts. Measurements of air quality over time based on a continuous night monitoring of a ship plume showed that dispersion and deposition processes caused about 20% decrease in concentrations over 3 h but an 1–1.3% h^{-1} SO_2 growth rate in particle phase sulphur, due to the transfer of gas phase SO_2 to hydrated aerosol surfaces followed by heterogeneous chemical reactions was observed. At the same time, they observed a net loss of volatile or semi-volatile organic compounds from the particle to the gas phase.

Poplawsky et al. (2011) investigated air quality levels of $PM_{2.5}$, NO_2 and SO_2 associated with cruise ships in James Bay, Victoria, British Columbia (Canada). Data were obtained over 4 years (2005–2008) by the public air quality network and during a specific campaign carried out from the 30th of May to the 24th of August 2009. While background emissions of NO_2 and $PM_{2.5}$ resulted quite high, so that ship contribution could not be properly assessed, the monitoring data showed 3 up to 5 folds increases in hourly SO_2 when ships were in port during all years.

The ENTEC (2007) reported that ships manoeuvring and hoteling in the Mediterranean sea contributed to about 14 kT/year. In 2014 Viana et al. (2014) reviewed data on the effects of ships emissions along European coastlines showing that shipping influenced atmospheric aerosol concentration within about 8% of PM_{10}, while data on $PM_{2.5}$ indicated larger effects up to 20% (Mazzei et al., 2008). A few data on PM_1 indicated that shipping contributed for a fraction between 8% and 11% in Barcelona (Amato et al., 2009) and Lampedusa (Becagli et al., 2012) with peaks up to 60% in Genova (Mazzei et al., 2008) where a correlation between PM_1 concentration and the traffic of passenger ships in the harbour during the holiday period, when peaks of about 5 $\mu g/m^3$ were observed. Maragkogianni and Papaefthimiou (2015) indicated that emissions of particulate matter from cruise ships in the Greek ports of Piraeus, Santorini, Mykonos, Corfu and Katakolo, for year 2013 amounted to 94.3 T and are related for 88.5% to the ships hoteling and 11.5% to their manoeuvering.

Ships emissions are relevant also in the northern sea, in higher industrial areas. Ships hoteling in the port of Bergen (Norway) generated 8.7 T of PM_{10} (8.67 T of $PM_{2.5}$) in 2010 (McArthur and Osland, 2013) while values of PM emissions up to 122 T/year 248 T/year were respectively reached in Kaohsiung in 2010 and Rotterdam in 2005 (Berechman and Tseng, 2012; Hulskotte and Denier van der Gon, 2010). Gibbs et al. (2014), on the bases of data for UK ports and statistical model for emission inventories, indicated that emissions generated by ships during their voyages between ports are of a far greater magnitude than those generated by the port activities.

Consequently, "the emissions from the maritime transport sector cannot be considered a negligible source of atmospheric pollutants in European coastal areas" (Viana et al., 2014).

As regards Asia, recent works were published by Fu et al. (2013), Zhao et al. (2013) and Kim et al. (2009). Kim et al. (2009) recognized significant anthropogenic influence even for the background value over the marine boundary layer in the Yellow Sea and the East China Sea. Fu et al. (2013) reported emission data of ships travelling along the Grand Canal of China by conducting on-board emission tests and derived distance-based and fuel-based emission factors on the basis of the cruise and manoeuvring operations. During manoeuvring, the number concentration of nucleation mode particles, having size lower than 10 nm, was more than 30 times that was observed during cruise mode. The emission factor, in terms of particles emitted per km was far higher during normal cruise, with the engines at high load, being from 10 to 100 times those emitted when the engines are operated at low or middle loads. A relevant finding of this study was the observation that the particle concentration is lower during fog periods, which the authors indicated as consequence of relatively less ageing aerosol processes within the marine boundary layer. Zhao et al. (2013) reported data on air quality of the Shanghai port area gathered in 2011, considering the Vanadium and the V/Ni ratio as tracers for ships emitted particles. The mean concentration of V was 15.84 ng/m^3 in coastal airflows, compared with the average 9.84 ng/m^3 in continental airflows. Average daily concentrations of TSP and $PM_{2.5}$ were 114.39 and 62.60 $\mu g/m^3$, comparable with those in Shanghai land area, being the ship emissions responsible for 4.23% of the total $PM_{2.5}$ concentration on average. This is similar to the case of Los Angeles harbour (Minguillón et al., 2008), for which the high intensity of anthropogenic activities makes the effect of ships much closer to the Northern hemisphere average ships contribution to air pollution (Bond et al., 2013; Eyring et al., 2010).

Song (2014) recently reported that the PM_{10} and $PM_{2.5}$ emission in the port area of Yangshan, one of the busiest container ports of China, was 1078 T/year and 859 T/year respectively, being about ten times those measured in Europe. Chang et al. (2014) indicated that, the vessels in the port of Incheon (Korea) were responsible for about 142 T of PM emitted in the 2012, mostly occurring during the cruise and the manoeuvring phases.

Environmental regulation

As formerly discussed, number concentration is the preferred metric for assessing particulate matter health effects. However, with the exception of diesel cars, current air quality standards refer to mass concentration, so that actual emissions are below limits although in practice they remain extremely hazardous.

However, the authors envisage that technical issues, as the lack of instrumentation for large-scale continuous monitoring of ultrafine particles and of specific techniques for their removal, are among the reasons why the application of air quality standards based on number of particles is beyond the horizon of the next future. In fact, instrumentations for particulate measurement (ELPI, laser scanners, SMPS, DMA, etc.,) are not sufficiently cost effective and robust to be applied in a conventional regional or urban air quality monitoring networks. The presence of a qualified operator and a significant maintenance are also needed to manage the data.

Besides, the learning curve of the new technologies to allow compliances with submicron and ultrafine particles number based standards is still at its beginning and their commercial diffusion is not sufficient to supply the introduction of regulations so far. The only exceptions to this discussion are car engines, which are currently equipped with very efficient diesel particulate filters (DPF) and diesel oxidation catalysts (DOC). This allowed the introduction of specific regulations for particle reduction.

A summary of the main regulations pertaining to air quality standards in EU and USA, European car regulations and IMO guidelines is described in Fig. 3.

The International Maritime Organization (IMO) did not define specific guidelines for particulate matter control. IMO MARPOL Annex VI Regulation 14 indicates that a reduction in mass emission of particulate matter is achieved by reducing the fuels sulphur content. This switching to lighter fuels is imposed in the so-called Emission Control Areas (ECA) that are actually in force in the Baltic Sea, North Sea and Channel. The USA and Canada coastlines became ECA in 2011 and there are considerations for applying an ECA for the Mediterranean Sea (Panagakos et al., 2014), the Turkish Marmara sea (Viana et al., 2015), the Japan coastline and parts of Australia. European Union also defined more stringent limits for shipping in the territorial waters and for cruises within European ports. Recently, IMO (BLG 15 & MEPC 62) started discussions on the introduction of measures to determine and reduce black carbon emissions from ships in the lights of its climate forcing action in the Arctic region (Lack and Corbett, 2012; Litehouz et al., 2012).

Particulate matter reduction approaches

There are four main strategies to reduce particulate matter emissions in ships exhausts:

1. Reduce fuel consumption by energy efficient operations.
2. Reduce fuel consumption by improving ships design.
3. Improve engine performance and use cleaner fuels.
4. Adopt a proper exhaust gas cleaning systems.

In the first two cases, the reduction of particulate emission is expected to be proportional to the reduction of specific fuel consumption (Corbett et al., 2010; Litehouz et al., 2012). In the third case, the reduction of particulate emission is related to specific effects of fuel composition and combustion conditions and this effect cannot be predicted directly, but must be assessed by field experience. In the last case, the actual removal efficiency should take into account of both the specific features of the adopted technology and of its influence on normal ship functioning.

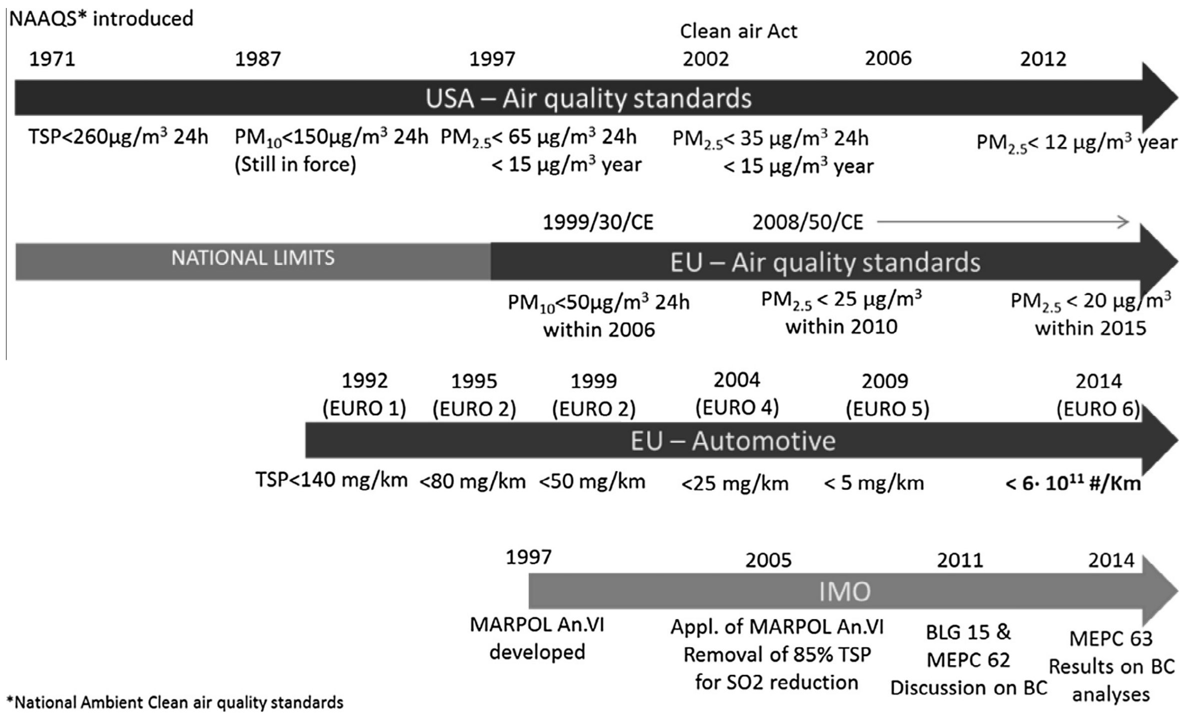

Fig. 3. Summary of air quality standards for particulate matter.

The details of the four strategies are reviewed in the following sections in terms of potential reduction of total particles (PM) and submicron (included ultrafine) particles (PM_1).

In these premises, however, it is important to remember that the design of more efficient ships and the assessment of optimal working procedures are two of the main paradigms of naval architecture towards its millenary history. In recent times, three main events gave rise to a significant improvement in energy efficient ship design: the oil shocks of the 1970s, the entry in force of the Kyoto Protocol on Greenhouse Gases Emission control in 2005, which was addressed by the IMO through different measure, among which the introduction of the Energy Efficient Design Index, EEDI, (International Maritime Organization, 2009; International Maritime Organization, 2014) and the financial crisis of 2008.

It is worth mentioning that the approach lying behind the EEDI parameter is that of establishing a roadmap towards a progressive reduction of ships emissions and of energy consumption compared with the 2008 reference conditions. A time route for the EEDI energy reductions goals was established for each class of ships, starting from the new units built from 01.01.2013 and organized in three steps: 0–10% for ships built within 01.01.2013 and 31.12.2019; 10–20% from 01.01.2020 to 31.12.2024 and 20–30% after 01.01.2025.

The EEDI is calculated according to a specific algorithm (International Maritime Organization, 2009; International Maritime Organization, 2014) that considers six main components:

- Ship design architecture.
- Main propulsion system.
- Auxiliary energy systems.
- Alternative propulsion system and energy recovery units.
- Fuel type.
- Routing and optimal operational procedures (SEEMP).

Among them, the fuel type contribution is meant as a measure of the C/H ratio, which correlates fuel consumption to CO_2 emissions. All the other measures directly affect the emission of all pollutants through a reduction of fuel consumption.

Energy efficient operations

This approach allows a simple and reliable way to reduce overall emissions from ships and can be classified under the "Ship Energy Efficient Management Plan, SEEMP" procedures (International Maritime Organization, 2012). The underlying principle of this approach consists in finding optimal operational practices that explicitly take into account the fuel costs and the environmental prescriptions while preserving the overall transport velocity of goods required by the markets. One of the most relevant parameter to assess this benefit is the so-called Fuel Operational Consumption (FOC) that is the actual fuel consumption per travelled route.

Among the most relevant ways to improve the FOC index, there are the improvement of ships routing, also accounting for weather conditions, and the so-called slow steaming.

The adoption of an optimal weather routing system that integrates GIS platforms and ships autopilot systems can allow operating under optimal weather conditions reducing fuel consumption and proportionally cutting all exhaust gas emissions. In this sense it is worth noticing that wave resistance is proportional to the six power of the speed while the frictional resistance is proportional to the third power of the speed (MAN Diesel and Turbo, 2011; Molland, 2008; Vergara et al., 2012) so that wave height is one of the most relevant parameter for actual ship consumption (Seo et al., 2013a,b; Vergara et al., 2012). Tsujimoto et al. (2013) showed, for example, that cruising a ship at 25 knots velocity in 3 or 4 m wave height seas requires from 22% to 33% more energy than operating into calm seas. However, weather routing effects should be considered in terms of actual FOC values and as part of an advanced routing system. In fact, optimal routing procedure aimed to fuel saving must consider both weather conditions and routing length. If, for example, it is planned to operate in optimal sea conditions so to save 20% fuel, but the new route length is increased by 20%, there will be no actual fuel gain from this procedure. Although potentially the weather routing can reduce high amount of fuel, advanced routing is a common practice on board ships and it is envisaged that its actual benefit can be in practice about 2–4% (International Council on Clean Transportation, 2011; Skjølsvik et al., 2000) and may be achieved by the introduction of auto-piloting systems (International Council on Clean Transportation, 2011).

The concept of slow steaming operation is mainly related to the functioning of the logistic chain: by minimizing berth time and defining just-in-time loading/unloading practices it is possible to reduce ship velocity with limited effects on the overall transport velocity of goods. According to the definition of "Admiralty coefficient", the propulsion power is proportional to about the third power of the ship velocity (MAN Diesel and Turbo, 2011). The specific fuel consumption, expressed as g/kW h of power was found to be a decreasing function of the fractional load that reaches an almost steady level (between 150 and 170 g/kW h) when the fractional load is over 20% (EPA, 2000). Therefore, in normal cruise conditions, the total fuel consumption almost linearly increases with the propulsion power and is almost proportional to the third power of the ship velocity (MAN Diesel and Turbo, 2011). Consequently, reducing ship velocity from 23 to 18 knots (in the so-called *slow steaming mode*) should theoretically cut more than 50% of the fuel consumption. The ICCT (2011) estimated a fuel saving from 15% to 19% for a 10% speed reduction and 36–39% for 20% speed reduction.

Chang et al. (2014) estimated that a 12 mile speed limit within the 25 mile zone around the Incheon port will cut PM emission by one third. Psaraftis and Kontovas (2013) recently reported a very detailed review on the optimization of ship speed as a market based practise, also considering emissions effects. From their survey, the authors concluded that slow steaming is largely practised on a voluntary basis but also pointed out the occurrence of several potential side effects from the introduction of specific speed limit regulations in the light of the projections on shipping market increases in the next future. Among them, the authors included the following:

– Building more ships to match demand throughput, with more CO_2 associated with shipbuilding and recycling.
– Increasing cargo inventory costs due to delayed delivery.
– Increasing freight rates due to a reduction in tonne-mile capacity.
– Inducing reverse modal shifts to land-based modes that would increase the overall CO_2 level.
– Implications on ship safety.

It must be noted that advanced routing and slow steaming may be less effective for ships travelling along coastlines with intense traffic and during manoeuvring in harbour areas, when low loads are compensated by emission factors (g/kW h) up until one order of magnitude higher than that at normal cruising (EPA, 2000; International Maritime Organization, 2009; Petzold et al., 2011) and distance from population is lower. However, optimal control of port use, in terms of optimal cargo handling and optimal berthing, mooring and anchoring, can lead to a fuel reduction of 1–5% and 1–2% respectively (Skjølsvik et al., 2000). The STATOIL recently demonstrated that optimizing sailing speeds and type of supply vessels can reduce emissions in the upstream supply chain to offshore installations by 10%, with peaks up to 25% (Norlund and Gribkovskaia, 2013; Tesfay, 2014). The potential particulate matter reductions related to the different energy efficient operations are resumed in Table 2 in terms of PM and PM_1 removal.

Expected PM reduction associated with fuel savings coming from the implementation SEEMP practices may potentially reach 40%. These comes mainly from the recourse to slow steaming and weather routing that, however, are strongly dependent on the actual ship route and on the logistic chain effects.

Improved ship design

Optimal ship design can generate significant benefit in terms of specific fuel consumption. One of the most interesting parameters to quantify the effectiveness of this method is the EEDI, since the reduction of PM emissions is directly proportional to the fuel saving related to the increase of efficiency. Several methods to accomplish reduction of fuel consumption by ship design improvements were proposed and their effects on PM and PM_1 removal are listed in Table 3 assuming them as proportional to observed fuel saving.

Improvement of ship design may be distinguished into two main categories: improvement of ship architecture and adoption of use of alternative propulsion system and/or energy recovery units.

In terms of ships architecture design, the main improvements are related to the following: (i) propeller design optimization; (ii) ship hydrodynamics and aerodynamics, included hull coating and cleaning; (iii) plant design; (iv) ballast water and trimming optimization; and (v) weight reduction.

Vergara et al. (2012), Seo et al. (2013a) and Skjølsvik (2000) indicated that improved propeller design can contribute to about 4–5% improvement of ships energy consumption. The ICCT (2011) reported 0.5–3% for propeller upgrade (nozzle and tip winglets), 1–3% for propeller boss cap with fins, 2–6% for complete propeller–rudder replacement.

Major ships hydrodynamics improvements include bow design, stern bulbs, stern flaps and slender hulls that reduce wave resistance and may lead to 5–20% reduction of fuel consumption (Skjølsvik et al., 2000). Reduction of skin friction can be instead achieved by the use of antifouling paints for hull coating/cleaning or by the use of air lubrication. Periodic hull cleaning reduces fuel consumption by up to 10% (Corbett et al., 2010; International Council on Clean Transportation, 2011) while anti-fouling coating was associated with fuel reductions from 0.5% to 5% according to the ICCT (2011) and from 2% to 9% according to Corbett et al. (2010). The air lubrication is more complex, but was associated with higher fuel reductions, between 10% and 15%, as reported by Mizokami et al. (2010), Kumagai et al. (2015) and by the ICCT (2011). Lower values were indicated by the ICCT (2011) for container vessels and by Corbett et al. (2010), who reported a minimum reduction of 3.5%.

Table 2
Particulate matter reduction potentialities for energy efficient operations.

Energy efficient operations		
Strategy	% PM reduction	% PM_1 reduction
Slow steaming	15–40	15–40
Weather routing	20–30	20–30
Advanced routing	2–4	2–4
Optimal cargo handling	1–5	1–5
Optimal berthing, mooring and anchoring	1–2	1–2

Table 3
Particulate matter reduction potentialities by ship's design improvements.

Ship design		
Strategy	% PM reduction	% PM_1 reduction
Ship's architecture		
Ballast water and trimming optimization	1–5	1–5
Plant design	1–4	1–4
Propeller design optimization		
– Propeller upgrade (nozzle and tip winglets)	0.5–3%	0.5–3%
– Propeller boss cap with fins	1–3%	1–3%
– Complete propeller–rudder replacement	2–6%	2–6%
Weight reduction	<5%	<5%
Air lubrication		
– Tanker/bulker	10–15%	10–15%
– Container	5–9%	5–9%
– Fishing boats	1–4%	1–4%
Aerodynamics	3–4%	3–4%
Hull coating	0.5–9%	0.5–9%
Hull cleaning	1–10%	1–10%
Power train optimization	<2%	<2%
Optimal hull design	5–20%	5–20%
Advanced autopilot	0.5–3%	0.5–3%
Auxiliaries		
Pumps and fan velocity control	<1%	<1%
Machinery monitoring	0.5–1%	0.5–1%
Alternative propulsion system and energy recovery units		
Hybrid battery and diesel–electric main propulsion system	<30%	<30%
Waste heat recovery	5–15%	5–15%
Waste heat recovery with organic ranking cycle	6–9%	6–9%
High efficient lightning	<1%	<1%
Solar energy	2–10%	2–10%
Wind assistance (sails/kites)	2–32%	2–32%
Wind assistance (Flettner rotors)	3.5–12.5%	3.5–12.5%

The optimization of power transmission system (Vergara et al., 2012), plant design and the weight reduction and balancing (International Council on Clean Transportation, 2011; Skjølsvik et al., 2000) was associated with additional fuel reductions between 1% and 5%. A small machinery monitoring (Skjølsvik et al., 2000) may lead to a fuel reduction of about 1%. Finally, an advanced adaptive autopilot system can be used to optimize rudder position considering winds, currents and ship yawing so as to minimize the ship resistance, which was associated with an energy efficiency potential between 0.5% and 3% (International Council on Clean Transportation, 2011).

A small contribution (<1%) to fuel reduction may come from auxiliary systems, by using lower lightning and pumps and fan velocity controls (International Council on Clean Transportation, 2011).

The use of alternative propulsion system and energy recovery units is an interesting option, although it may require high investment costs and specific vessel design. Wind propulsion, solar energy, hybrid energy storage and waste heat recovery processes are the most promising techniques.

Wind and solar powers are potentially very interesting, but there is a strong correlation among their effectiveness and the specific ships routes. In this sense, the overall economy of these approaches should present a break-even point strongly dependent on climatic condition under which the ship operated. Unfortunately, this information is not available to us at date.

Wind assisted navigation was first tested in the 1980s (Smith et al., 2013) and proved to reach a fuel savings from 10% to 50%. The ICCT (2011) and the Litehouz et al. (2012) reported an average energy savings from 3.5% to 12% from Flettner rotor and 2–26% for kites. Nuttal et al. (2014) reported results on the retrofitting of two small passenger/cargo ships with auxiliary sail rigs, which resulted in fuel savings of 23–30%, increased stability, increased passage speeds, and reduced engine wear. Traut et al. (2014) reported numerical performance models of two wind power technologies, Flettner rotors and kites, showing that Flettner rotor may provide from 2% to 21% of the engine power while the kite contributed from 1% to 32% of the engine power. Traut et al. (2014) highlighted that kites require less deck space but are less stable than the Flettner rotors.

Solar power was proposed in combination with either the auxiliary or the main engines. Data on potential fuel recovery were largely scattered, resulting between 2% and 3.5% (International Council on Clean Transportation, 2011), 5% (Cheng and Hirdaris, 2012) or within 5–17% (Litehouz et al., 2012). Of course, these percentages depended on the actual panelled area and by the total ship power. Therefore, in general, it is possible that larger ships exploited lower percentage power reduction. The first cargo ship propelled by solar panel was the M/V Auriga Leader, that provided about 10% of the total electrical needs

(Pearsons, 2009). Solar panels were mounted on the Nichioh-Maru, a Nissan car transporter ship, providing fuel savings of 1400 tons per year (Quick, 2012).

Dedes et al. (2012) investigated the economical reliability of integrating hybrid batteries and diesel–electric main propulsion system for different kinds of ships, calculating fuel savings up to 28% for ships travelling in ballast and up to 19% for loaded ships. Baldi and Gabrieli (2015) indicated that waste heat recovery may achieve fuel savings between 5% and 15%, depending on the sources of waste heat used and on the efficiency of the waste heat recovery system. Larsen et al. (in press) calculated that a system including low-speed two-stroke diesel engine, turbochargers and an Organic Rankine Cycle may achieve energy reductions up to 9%.

In summary, several ship design improvement can lead to reduction of fuel consumption, and of associated PM emissions up to 35%. Better performances can be achieved by integration of more technical improvement. These can be valuable for new construction, but are less feasible for retrofit of existing units.

Engine and fuels modifications

As discussed in former sections, the emissions of particulate matter depend on fuel composition and quality of combustion. Therefore, a first possible approach to reduce PM emissions is to improve specific fuel consumption and/or adopt cleaner fuels as alternatives to IFO. These approaches guided the development of new marine diesel engines with higher efficiencies and able to burn lighter fuels. In this sense it is worth noticing that the last 50 years led to a significant reduction of marine engine specific fuel consumption. The main spring to this reduction was the fuel crisis of 1970s: the fuel consumption passed from about 210 g/kW h to about 170 g/kW h in the 1980s (della Volpe, 2007). In recent times, the websites of the main engine producers indicate fuel consumptions at about 155–170 g/kW h.

The diesel engines should be modified to proficiently improve combustion and reduce PM by modifications of injectors, slide valves, engine de-rating for slow-steaming, etc. and it was estimated (Entec, 2005; Litehouz et al., 2012) that combination of these techniques can save up to 50% of particulate emissions by weight. Use of humid air motors or direct water injection in the combustion chamber was also proposed (Entec, 2005), but its effect was limited to the reduction of NO_x emissions. The ICCT (2011) estimated that a new tuning of the main engine may lead to a reduction of fuel consumption from 0.1% to 0.8%. With this retrofit, the most commonly used load range is determined and then the engine is optimized for operation at that load. This requires a different engine mapping and entails changes in injection timing. An additional upgrade to a common rail system can lead to a further improvement of 0.1–0.5% (International Council on Clean Transportation, 2011). Recently, the Wärstilä (2013) introduced a new kind of gearbox that can reduce ships consumption for about 8% and potentially up to 15%. Table 4 resumes the estimated effects of engine improvements on particulate matter reduction.

The use of alternative, low sulphur, liquid fuels (as diesel or biodiesel) or synthetic fuels (as water in fuels emulsions) is actually the main emission control strategy applied by the IMO (MARPOL Annex VI Reg. 14) to reduce sulphur based emissions, and included those related to sulphates particles, which compose the majority of IFO fuelled ships emissions in weight. This decision was based on several studies dating back to the beginning of the century (Entec, 2005) that selected this method as the most feasible political means to produce reduction of SO_2 and PM emissions.

When these fuels are adopted, only a retrofit of the engine is required to assure a proper system tuning that accounts for the different viscosity of the fuels and for the optimized combustion conditions. Additionally, different formulation of the lubricating oils is required to take into account the different acidity compared with IFO fuels (della Volpe, 2007; Woodyard, 2004). Since the last months of 2014, the so-called hybrid-fuels, i.e., blends of IFO and MDO with sulphur weight content close to 0.1%, were commercialized. Ash contents spanned between 0.003% and 0.07% (Lloyd's Register Marine – Fuel Oil Bunker Advisory Service, 2014). These were called Ultra Low Sulphur Fuel Oils, ULSFO 0.1%. One of the main R&D topics of the last years is the design and construction of LNG fuelled ships (Brynolf et al., 2014). The use of LNG ships is a groundbreaking proposal since the LNG provided a substantial reduction of pollutants because the emissions of SO_2, NO_x and PM are negligible. However, methane and hydrocarbons slip and CO emissions must be considerably higher than that of diesel engines (Abdelaal and Hegab, 2012; Cheenkachorn et al., 2013; Kado et al., 2005). These emissions of hydrocarbons may be reduced by introduction of exhaust gas recirculation systems.

The use of LNG suffers for several drawbacks among which the need for severe modification of the harbours infrastructures, the introduction of new safety and logistics practices as well as the development of specific skills for service and

Table 4
Particulate matter reduction potentialities by engine modifications.

Engine modifications		
Strategy	% PM reduction	% PM_1 reduction
Slide valves	10–50	n.a.
Slow steaming with engine de-rating	1–4	1–4
Humid air motor	n.a.	n.a.
Main engine retrofit – tuning	0.1–0.8%	0.1–0.8%
Main engine retrofit – tuning + common rail	0.1–1.3%	0.1–1.3%
Improved gear system	8–15%	8–15%

maintenance teams and for seafarers. The same ship design must consider new safety aspects, space allocation for LNG tanks, which cannot be stored as a liquid fuel. Recently Acciaro (2014) discussed the economic aspects of investments on LNG retrofit to comply with ECA regulations. The author believes that such investment carries substantial risk due to the significant upfront costs and the high degree of uncertainties on the price differential between LNG and conventional maritime fuels, as well as on the availability of LNG and the reliability of its supply chain.

Recently, the use of methanol (both from natural gas and from biomass) was proposed as a mean to cut gaseous emissions (Brynolf et al., 2014), but data on the particulate emissions from ship engines are not yet available.

An interesting strategy adopted by ship manufacturers is the development of dual fuels engines that allow operating with either liquid or gas fuels and/or switching from one another at certain engine load. In most applications of this technique, natural gas is inducted or injected in the intake manifold to mix uniformly with air, and the mixture is then introduced to the cylinder as a result of the engine suction (Abdelaal and Hegab, 2012). Usually, these engines are started with diesel fuels and are operated with gas over a certain engine load. Biofuels were also proposed as pilot fuels for the engine starting phase (Namasivayam et al., 2010; Park et al., 2014).

The sustainable use of alternative fuels must take into account several specific issues regarding the effects on PM emission, the costs and the availability of the fuels that are discussed in the following subparagraphs.

Effects on PM emissions

The use of lighter fossil fuels, with lower sulphur content, is known to reduce the emission of sulphate particulate matter only, leaving almost unchanged the emissions of soot particles (e.g., International Maritime Organization, 2009), which are the most toxic. Referring to tests carried out on marine diesel engines fuelled with IFO (1.6% S) and MGO/ULSF (ultra low sulphur fuel 0.03% S), it was demonstrated that "*Specific PM emissions were generally higher for IFO than for the MGO*; *however, for the smallest size-fraction measured, containing particles 0.30–0.40 μm in diameter, the opposite was observed. This finding emphasizes that to minimize negative health effects of particles from ships, further regulation may be needed to reduce small-sized particles; a fuel shift to low sulphur fuel alone does not seem to accomplish this reduction*" (Winnes and Fridell, 2009).

Winnes and Fridell (2009) noticed that for an engine load between 65% and 70%, the weight emissions decreased from 0.53 to 0.37 g/kW h passing from the IFO to the ULSF, but the total particle number concentration increased by a 10%. The authors also analysed, on the bases of literature data, a trend of emissions factors, in g/kW h, for diesel fuels with sulphur content from nearly 0% to 2.5%. The authors noticed that particle emissions were about 0.2–0.4 g/kW h for ULSF, remained in the range 0.4–0.6 for sulphur content lower than 2%, while for larger values, more scattered data, varying from 0.5 to 3 g/kW h were registered. Verbeek et al. (2011) indicated a baseline value of PM emissions from diesel fuels of 0.2 g/kW h plus an additional contribution proportional to the sulphur content in the fuel (in grams of sulphur per gram of fuel) through a factor between 0.138 and 0.184 g of PM per gram of sulphur. Petzold et al. (2011) observed that at 75% load, total PM decreased by 90%, and EC and OM contents lowered by about 50% by using a distillate fuel (<0.1% S) in place of an IFO fuel (2.17% S): The emission factors for the distillate fuels for PM, EC and OM were, respectively, 0.072, 0.007 and 0.058 g/kW h. Ushakov et al. (2013a), compared a conventional marine gas oil and gas-to-liquid Fischer–Tropsch (GTL) fuel, showing that the number of emitted particles increased for the GTL is 21% higher than for the MGO, being the particle size distribution almost unchanged.

In a recent review, Lack and Corbett (2012) indicated that, improvements to fuel quality (from residual to distillate fuels) can reduce BC emissions by an average of 30% and potentially up to 80% but specific consideration on the effect of distillate fuels on each component of particulate matter must be carefully assessed. Jayaram et al. (2011) measured PM emissions of an ULSF (10 ppm S) of about 0.174 g/kW h on a four strokes 500 hp Tier II marine diesel engine: the contributions of EC and OM were 0.080 and 0.094 g/kW h respectively. Tests ULSFO 0.1% are not still available on ships, but Anderson et al. (2015) recently reported the results of tests on a IFO fuel blend containing 0.12% sulphur in a test-bed engine laboratory equipped with a 4-stroke, turbocharged Volvo Penta D3-110 marine diesel engine. The authors found that no or small differences appeared in terms of particles number between the 0.12% sulphur IFO fuel and MDO, but larger emissions appeared for particle diameters larger than 50 nm. Sulphate and ashes emissions are reduced due to their small concentration in the fuel.

A possible alternative is the use of water in fuel emulsions (WIFE), a suspension of water in fuel produced on-site starting from fuel oil or diesel water and proper additives. Several commercial applications on WIFEs are available for off-road diesel engines. Results of trial tests of a WIFE in an ferry vessel engine (Corbett et al., 2010; Lyons, 2003) indicated that a reduction of PM emission of 42% in mass was achieved, but an energy loss (and an opposite increase of fuel consumption) from 8% to 12% was also registered. Tests on the air quality in an underground mine contaminated by an off-road diesel engine (Noll, 2006) showed that when the same engine was fuelled with a WIFE, the mass concentration of EC reduced by a factor within 70–85% and total PM reduced within 50–70%. However, a corresponding increase of OM up to 40% was also shown. Unfortunately, there are no data regarding the number concentration of the emitted particles, leaving lack of information on the actual effects of WIFE on the ultrafine soot particles concentration. The reduction of EC and PM associated with WIFEs is likely to be related to the occurrence of competitive partial oxidation and gasification reactions reducing the production of pyrolysis solid by-products. The same phenomena are expected to increase the emissions of OM, as shown by Gualtieri et al. (2014). WIFE also improves the emission of NO_x by reducing the combustion temperature.

Recently Petzold et al. (2011) analysed the emission profiles of several biogenic fuels (soybean, sunflower, palm oils and of animal fat) in a 400 kW single cylinder engine, comparing the data with the emissions of MGO (ULSF < 0.1% S) and IFO (2.17% S). The fuels were de-gummed and de-acidified but not trans-esterified, so appearing as biodiesel precursors, which can be directly used on marine diesel engines designed to operate with high viscosity fuels. The experimental results pointed out that while the emissions of gaseous pollutants for the biofuels are usually lower than those observed for the ULSF, particulate matter emission is higher for all the PM constituents (OM, EC/BC). With reference to IFO fuels, the first advantage of biofuels is that ashes and sulphate fractions are absent in their exhaust gases. The emission of OM results is comparable with that of IFO while the EC is up to 30% lower for palm oil and animal fat but is higher for soybean and sunflower oils. In terms of total emitted particles, biofuels emissions were similar to the IFO fuel emissions, although number of non-volatile particles (EC) is about 50% lower. Investigations on the exhaust gas toxicity are under development and Bunger et al. (2012) reported that toxicity may be substantially higher for biogenic rather than for conventional diesel, mainly due to the higher NO_x emissions. In this sense, it is worth remembering that Zhang and Balasubramanian (2014) indicated the need to understand the role of lubricating oil in determining particles emissions and toxicity.

In a recent paper, Anderson et al. (2015) specifically investigated the effects of fuel composition on particulate emissions from marine engines at low (from idle to 35%) loads. The authors pointed out that the number of emitted particles increases by one order of magnitude by reducing the load from 35% to idle, probably because the low temperatures favoured the condensation of heavy molecular weight organics deriving from the unburnt fuel and oil. Moreover, the author showed that the particles number emissions of MDO (0.52 wt% S), MK1 (<0.0003 wt% S) and MK3 (0.0003 wt% S) fuels were substantially lower, being about 50%, <3% and <4% of that emitted by using an IFO fuel. The particle size distribution showed peaks at 10 nm and 45–50 nm for distillates and 10 and 100–110 nm for HFO. Moreover, the authors monitored the concentration of heavy metals in the particles, showing how these can be related to both the fuel composition and the lube oil as well as to engine wear phenomena.

Potential reductions in particulate matter emissions deriving from the use of biodiesels (B20, B50, B100), E85 (ethanol–gasoline 85:15 blends) and LNG in heavy-duty diesel engines, are currently available only in comparison with ULSF.

Jayaram et al. (2011), reported tests on ULSF and soybean biodiesel B20 and B50 blends in marine diesel engine. At 75% load, the use of B20 and B50 led to reduce PM emissions from 0.178 g/kW h to 1.34 and 1.14 g/kW h; EC emissions reduced from 20% to 42%, while OM emissions lowered from 20% to 30%. However, the biodiesels provided lower particles sizes that lead to more conspicuous emissions of particles in number: +170% for B20 and +350% for B50.

Betha and Balasubramanian (2011) compared the emissions of ULSF and biodiesel blends (B20, B50 and B100) in a single-cylinder, four-stroke, air-cooled, direct fuel injection, diesel backup power generator (L70AE; Yanmar Corp, China). It was found that with an increase in percentage of biodiesel in the fuel mixture, particle mass was reduced by 16.3% for B20, 20.2% for B50, and 35.1% for B100 during idle mode and by 12.2%, 26.9% and 36.6% respectively at full load. However, the particle size distribution shifted to smaller diameters (from 52–100 nm for ULFS to 34–52 for B100 at 20 and 100% load), so that the particle number remained unchanged. Lapuerta et al. (2012) on the basis of statistical data showed that the logarithm of the ratio between total PM emitted by a biodiesel blend and the total PM emitted by the pure reference fossil fuels is proportional to the percentage of biodiesel in the blend multiplied for a factor equal to -0.006388. The highest reduction, for a pure B100 was assessed at about 50%, but lower efficiencies were expected for heavy-duty diesel engines.

Sarjovaara and Larmi (2015) found that the filter smoke number of the exhaust gas coming from a heavy-duty turbocharger with common rail engine remained unchanged for gasoline and E85.

Finally, with reference to LNG, Holmen and Ayala (2002) and Kado et al. (2005) highlighted that the particulate emissions of heavy duty engines fuelled with LNG were comparable with those achieved for ULSF engines equipped with DPF (almost 1/10 of the ULSF emissions). Verbeek et al. (2011) reported a similar estimation (0.02 g/kW h) for LNG ships.

The effects of adopted fuel on particulate matter emission are resumed in Table 5. In this case, data for the referenced authors were used to estimate potential reduction efficiency towards different components of the fuels using both IFO with 2.17% sulphur – the same adopted by Petzold et al. (2011) – and ULSF as reference baselines and considering data for engines at 75% load.

Fuel cost

Updated information on fuel price can be gathered by specific website as that of the United States Department of Energy (United States Department of Energy, 2014) or from the United States Energy Information Administration (United States Energy Information Administration, 2014). At date, the average price differential between IFO fuels (either IFO 380 or fuels with higher viscosity) and lighter marine gasoil (MDO or MGO) is around 300–350 $/kg. Similar differences applies with bio oils (palm, soybean, sunflower oils, etc.) while biofuel blends as E85 (85% ethanol–15% gasoline) or pure biodiesel (B5, B20, B100) have price differentials of about 400–500$/kg. LNG/LPG are a little less costly (around −60$/kg) than IFO. Price differential with respect to IFO is also reported in Table 5.

The price differential is a critical issue for shipping since the same price of IFO fuels was already a critical OPEX costs for ship-owners and charterers. A report of the United Nations Conference on Trade and Development (United Nations Conference on Trade and Development, 2012) claimed: "*In 2008 the annual capital cost of a new Panamax bulker was 6 million of USD and the annual bunker cost was 3.3 million of USD. In 2011, the capital cost lowered to 2 million USD and the bunker cost (IFO 380) was 5.5 million USD. The fuel costs are estimated to have made up 60% of the total freight earnings on the benchmark very large crude carrier (VLCC) Western Asia to Far East voyage*". It is clear that shifting to lighter fuels may further increase market pressure on the maritime sector potentially leading to an increased cost of transported goods. Besides, this shift can frustrate political actions aimed to transport modal shifting towards marine/inland waterways, as for the European Union, where positive actions have been launched to move more road and rail transport onto short sea and inland routes,

Table 5
Particulate matter reduction potentials by fuel modifications compared with IFO (2.2% S) in Petzold et al. (2011) and ULSF (<30 ppm S). Price differentials from United States Energy Information Administration (2014) and U.S. Dept. Energy (2015).

		Alternative fuels								
		EC, g/kW h	OM, g/kW h	Ash, g/kW h	Sulphates, g/kW h	BC, g/kW h	Total number $\times 10^{14}$ 1/m^3	PM, g/kW h	IFO Price differential, $/Tonne	References
Pseudo-ULSFO 0.1%S (IFO 0.12%S)	IFO	–	–	95,0	99,0	n.a.	–	>90	180–200	Anderson et al. (2015)
	ULSF	–	–	–	–	–	–	–		
ULSF	IFO	40–50	50–60	95,0	99,0	87,1	<10	>90	400–500	Petzold et al. (2011) and Winnes and Fridell (2009) Jayaram et al. (2011)
	ULSF	–	–	–	–	–	–	–		
MDO (1%)	IFO	<10	<10	50–60	50–60	n.a.	<10	75–80	300–350	Winnes and Fridell (2009)
	ULSF	–	–	–	–	–	–	–		
Palm Oil	IFO	38	−38	91	100	84	−220	75	300–350	Petzold et al. (2011)
	ULSF	−14	−207	−50	60	−23	−220	−165		
Animal fat	IFO	38	−22	91	100	81	−274	78	300–350	Petzold et al. (2011)
	ULSF	−14	−171	−50	80	−46	−274	−135		
Soybean oil	IFO	−15	17	94	100	67	−95	83	300–350	Petzold et al. (2011)
	ULSF	−114	−84	0	60	−154	−95	−75		
Sunflower oil	IFO	−46	−26	94	99	82	−133	75	300–350	Petzold et al. (2011)
	ULSF	−171	−179	0	0	−38	−133	−161		
WIFE	IFO	70–85	−40	>98	>99	50–90	n.a.	n.a.	n.a.	Corbett et al. (2010), Noll (2006), and Lyons (2003)
	ULSF	70–85	−40	n.a.	n.a.	50–90	n.a.	30–45		
B20	IFO	53–55	63–65	>98	>99	100	n.a.	90–92	400–500	Jayaram et al. (2011) Betha and Balasubramanian (2011) Sarjovaara and Larmi (2015)
	ULSF	12–17	18–21	n.a.	n.a.	100	−170	12–19		
B50	IFO	60–70	65–68	>98	>99	100	n.a.	94–96	400–500	Jayaram et al. (2011) Betha and Balasubramanian (2011) Sarjovaara and Larmi (2015)
	ULSF	27–42	27–29	n.a.	n.a.	100	−350	35–37		
B100	IFO	65–73	70–78	>98	>99	100	n.a.	95	400–500	Betha and Balasubramanian (2011) Sarjovaara and Larmi (2015)
	ULSF	36–50	36–50	n.a.	n.a.	100	n.a.	36–50		
E85	IFO	40–50	50–60	95	99	87	<10	>90	400–500	U.S. Dept. Energy (2014)
	ULSF	–	–	–	–	–	–	–		
LNG	IFO	95	96	>98	>99	99	n.a.	99	−50	Acciaro (2014), Verbeek et al. (2011) U.S. Energy Information Agency (2014)
	ULSF	90	90	n.a.	n.a.	90	90	90		

which are more cost effective and emit less pollutants and GHGs per ton-mile compared with aviation, rail and car transport (European Environment Agency, 2014; Natural Resources Defence Council, 2014).

The potential shifting of shipping from IFO to distillate fuels should also take into account the effects on refined products demand and on oil refining industry. With current crude oils, conventional distillation techniques produce up to 50% of residue oils, while gasoil yields are about 20% – see for example the BP crude assay website (British Petroleum, 2015). Diesel demand is now around 35% and projections indicate that 40% level will be required in 2030 (Calzado Catalá et al., 2013).

In order to comply with this requirement, hydrocracking, vacuum distillation, cocking, hydro-treating processes are used, generating additional fuel costs. To date, the IFO market is actually a niche market mainly devoted to marine applications. If fuel switching is applied at large scale to date, an additional 15% diesel yield increment is needed (Fig. 2). This requires further investments on refineries processes. Moreover, the maritime transport will become one of the market competitors for diesel fuels and may be exposed to higher market pressure. Similarly, we envisage that, although the development of LNG is a valid alternative in the light of an energetic mix approach, the conversion of the overall fleet to LNG and the construction of proper infrastructure still need a detailed political discussion at national and international levels.

Exhaust gas cleaning systems

The removal of particulate matter in marine diesel engine exhaust is a complex issue related to its tri-modal particle size distribution. As a general rule, two main particle fractions can be considered: (i) a fraction coarser than 2 μm, mainly composed by ashes and sulphate particles and (ii) a finer fraction represented by particles in the so-called Greenfield Gap particles (0.1–2 μm), composed by soot, finer ashes and sulphate particles, and by the soot particles in the ultrafine-nanometric range (<200 nm). The removal efficiencies for these technologies are resumed in the last columns of Table 6, which also reports an estimation of the corresponding Technology Readiness Level (TRL) for marine application based on the European Union definition (European Commission, 2014). The TRL is a reliable parameter to evaluate the stage of development and maturity of each of the after-treatment technologies, some of which are still at laboratory scale demonstration level.

Removal of particles larger than 2 μm

These particles have sufficient inertia to be collected by wet scrubbers (WS) or cyclones (CYC). The mechanisms that govern particles capture are related to the hydrodynamic interactions between the particles and the collecting surfaces via the gas streamlines in the surface boundary layer.

With specific reference to existing on-board exhaust gas cleaning systems, the coarse particles may partially be removed by selective catalytic reduction (SCR) reactors for NO_x reduction and by dry or wet scrubbers for SO_2 capture.

SCR units are monolithic fixed beds with square holes of length larger than some millimetres. This size allows avoiding catalyst plugging and poisoning related to particles entrained in the gas stream. Therefore, although a partial deposition of coarse particles in the SCR can be achieved since each hole acts as a small and long sedimentation chamber, the removal efficiency was negligible and limited to the largest particles only. In a recent study, Hallquist et al. (2012) showed that the number of particles emitted from a ship (including the 12.6 MW main engine plus auxiliaries) fuelled with MDO (0.5% S) and equipped with a SCR, was comparable to ships running on higher fuel sulphur content, being about $10.4 \pm 1.6 \times 10^{16}$ particles per kg of burned fuel and mainly in the range of ultrafine particles. In addition, the authors found that a clear connection with the sulphur content could not be established. The SCR is actually ineffective in removing soot particles.

Wet scrubbers are spray towers loaded with different liquid-to-gas mass ratio aimed to quench the gas stream and abate SO_2 according to the MARPOL Annex VI Reg.14. To this end, seawater or chemical solutions of basic/alkaline compounds in either freshwater or seawater are used (Gregory, 2012). When simple seawater is used, higher water loading is required to comply with SO_2 emission requirements compared with chemical scrubbing. However, particle removal efficiency in

Table 6
Particulate matter reduction efficiencies for exhaust gas cleaning systems.

Exhaust gas cleaning systems			
Strategy	% PM reduction	% PM_1 reduction	TRL
Selective catalytic reduction	n.a.	Negligible	9
Wet scrubbers	<85	n.a.	9
Venturi scrubbers	>90	<50	9
Fabric filters	>90	>90	5
Diesel particulate filters	<95	<95	5–6
Diesel oxidation catalysts	<95	<95	5
Electrostatic precipitators	>90	60–80	5
Particle agglomerators	n.a.	90	4–5
Wet ESP	n.a.	n.a.	5
Wet electrostatic scrubbers	<85%	>90 in number	5
Heterogeneous condensation assisted scrubbers	n.a.	n.a.	3–4
Bubble towers	>90	>90	3–4

scrubbers is an exponential function of liquid-to-gas ratio and, therefore, particle capture is more effective for seawater scrubber than for chemical scrubbing (Carotenuto et al., 2010b).

The majority of marine scrubbers include a Venturi unit prior to a SO_2 absorption tower. The Venturi scrubber favours hydrodynamic interactions among particles and sprayed droplets thus allowing appreciable removal efficiency for particles larger than 1 μm (Hesketh, 1996). Special cyclonic scrubber designs as that developed by Cleanmarine, allow reducing the larger soot particles, with size as low as 500 nm. Dry scrubbers, instead, remove particles by filtrations since these units are composed by long fixed bed columns, several metres high, with particles with size from 1 to 10 mm. Dry scrubbers for marine applications were developed by Couple Systems, but unfortunately, there are no available data on particle abatement from the unit installed on the Oceanex MV Connaigra in 2013. It is worth mentioning that, conceptually, both wet and dry scrubbers may exploit a small reduction of nanometric particles as a consequence of their Brownian diffusivity (Carotenuto et al., 2010b), but there is no available data on such phenomena in commercial marine scrubbers.

Removal of particles finer than 2 μm

For particles finer than 2 μm, inertial effects are useless and particle capture can be achieved by means of Brownian diffusion phenomena or phoretic effects or by applying electromagnetic forces.

In particular, particles in the Greenfield gap are difficult to capture since both inertial properties and diffusional behaviour are insufficient to assure proper capture efficiencies by hydrodynamic interactions. In this range, electrostatic precipitators ESP and fabric filters FF are commonly adopted for land-based applications. A longstanding industrial experience in these technologies is available for the removal of $PM_{2.5}$ particles to comply with air quality standards regulations, but both ESP and FF are far less effective when diesel soot particles are considered.

As regards filtration, the commercial units for land-based applications are very effective in removing particulate matters but are susceptible to gas humidity or the presence of sticky oil droplets and tend to generate high-pressure drop to remove particles finer than 200 nm with high load. This makes unreliable the direct application of industrial FF to the treatment of exhaust gas from ships engines. Specific filter designs were developed for the automotive industry, allowing the introduction of stricter environmental regulations for diesel cars. The two most adopted units for diesel particle filtration are diesel particulate filters (DPF) and diesel oxidation catalysts (DOC), both of which are expected to reduce more than 90% of emitted particles. Catalytic oxidation or thermal decomposition of the soot deposited on the filter is periodically carried out to avoid filter clogging and, consequently, to contain pressure drops that can also reach 100 mbar before regeneration (Adler and Petasch, 2013; Mokhri et al., 2012). Concerns on particulate and gaseous emissions during filters regenerations were also reported (Mamakos and Martini, 2011). When operated with IFO or with MDO/MGO fuels, the DPF and the DOC become largely ineffective because of the progressive filter pores clogging by the coarse particles. In addition, catalysts have to be periodically replaced to preserve system functionalities and require clean hydrocarbons for regeneration. This further complicates the DOC application on board ships. Besides, concerns on filter size and pressured drops must be considered for on-board application.

In 2012, the Mitsui O.S.K. provided evidences of trial tests on the application of a DPF system on a 9 MW engine of a ferry fuelled with IFO, reporting a particle removal efficiency of 80% in mass and an operation time of 500 h, but without indication on system operation in terms of filter regeneration practices (Mitsui and Lines, 2012). To avoid filter clogging, DPF can be adopted only in conjunction with fuels containing less than 500 ppm (Corbett et al., 2010), but studies to increase this level up to 2000 ppm (0.2% w/w) are ongoing (Mayer et al., 2011).

Electrostatic precipitators (ESP) are based on the use of corona effects to charge the particles and collect them on grounded plates. The particles are attracted towards the plates with an electric drift velocity proportional to the acquired charge. The system is very useful in collecting particles with low inertia but particle conductivity and particle cohesiveness play a key role in determining particle re-entrainment from the collection electrode. The removal of ultrafine soot particles in ESP is a challenging process because of the physical limitations in effectively charging these particles, so that electrical drift velocity is low and a reduction of collectors plate distance and/or a higher residence times is required to make the ESP effective (Huang and Chen, 2002; Jaworek et al., 2007; Yoo et al., 1997; Zhuang et al., 2000). Jaworek et al. (2007) reviewed conventional and innovative technologies for electrostatic precipitation with specific reference to submicron particles. The authors described several innovations to conventional ESP, such as particle agglomerators, multi-stage ESPs, electro-cyclones and use of complex electrodes design. In particular, tests on diesel particles were successfully carried out with particles agglomerators, PA (Masuda and Jae-Duk, 1983; Zukeran et al., 2000), for which removal efficiency of more than 90% in mass was achieved for 1 μm particles. Lower efficiency (60–80% in mass) was reached with an ESP equipped with a barrier discharge dielectric layer (Kuroda et al., 2003).

A hybrid technologies, that combine ESP and FF is the so-called electrostatic fibrous filter (EsFF). EsFF are filters made of textiles that enclose charging wires to couple filtration and electrostatic deposition. These units are very effective, but the textiles are prone to burn due to the effect of electric discharge. Therefore, the application of EsFF is now mainly focused on indoor air quality control and bioaerosol removal (Ardkapan et al., 2014; Eyring et al., 2005b).

Innovative "wet" technologies appear as promising alternatives to ESP and DPF/DOC for the capture of submicron particles. Among them the Wet Electrostatic Precipitators (WESP), the Wet Electrostatic Scrubbers (WES), the Heterogeneous Condensation Scrubber (HCS) and the Bubble Towers (BT) emerged as reliable options. However, one fundamental drawback of all these water-based techniques is that proper washwater treatment units are required to minimize any liquid discharge and to assure they are harmless to human and marine environment.

The WESP (Environmental Protection Agency, 2003; Kim et al., 2012) consists in an ESP whose collecting walls are wet by a film of liquid that collect the particles once they impact with its surface. The WESP units were developed to treat gas streams up to 180.000 m^3/h, which are reliable for ship's engines application. The issues related to particle conductivity, stickiness and cohesiveness are not relevant for WESPs since the collectors are wetted by a liquid that remove the particles. However, the system is not able to solve those issues related to the insufficient level of ultrafine particle charging.

Wet electrostatic scrubbing (WES) allowed reducing the issues of a limited particle charging efficiency by using sprayed droplets as diffused particles collectors, in place of the ESP plates. A wet electrostatic scrubber is a spray tower equipped with an electrified spray unit for electrically charged water droplets generation and, optionally, a particle pre-charging unit. The electrostatic forces among droplets and particles lead to a more rapid and effective particle capture compared with conventional spray scrubbers. In addition, wet electrostatic scrubbers inherit all the structural and process advantages of conventional scrubbers, such as low-pressure drop, simple design and operation, as well as the ability to remove simultaneously soluble gases. After intense scientific study and tests at laboratory scale (Balachandran et al., 2003; Carotenuto et al., 2010a,b; D'Addio et al., 2014; D'Addio et al., 2013; Di Natale et al., 2013; Ha et al., 2010; Jaworek et al., 2006), application of an innovative wet electrostatic scrubber for diesel particle capture at pilot scale was recently developed and tested within the activity of the FP7 project DEECON. Trial tests on model flue gases (Di Natale et al., 2015) and with mono-cylinder diesel engine test rig (Antes et al., 2013; Jaworek et al., 2014) indicated that the WES unit removes particles as fine as 10 nm and reduces total particle concentration in the range 10–500 nm with efficiencies between 70% and 95% in number. Tests on an old 200 kW heavy duty diesel engine using a 2% sulphur fuel demonstrated an SO_2 reduction of 56% or more and PM number reduction of 90% or more during tests running the engine at 40% load, with a liquid to gas ratio of 1.2 l per cubic metre (Einemo, 2014). During these experiments, the particle size distribution in the submicron range spanned from 10 to 400 nm, with peak value at about 30 nm. When the same unit was tested as a conventional scrubber, particle removal efficiency resulted negligible for liquid-to-gas ratio as low as 10 L/m^3.

Recently, other wet processes for ultrafine particle capture were under development and examined at laboratory scale. One of these methods is the heterogeneous condensation scrubbing (HCS), which is based on the ability of particles to act as condensation nuclei for water vapour. By this way, soot particles are enclosed in water droplets making them easy collectable in a subsequent conventional scrubber (Fan et al., 2009; Tammaro et al., 2012; Yan et al., 2008). Bubble towers (BT) for nanometric particle capture were also investigated (Calvert and Parker, 1978; Charvet et al., 2011; Koch and Weber, 2012) and experimental results at laboratory scale revealed high removal efficiency for submicron particle. However, the system may present high pressure drops related to the high water holdup (up to 1 m) and a significant device footprint needed to optimize gas bubbling inside the column.

Final remarks

In the last 10 years, the number of scientific and newspaper articles concerning particulate emissions from ships rapidly increased, testifying the growing interest of scientific community and of society on this topic. With reference to the scientific articles, researches spread along three main directions:

- the assessment of emission factors from ships;
- the assessment of shipping influence on climate forcing;
- the impact of shipping on the air quality in urban areas.

This work reviewed the most recent literature pertaining particulate matter emissions from ships, analysing both the data related to the emission profiles and the different options proposed for the emission control. The vast majority of the available studies agreed in attributing to shipping a major role in producing particulate matter whose characteristics and quantities have significant impacts on the environment and the air quality in regions close to large harbours and in coastal areas of intense naval traffic.

As regards the ships emitted particles, the main findings can be summarized as follows:

1. Particle emissions from ships are formed by four major constituents: ashes, sulphates-related particles, elemental carbon (EC) and organic matter (OM). The emission factors can be estimated to be in the ranges: Ashes + sulphates = 138–185% S mg/kW h; EC = 10–100 mg/kW h; OM = 100–1000 mg/kW h. The number of total particles emitted is in the order of range 10^{15}–10^{16} particles/kW h. These emissions are comparable to those measured for heavy duty truck engines (Ban-Weiss et al., 2009) in terms of total emitted number and, at comparable sulphur fuel content, also in terms of mass emissions. However, the emission factors of modern coal power plant (Chen et al., 2014; Yi et al., 2008; Zhang et al., 2005) and cars are lower thanks to the introduction of proper after-treatment systems thanks to more restrictive environmental rules.
2. In spite of the emission factors, the lower amount of fuel consumption associated with ships (see Fig. 2) makes the contribution of shipping to the overall particulate matter emissions limited to 1–2% of global emissions. This low percentage indicates that the overall contribution of shipping to climate change is limited; however, the distribution of commercial routes concentrates most of these emissions in the areas with the highest population densities and several researches associated the presence of large ports with a significant contribution to air pollution in the hosting city. Although

sometimes these emissions represent a fraction of the emissions of PM from cars, the associated impact on population cannot be neglected. In this sense, it is worth noticing that Winijkul et al. (2015) and Yan et al. (2014) indicated that transportation accounts for about 5% of the total emitted particles, but about 50% of the particles in the ultrafine size range. Balkanski et al. (2010) found that shipping is responsible for small fractions (about 5%) of BC, but for relevant fractions of OM (about 25%) and for the majority (about 75%) of sulphates associated with worldwide emissions from road vehicles, ships and aviation.

3. Exposure to particulate matter was associated with severe diseases, and eventually death, upon chronic exposure. It also appeared that the particles emitted by ships are potentially more dangerous than those emitted by other fossil fuels due to the amount of transition metals and to the role of additives in the fuel. In addition, the role of lube oils is relevant in determining a higher toxicity potential of ship emitted particles compared with the particles emitted by coal or wood combustion. At the best of our knowledge, there are no studies on the epidemiological effects of ships exhaust exposure.

All these evidences suggest the necessity to introduce specific regulations to control emission of particulate matter from ships, similar to what happened in the past for the industrial and the automotive sectors.

To date, IMO guidelines and national flags regulations are focusing on the emissions of the coarse, by far less toxic, fraction of particulate matter, while the ultrafine particle is mainly still considered in relation to their impact of black carbon on climate.

The shipping industry is currently facing a tremendous pressure due to the introduction of regulations to control the emissions of sulphur dioxides and nitrogen oxides; therefore, the application of any new regulation should carefully take into account the technical and economic feasibility of emission control strategies. To this end, it is worth remembering that any strategy aimed to increase ship efficiency has beneficial effects on the overall emission of particulate matter, as for all the other pollutants. In this sense, the introduction of EEDI index will have a proportional benefit on PM emissions.

However, any other control strategy has to consider the different nature of particles. Sulphates and ashes are effectively reduced by the same strategies currently adopted to control sulphur dioxide emissions: the use of low sulphur fuels and the conventional scrubbers proved to be effective in reducing the emissions of particles with size as low as 500 nm. These effects were already forsaken in the Marpol Annex VI reg. 14, where the reduction of sulphur was expected to generate a reduction of PM emission by 85% in mass.

On the contrary, scientific evidences indicated that the soot fractions of EC and OM could not be reduced by changing the sulphur level in the diesel fuels, while only the LNG and potentially the WIFEs can lead to a relevant cut in the emissions of these kinds of particles. While the LNG emissions are well investigated, the actual effectiveness of the WIFEs in reducing PM emissions has to be still addressed in practice. The removal of EC and OM requires specific after-treatment systems to achieve significant reductions. Among them, those providing the highest potentialities are the diesel particulate filters, which are effective only in conjunction with fuels with sulphur content below 500 ppm, and the wet electrostatic scrubbers. These are ready at pilot scale and achieve very high removal efficiencies whatever the sulphur level is. At the same time, it must be considered that the introduction of any after-treatment unit, as the scrubbers or the DPF, introduces fuel penalties as a consequence of backpressure and of the additional costs of system auxiliaries (e.g., pumps, additional fans). These fuel penalties partially compensate for the benefits of the gas cleaning system, but only DPF and WES seem currently able to achieve removal efficiencies higher than 90%.

The review of available technologies suggests that, differently from the early years of the century, when the MARPOL Annex VI was introduced, scientific understating and technological development are now ready to properly support the introduction of specific regulations to reduce the emission of particulate matter at levels low enough to become harmless for population and the environment.

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