Assessing particle contributions of ship exhaust gases to the atmosphere in port cities (Review)

Abstract: The potential health and environmental implications associated with the exhaust gases of marine engines have led to significant recent research activities. Although knowledge of the characteristics of emissions from marine engines has improved considerably due to an ever-growing interest in the scientific community, there is still lack of the aggregate assessment to fully understand about aspects of the emissions from ships. This review summarizes the existing knowledge of emission from ships, highlights problems relating to particle emission such as particle emission factors, chemical compositions, particle distribution, factors affecting particle emissions, the contribution of ship emission to the environment, as well as the impact of ship emission on air quality, ecosystems, and human health. Additionally, this article also mentions emission regulations, study methods and evaluation of exhaust emissions and method reducing emission from the ship

Keywords: ship emission, environmental pollution, exhaust gas treatment.

Introduction

The ship is one of the friendly ecological means of transport, which have a low level of emissions per ton mile compared with aviation, rail and road transport [23]. For this reason, most of the cargo in the world is transported by ship (about 85%) [18]. Therefore, seaborne transportation plays a major role in global industries and society, enabling the transport of large volumes of raw and pro-
cessed material, as well as food, water and technological products [24]. Over the years, although experiencing a slightly slower growth rate compared to the long-term historical average, global water-borne trade is forecasted to rise to an excess of 15,000 mln tons in 2035 [24].

Like other means of transportation, the ship transportation always accompanies emissions. Exhaust gas from ships contains many toxic substances such as carbon dioxide (CO$_2$), carbon monoxide (CO), sulphur oxides (SO$_x$), nitrogen oxides (NO$_x$), volatile organic compounds (VOC), particulate matter (PM). These toxicity adversely impact human health and environment [45]. It is estimated that shipping-related PM2.5 emissions are responsible for approximately 60,000 premature cardiopulmonary and lung cancer deaths annually at a global scale [47]. Recent other studies have found that emissions from active vessels are primarily responsible for their impact on the climate and chemistry of the marine atmosphere [16].

Besides that, it has been confirmed that most of the ship emissions (approximately 90%) occur concentrated in urban areas along the coast within 45 km (24 nautical miles) from the coast and at least 70% of emissions from ships in international routes happen about 400 km of the coast [21]. However, these pollutants can spread out to hundreds of kilometers towards the mainland, which causes air quality problems even in very remote areas of the coastal zone. Emissions from ships affect air quality at large distances, and some pollutants have a worldwide dispersion [7]. Some studies indicated that shipping emissions accounted for the 9–15% of the total PM mass concentration at port areas [12].

**Study methods and evaluation of exhaust emissions from ship**

Although most of the previous studies focused on NO$_x$, SO, and PM emissions [47], recent publications have begun to mention nanoparticle emissions and their measurements [36]. The studies have carried out tests in the many different ways such as on engine test beds in laboratory conditions [34], on-board ship from the exhaust pipe [3], by taking measurements from ship plumes by aircraft [9] or by ship [37] and by performing stationary measurements in port areas [4]. In general, the ship emission research method includes following steps: 1) collecting, handling and preserving samples according to the relevant test methods, 2) sample analysis, 3) evaluate the sample based on the analyzed results.

Collecting samples should be conducted at locations which are determined by relevant regulations or by statutory instruments. If the sampling locations are not mentioned in the regulations or in instrument instructions, they will be selected to represent the total or the known portion of emission sources [44]. Monitoring site is chosen based on many parameters such as locality, terrain, meteorology, emission sources, possible chemical or physical interference, availability of services and security level [44].

Analyses should be conducted in an accredited laboratory by an independent accreditation body. In most cases, standard methods are referenced, with additional guidance or clarification given if needed. Where there is no suitable published method, a complete description of the approved method should be given [44].

Emissions factors for PM and trace gases (CO, SO, HCHO and NO) also may be measured using the plume intercept method (also known as the “sniffer” method, see Fig.1), wherein concentrations of the pollutants are measured in a plume that is intercepted downwind of the vessel of interest. One benefit of the plume intercept method is that atmospherically relevant dilution factors and temperatures are achieved by the time the plume is sampled, although the exact extent of dilution varies for each plume and the plume must be identifiable above background concentrations [10]. Whenever weight restrictions allow, multiple independent EMSUs are operated in parallel to increase the accuracy of measurements, ensure system redundancy, and secure additional evidence [14]. Basically, the results from individual studies may depend on the measurement methodology used i.e.plume intercept [10, 37], in-use stack sampling [2], or test-rig sampling [34].
Particle emission

The particle emissions depend on engine load and span over a wide range from 0.3 up to 10 g/kWh [31]. Early tests on particle emission from IFO and MDO fueled ships at berth reported that total particle emissions are from 0.48 to 0.67 g/kWh and from 0.14 to 0.48 g/kWh respectively [20]. Other study was carried out to investigate the emissions of 139 vessels on the banks of the river Elbe in Germany. Its results found that a total number of emitted particle was $2.55 \pm 1.91 \cdot 10^{16}$ particle/kg$_{fuel}$, and the mean emission of PM1 is about $2.4 \pm 1.8$ g/kg$_{fuel}$, in which BC took account for $0.15 \pm 0.17$ g/kg$_{fuel}$ [22].

Chemical composition and Morphology of PM

Particles emitted from ship have complex components consisted of sulphate, organic carbon (OC), elemental carbon (EC), and other inorganic compounds originating from the fuel and lubrication oil [37].

Organic carbon takes account of the highest percentage in PM component with about 45–65%, followed by sulphate (2–15%) and EC (1–20%) [21]. The OC emission is influenced by the fuel type and engine mode. The OC emission factor at berth and during maneuvering was 31% higher than those measured during cruising [30]. Elemental carbon emissions primarily depend on the engine type. The EC/PM mass ratio in exhaust gases of the auxiliary engines was 20%, while the ratio was 1–6% for main engines. The EC emission factor for the auxiliary engine was 6.7 times the average emission factor for the main engine. The EC emission factor tends to decrease with decreasing engine load and then significantly increase at 15% engine load [30].

The elemental compositions in PM also depend strongly on fuel type. Vanadium and Ni dominate emissions from burning HSF, and they have much lower concentrations in LSF emissions, which consist primarily of Si, Fe, Sn, Ba, Al, and Zn. During maneuvering, switching from HSF to LSF reduces the EFs of V and Ni by 85% and 80%, respectively. Calcium, Mg, Si are derived from the lubricating oil, and the higher concentration of these metals are found in the larger particle fractions, when engine load increases [51]. This phenomenon is explained by the fact that higher temperatures increase the consumption of lubrication oil [46].

Ionic components account for 8–27% in the PM emissions, and sulphate is the most abundant component. The sulphate fraction increases from 2.7% to 12.2% during switching from LSF to HSF. The sulphate emission factor of the ship using HSF during maneuvering was 3.4 times higher in comparison with the sulphate emission when using LSF. Other research indicated that the measured emission factors for sulphate particles such as SO$_2^{3-}$ are low, about 0.1–0.2 g/kg$_{fuel}$ for HFO with 1% sulphur, 0.07–0.09 g/kg$_{fuel}$ for HFO with 0.5% sulphur and 0.003–0.006 g/kg$_{fuel}$ for MGO. This corresponds to 0.1–0.8% and 0.1–0.6% of fuel S converted to PM sulphate for HFO and MGO, respectively [42]. Beyond sulphate, the ionic content largely consists of Ca$^{2+}$, NH$^{4+}$, NO$_2^{-}$, NO$_3^{-}$, and PO$_4^{3-}$, which account for 3.1%, 2.2, 1.3, 1.3%, and 0.9% of mass, respectively [30].
The PAHs are predominantly present in an alkylated form, and the composition of the aromatic OM in emissions clearly resembles that of fuel. Burning HFO emits significantly more species known to be hazardous to health (PAH, Oxy-PAH, N-PAH, transition metals) in comparison with burning LFO at all operating modes of engine [22].

A study showed that the average PM2.5 emission factor from ship using HFO and LFO is 520 mg/kWh, and 120 mg/kWh, respectively. The average emission factors for the major species of the particulate fraction in the exhaust gas are 320 mg/kWh, 86 mg/kWh, 57 mg/kWh, and 44 mg/kWh for OM, SO$_4^{2-}$, BC and inorganic elements, respectively. The main components of the inorganic elements are S (44%), V (18%), and Fe (12%) [43].

Size distribution

The particle size distribution depends on engines and operating mode (i.e. load) [25]. Some studies showed that the particle size distribution in exhaust gas of marine diesel engines is similar to that of other diesel engines used on land [29]. This distribution is bimodal with a large peak at around 12 nm (nucleation mode) and a smaller peak at 30–40 nm (soot/accumulation mode) from on-board measurements [29], but measurements on a test-bed engine showed a major peak at 15 nm and a minor peak at 50 nm [46]. By contrast, other research indicated that the particle size distributions showed only one mode for different operating conditions of the ship with a peak at around 40–50 nm [29, 34]. However, some studies found that particle size distributions for HFO at lower loads usually show a bimodal distribution with a significant amount of particles in the nucleation mode [29]. At higher loads the distribution is monomodal [34] and centered around 50 nm. This has been observed both for test-bed measurements [35] as well as for onboard measurements [51].

Generally, the particles in exhaust gas of marine diesel engines have smaller size (mean diameter of 20–40 nm) compared to particles emitted by automobile diesel vehicles (~60 nm) [40], which mainly emit particles in accumulation mode [34]. The size differences are associated with the differences in fuel sulphur content between marine fuel oils and fuels for automobile diesel vehicles.

Furthermore, the mass distribution also has a bimodal character [29], with one peak in accumulation mode and one in coarse mode [39, 42]. Previous measurements have observed peaks at 0.5 µm and 7 µm [42], and in other measurement indicated one peak at 60–90 nm and the second peak at 7–10 µm [39].

Contribution of ship emission to environment

Many studies indicated that emission from ships contribute to environmental pollution not only of harbours but also of inland areas [5]. The concentrations of PM2.5 could reach up to 2.9–45.0% due to ship emissions [12]. This contribution fluctuates in wide range, which is explained by following two reasons.

Firstly, it is geographical distance of experimental areas. This contribution is larger in areas nearby coastline (Fig. 2) [12]. As shown in figure 4.1, the contribution of ship emissions is the highest at distance 10 km (10.7%), with a value of 11.3 times as high as a value at distance 200 km. In other study [49] also indicated the similar trend with PM2.5 contribution at port and in city center of the port city of Thessalonki about 13.4% and 9.4%, respectively. The result of this study showed that the high percentages of Ni and V, along with the significant shares of SO$_4^{2-}$ and EC in PM 2.5 [49]. However, a reverse trend was seen in contribution of secondary sulphate and organic particles, whose main components are SO$_4^{2-}$, NH$_4^+$ and OC. The secondary particle contribution is 13.7% at the port and 34% in the city central [49]. This phenomenon is explained by the fact that secondary sulphate is typically associated with long range transport events, as it is considered as an ingredient of aged air masses, because the oxidation of SO$_2$ to SO$_4^{2-}$ is slow and thus this aerosol component is more related to transportation than local pollution [49].
Secondly, contributions of ship emissions considerably vary according to seasons [12]. This change can be caused by the change of ship emissions as well as meteorological conditions (e.g. wind speed, wind direction, temperature, and humidity) in different seasons. For example, the study conducted in Bohai Rim Region of China showed that the proportion of ship emissions among the total anthropogenic sources is the highest in autumn (0.1–7.8%), followed by spring (0.1–6.8%), and summer (0.1–5.6%), while in the winter it is lowest (0.1–4.9%) [12]. Meteorological conditions also play a non-negligible role. Statistical analysis of meteorological data indicates that the frequency of wind directions significantly affect PM2.5 dispersion. In addition, a relatively higher temperature and humidity could facilitate the transformation of precursor gases (e.g., SO$_2$, NO$_x$, and VOCs) emitted from ships to secondary particle (e.g. SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and secondary organic aerosol), leading to the increase of PM2.5 concentrations [12].

**Impact on environment**

The particles, especially sulphate particles affect significantly cloud formation since they act as water condensation nuclei [23]. Furthermore, fine particles may have a greater potential for adverse environmental impacts, because of some following reasons. Firstly, fine particles have a residence time in the atmosphere up to days and even weeks, so local emissions can become a regional issue and even global concern [15]. Secondly, these particles can affect the air quality in other countries through boundary transport and have global climate change implications [27]. Finally, fine particles such as PM2.5 are considered as a primary cause for the scattering of visible light, the degradation of visibility, the formation and evolution of haze [33].

Furthermore, the particles have direct and indirect effects on radiative forcing (RF), with different impacts on the climate. The net effect of emissions from ship operations on the RF estimates is considered to be negative (-0.408 W.m$^{-2}$) due to a high content of sulphate particles in the emissions compared to other combustion sources and the significant indirect effects of particles emitted by operating ships [32]. Besides that, black carbon deposits on snow and ice will reduce the albedo of the surface, which enhances the melting of snow and ice [32]. This is particular interest for the Arctic when BC is expected to increase due to an increase in ship operations, anthropogenic activity in this area, and the amount of BC transported to the Arctic from other areas [19].

**Impact on human health**

Some studies show that maritime emissions have less health and environmental impacts compared to the land-based source since they are released sometimes far away from populated areas or sensitive ecosystems. However, in harbour cities, ship emissions are in many cases a dominant source of urban pollution and need to be addressed when compliance air quality limit values [13].

Epidemiological studies also show a consistently link between the ambient concentrations of NO$_x$, SO$_x$, and PM with a variety of adverse public health outcomes, including increased risk of premature death from heart and pulmonary diseases and worsened respiratory disease [41]. Even in low doses, chronic exposure to soot particles causes several respiratory pathologies, such as bronchitis, pneumonia, tracheitis, and asthma [38] as well as to the occurrence of disruption of vascular
functions [48], ischaemic heart diseases and neurological effects at some brain regions [48]. Especially, the presence of several transition metals such as V, Pb and Ni in heavy fuel oil makes the marine diesel particles potentially more harmful than those produced by other sources, such as gasoline or NG [30].

Recent studies [17] indicate that shipping-related PM emissions are responsible for around 60,000 cardiopulmonary and lung cancer deaths annually, with most deaths occurring near coastlines in Europe, East Asia, and South Asia. Based on previous estimates of global PM2.5-related mortalities [14], it is estimated that 3–8% of these mortalities are attributable to marine shipping [17]. Another study by the California Air Resources Board (CARB) estimated that diesel PM emissions from the Port of Los Angeles and Long Beach increase the cancer risk for 60% of the neighboring population [26]. And diesel PM emissions are considered to be responsible for 14–43 additional premature deaths as well as 180–1,300 additional asthma attacks from these ports each year [26].

**Emission Regulation**

For marine engines, to restrict the negative impacts of emissions on the environment, IMO has set emissions standards for ships (including NOx content in engine exhaust and sulphur content in fuel), and technical measures to minimize emissions. These regulations are shown in the regulations in the annexes of MARPOL 73/78. SOx emissions are limited through the fuel sulphur content (FSC), which was regulated by Regulation 14 in MARPOL 73/78 Annex VI. The title of Regulation 14 is ‘Sulphur oxides (SOx) and PM’. Unluckily, the regulation does not mention any limits on particle emission, instead they are indirectly regulated by limits imposed on the FSC. Although there are no international regulations that directly control or limit the particle emission from ocean-going ships, it is believed that an increased use of fuel with low sulphur content (LSC) will reduce the PM2.5 emissions by 80% from 2000 to 2020 [1].

In Russia, emission standards were introduced in 1980–1981. Over the past 30 years, these standards have been revised several times and significantly changed. As a rule, the changes were related to the list of monitored parameters, from the values and methods of testing and calculation. Up to now, the main national documents limiting harmful emissions from ship, diesel and stationary internal combustion engines are:

- GOST R 51250-99. Marine, locomotive and industrial diesel engines. Exhaust smoke opacity Standard values and testing methods.
- GOST R 51249-99. Internal combustion reciprocating engines. Emissions of harmful substances with the exhaust gases. Limit values and test methods.

There is a tendency of approximation of the domestic regulatory framework to the standards of the international organization for standardization (ISO). National GOST ISO 8178 has already been developed, which is identical to the international standard ISO 8178, developed by ISO / TC technical committee.

**NOx Reduction**

Humid Air Motors(HAM) prevents NOx-formation during combustion by adding water vapors to the combustion air. The method is able to reduce NOx by 70–85% [8].

Selective catalytic reduction (SCR) is used to convert NOx molecules back to harmless O2 and N2 molecules by urea with conversion efficiency about 90% at temperatures above 300ºC [28]. This system operates reliably in the temperature range between 250 and 400ºC with low-sulphur fuel oil [49]. Nowadays, there are already around 500 ships fitted with SCR. However, a drawback of SCR technology is the risk of ammonia appearing in the exhaust system when the engine is working in variable loads [49].

Using Liquefied natural gas (LNG) as an alternative fuel is a way to reduce effectively NOx, SO2 emissions because this fuel contains no sulphur. NOx and PM emissions reduce significantly by 80 per cent and even more.
SOx Reduction

*Low-sulphur fuel:* Sea-going ships usually use high sulphur fuels with average content about 2.5–3 per cent, which is higher 3,000 times than that in road fuel in Europe. Therefore, the simplest way of reducing SO2 emissions is to switch to low sulphur fuel [8].

*Scrubbers:* Using scrubber is an effective method to reduce SO2 and PM emissions with removing efficiency up to 99% and 88%, respectively. Most marine scrubbers have a venturi block before an SO2 absorption column. The Venturi scrubber promotes hydrodynamic interaction between particles and sprayed droplets, which increases the removal efficiency of particles larger than 1 micron. Special cyclonic scrubber designs developed by Clean marine allow to reduce the larger soot particles with size as low as 500 nm [11].

PM Reduction

Nowadays, instruments, which include Diesel Particulate Filter (DPF), Diesel Oxidation Catalyst (DOC) and Continuous Regeneration Trap (CRT), are popularly used to remove particles, CO and HCs from exhaust gas. However, they only operate effectively with low sulphur fuels, so they are popularly used for road vehicles. They will have a bright future when the heavy diesel oil standard is more strict [28].

Conclusion

As it has been shown in this review, ships are significant particle emitting sources, which adversely impact the environment and human health in coastal areas, especially in harbour regions with particle contribution up to 45%. Chemical components, morphology, and emission factor of particles depend strongly on fuel types, lubrication oil, operating modes. Besides, impact levels of ship emissions on coastal regions can vary widely with many different factors, such as the geographic location, meteorological conditions, and ship density etc. Therefore, each coastal country, as well as harbour region, should conduct specific studies to accurately assess the impact of ship emission, which helps authorities plan appropriate economic development policies as well as find effective solution to reduce toxicity and negative impact of exhaust gas from ships.

Nowadays, there are major three groups of method popularly being applied to reducing pollutant emissions from ships: 1) using exhaust treatment systems such as SCR or scrubber; 2) using low sulphur fuels or alternative fuel such as LNG etc... in combination with the use of lubricating oils containing few toxic additives (toxic substances and heavy metals etc); 3) improving and optimizing engine operation to reduce fuel and lubrication oil consumption. In general, these methods are combined together to comply with emission regulation of MARPOL 73/78.

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Оценка вклада частиц выхлопных газов от судовых двигателей в атмосферу в портовых городах (обзор)

Аннотация: Возможные последствия для здоровья и окружающей среды, связанные с выхлопными газами судовых двигателей, вызвали значительную исследовательскую деятельность в последнее время. Хотя знания о характеристиках выбросов, производимых морскими двигателями, значительно улучшились в связи с постоянно растущим интересом к ним со стороны научного сообщества, по-прежнему отсутствует агрегированная оценка, позволяющая в полной мере понять аспекты выбросов с судов. В настоящем обзоре обобщаются существующие знания о выбросах с судов, освещаются проблемы, связанные с выбросами частиц, такие, как факторы выбросов частиц, химический состав, распределение частиц, факторы, влияющие на выбросы частиц, вклад выбросов судов в окружающую среду, а также влияние выбросов судов на качество воздуха, экосистемы и здоровье человека. Кроме того, в этой статье также упоминаются правила выбросов, методы исследования и оценки выбросов выхлопных газов и метод сокращения выбросов с судна.

Ключевые слова: выброс судов, загрязнение окружающей среды, обработка выхлопных газов.