Aerosol emissions from a marine diesel engine running on different fuels and effects of exhaust gas cleaning measures

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Abstract

The emissions of marine diesel engines have gained both global and regional attentions because of their impact on human health and climate change. To reduce ship emissions, the International Maritime Organization capped the fuel sulfur content of marine fuels. Consequently, either low-sulfur fuels or additional exhaust gas cleaning devices for the reduction in sulfur dioxide (SO_2) emissions became mandatory. Although a wet scrubber reduces the amount of SO_2 significantly, there is still a need to consider the reduction in particle emissions directly.

We present data on the particle removal efficiency of a scrubber regarding particle number and mass concentration with different marine fuel types, marine gas oil, and two heavy fuel oils (HFOs). An open-loop sulfur scrubber was installed in the exhaust line of a marine diesel test engine. Fine particulate matter was comprehensively characterized in terms of its physical and chemical properties. The wet scrubber led up to a 40% reduction in particle number, whereas a reduction in particle mass emissions was not generally determined. We observed a shift in the size distribution by the scrubber to larger particle diameters when the engine was operated on conventional HFOs.

The reduction in particle number concentrations and shift in particle size were caused by the coagulation of soot particles and formation/growing of sulfur-containing particles. Combining the scrubber with a wet electrostatic precipitator as an additional abatement system showed a reduction in particle number and mass emission factors by >98%.

Therefore, the application of a wet scrubber for the after-treatment of marine fuel oil combustion will reduce SO_2 emissions, but it does not substantially affect the number and mass concentration of respirable particulate matters. To reduce particle emission, the scrubber should be combined with additional abatement systems.

Keywords

Abatement system; Marine fuel types; Particulate matter (PM) reduction; Ship emission; SO_2 reduction; Wet sulfur Scrubber

Abbreviations

CMD, count median diameter; DF, dilution factor; eBC, equivalent black carbon; EC, elemental carbon; EF, emission factor; FID, flame ionization detector; FSC, fuel sulfur content; FTIR, Fourier transform infrared spectroscopy; HFO, heavy fuel oil; HIA, health impact assessment; IMO, International Maritime Organization; MCR, maximum continuous rate; MGO, marine gas oil; OC, organic carbon; PAHs, polycyclic aromatic hydrocarbons; PM, particulate matter; PSU, practical salinity unit; SCR, selective catalytic reduction; SECA, sulfur emission control area; SMPS, scanning mobility particle sizer; TEOM, tapered element oscillating microbalance; UV, ultraviolet; WESP, wet electrostatic precipitator

1. Introduction

Maritime transport logistics play an essential role in the international freight system. In 2015, more than 80% of worldwide transport supply was conducted by shipping (UNCTAD, 2017). The increasing demand for shipping conveyance and lack of tight regulations regarding shipping emission are the main contributors to air pollution (Blasco et al., 2014). According to a recently conducted health impact assessment (HIA), on average, 5.5 premature deaths per year for every 100,000 inhabitants in Mediterranean cities are attributed to shipping emissions (Viana et al., 2020). As a result, ship emissions and their deposition from the atmosphere into marine sectors are defined as hazardous and polluting substances in the Marine Strategy Framework Directive 2008/56/EC of the European Parliament and the Council (European Parliament and the Council of the European Union, 2008). One of the ship emission's pollutants is particulate matter (PM), which refers to diverse substances existing as liquid and/or solid matter (EPA, 2008). Together with PM, nitrogen oxides (NO_x) and sulfur oxides (SO_x) play a role in the environment. Direct and indirect effects of SO_x and NO_x emissions include acidification of water and soil, changes in the atmospheric radiation budget, the formation for secondary aerosols in the atmosphere (Fuglestvedt et al., 2009; Winnes and Fridell, 2009). Therefore, the International Maritime Organization (IMO) adopted specific guidelines in Regulations 13 and 14 of Annex VI of the International Convention for the Prevention of Pollution from Ships (MARPOL). In 2005, the IMO widened its measures by introducing sulfur emission control areas (SECAs) and defining the maximum fuel sulfur content (FSC), which is currently 0.1% m/m inside and 0.5% m/m outside of SECAs (IMO, 2008). Consequently, conventional low-grade HFOs have been replaced with high-quality distillate fuels or low-sulfur heavy fuel oils. This leads to a lower number and mass of sulfur and particle emissions (Kuittinen et al., 2021; Moldanova et al., 2013). Alternatively, conventional low-grade HFOs with a high sulfur content can be used in combination with an abatement system, such as a wet scrubber, to reduce the SO_2 emissions in exhaust gas.

Therefore, ship owners must decide either to use fuels with a low-sulfur content or to install an abatement system by considering the navigating routes and capital investment (Carr and Corbett, 2015; Le et al., 2021). However, despite a possibly reasonable use from an economic perspective, the environmental benefit of open-loop wet scrubbers is questionable as air pollutants are locally concentrated in the hydrosphere at the air–sea interface (Endres et al., 2018; Turner et al., 2017). Consequently, the discharge of polluted water from the scrubber system is prohibited in vulnerable areas to pollution such as ports and estuaries (Hermansson et al., 2021).

In recent years, several studies have proven that a wet scrubber can reduce the SO_2 content of exhaust gas by 90%–99% to meet the IMO regulations. In addition to SO_2 reduction, these studies have led to the potential reduction in gaseous and particulate exhaust emissions by wet scrubbers (Karjalainen et al., 2022; Lehtoranta et al., 2019; Yang et al., 2021). Although these studies agree well on SO_2 reduction, the results associated with particle emissions show wide variations of reduction, which are highly dependent on the geometry of the scrubber and the used technology. Furthermore, although all scrubbers provide relatively high humidity and a low-temperature condition, several factors such as sampling methods, fuel types, particle size distribution, and the specifications of the engine play an essential role in the removal of the particle phase (Fridell and Salo, 2016; Ritchie et al., 2005; Yang et al., 2021). Among others, Fridell and Salo (2016) observed a reduction of 75% of the total particulate mass emission and 92% of the total particle number concentration by the scrubber when using an HFO with 2.3% m/m of sulfur. Compared to these results, a test ship engine experiment by Zhou demonstrated different removal efficiencies of particle mass

from 5% to 50% using a scrubber, depending on the particle size of interest (Zhou et al., 2017). Similar results were reported by an onboard experiment, where a scrubber removed only 10% of particle mass of $PM_{2.5}$ when running an HFO with 1.89% m/m sulfur (Yang et al., 2021). In addition, Lehtoranta et al. (2019) found that particle number concentration was not affected by a scrubber, concluding that submicron particles are not effectively removed. Hence, there is still a need to understand particle removal using marine scrubbers based on physicochemical characterization.

Therefore, a wet scrubber in pilot plant scale was connected to a common rail research ship engine, and PM and gaseous emissions were analyzed as untreated raw gas after exhaust gas cleaning. Fuel types were specifically selected in consideration of currently existing regulations to investigate emission profiles depending on fuel properties. Moreover, the present study provides evidence on how the composition of fuel types changes the physical and chemical properties of particle emission with respect to the properties of the wet scrubber. As a proof-of-concept experiment, we conducted a further exhaust gas cleaning step by applying an additional wet electrostatic precipitator (WESP) after the scrubber to investigate its potential to reduce environmental and health-related burden.

2. Materials and Methods

2.1 Engine description and fuel properties

In the experiments, a single-cylinder four-stroke research and development engine of the Institute of Piston Machines and Internal Combustion Engines at the University of Rostock, Germany, was used. The research engine has a large displacement of 3.18 L, has a rated power of 80 kW at 1500 rpm, and is capable of running on various fuels from the distillate, paraffinic to high sulfur and highly viscous marine fuels. It is equipped with a common rail injection system and adjustable external charge air compression.

The engine was operated at 25% (20 kW) and 75% (60 kW) of the maximum continuous rate (MCR) according to the E2 test cycle for heavy duty, constant speed engines for marine propulsion based on ISO 8178-4. The 75% engine load stands for the typical optimum fuel oil consumption point. Therefore, this engine load was considered representative for cruising ships at open-sea operation or for stationary power generation operation (Woud and Stapersma, 2002). In addition, the engine was operated with 25% of the MCR, which represents a typical maneuvering operation at ports and harbors. Previous studies have demonstrated detailed information on the engine and have characterized the emission with different fuel types (Mueller et al., 2015; Sippula et al., 2014; Streibel et al., 2017). However, we emphasize that the engine parameters during our campaign had to be set differently from those from the previous studies.

In this study, we used fuels with different sulfur contents, MGO, and two different HFOs (HFO A and HFO B). MGO is a distillate fuel with a low-sulfur content and can be used without a scrubber on the open sea as well as within the SECA. Here MGO represents the reference compliant fuel for the current sulfur legislation. HFO A and HFO B are noncompliant high-sulfur fuels with sulfur contents of 1.02% and 2.16% m/m, respectively. As typical residual fuel oils (classified as RMK 380), both HFOs are only compliant when a scrubber is installed. A detailed physicochemical characterization of the three fuels was conducted according to ISO 8217, and it is presented in Table 1.

2.2 Sampling set-up and sample dilution

The sampling system included numerous instruments, but their analyses are out of the scope of this article. A simplified scheme of the set-up system is shown in Fig. 1 (for detailed information, see Fig. S1).

Two sampling points at the stack were used for the scrubber up- and downstream comparison consisting of two identical perforated sampling probes and two identical heated transfer lines (Fig. 1). Heat-resistant ball valves and an isolated Y-connector were used to switch the sampling points between up- and downstream of the scrubber for both particulate and gaseous phase sampling.

In the case of the scrubber upstream measurements, the heated transfer line, precyclone used to remove coarse particles larger than 10 µm, ball valve, and dilution air were heated to 200°C to minimize losses of particles. This high-temperature setting was able to avoid condensation and to keep the condition of the sampling lines above the dew point of sulfuric acid. The same type of ball valve was used for gaseous phase sampling, and the temperatures of the two heated transfer lines were set to 250°C and 180°C, respectively, for both up- and downstream measurements. The primary dilution factor (DF) from the main engine stack was adjusted depending on the fuel types using a two-stage ejector diluter system (eDiluter, Dekati Ltd., Finland). It was set to a factor of 25 for MGO and between 50 and 100 for the HFOs. After the primary dilution system, the sample stream was split into the sampling line of the filter sampling system and the online measurement instruments. To accomplish the desired concentration range of particle number and mass concentration related to the online measurement instruments, the sample stream was diluted further by two ejector diluters (Palas, Germany), each with constant 1:10 dilution.

The same sampling system was used for the emissions after the scrubber. To minimize the condensation of water vapor in the sampling line as well as to mimic the emission of the scrubber as a preconditioner for a WESP, the samples from the scrubber were taken through a heated transfer line with a temperature of 60°C. Emissions from the scrubber downstream and WESP were sampled at the same position, bypassing the WESP for the scrubber downstream measurement.

2.3 Gas-phase sampling

To avoid possible interferences with particle sampling, the gaseous compounds of exhaust gas were sampled directly after each particle sampling point. The sampled exhaust gas was brought to a heated filter, and Fourier transform infrared spectroscopy (FTIR, DX4000 gas analyzer, Gasmet Technologies Oy, Vantaa, Finland) was performed with 3 L/min of inlet flow. The software Calcmet (Gasmet, Vantaa, Finland) was used for spectrum analysis, including the blank subtraction to acquire the absorbance of sampled gaseous compounds and to correct the baseline absorbance.

2.4 Online particle-phase sampling

Comprehensive analyses of particle number and mass concentration were performed using different instruments such as a scanning mobility particle sizer (SMPS 3082, TSI, USA), a condensation particle counter (CPC3750, TSI, USA), a tapered element oscillating microbalance (TEOM 1400a, Thermo Fisher Scientific, USA), and an aethalometer (AE33, Magee Scientific, Aerosol, d.o.o., Slovenia).

SMPS has been used widely for the online measurement of the physical characteristics of aerosols from ship engines (Kasper et al., 2007). An X-ray neutralizer was built in, and the aerosol flow was set to 0.3 L/min. A TEOM was set for real-time monitoring of particle mass concentration (Patashnick and Rupprecht, 1991). In our study, the standard operation was set to a sample flow rate of 3 L/min and a temperature of the filter as well as sampling inlet tube of 50°C. In addition, the light absorption properties of ship engine exhaust aerosols were measured online using an aethalometer that determines the equivalent mass concentration of black carbon (eBC) at the wavelength of 880 nm.

2.5 Coagulation model

The contribution of a pure physical coagulation process in the scrubber was investigated by calculating the average coagulation coefficient ($\underline{K}$) in Equation 1 and by applying it in the coagulation model in Equation 2 (Hinds, 2011):

$$\underline{K} = \frac{2kT}{3\eta}\left[1 + \exp\left(ln^2\sigma_g\right) + \left(\frac{2.49\lambda}{CMD}\right) \times \left[\exp\left(0.5ln^2\sigma_g\right) + \exp(2.5ln^2\sigma_g)\right]\right], 1$$

where η is gas viscosity in Pa s, T is temperature in Kelvin, and k is the Boltzmann's constant (1.38×10^{-16} dyne cm/K). In addition, the count median diameter (CMD), geometric standard deviation (σ_g), and particle mean free path (λ) were considered for the polydisperse coagulation process (Lee and Chen, 1984). On the basis of the dimension of the scrubber and the volume flow of the exhaust gas from the motor, residence times (t) of 4 and 8 s and a geometric standard deviation (σ_g) of 2 for all fuel types were set in the model for engine loads of 60 and 20 kW, except that the size distribution of HFO B at 60 kW had a geometric standard deviation of 1. Here, we assumed that the particle-to-particle coagulation process takes place in an empty and dry scrubber at normal temperature and pressure, which is one of the main interparticle phenomena for particles with a diameter smaller than 1 µm (Walter, 2011).

$$N(t) = \frac{N_0}{1 + N_0\underline{K}t}, 2$$

where $N(t)$ is the number concentration at residential time t and N_0 is the initial number concentration in particles/m^3 measured by SMPS. To calculate the total particle number concentration and count median diameter after the coagulation processes in the scrubber, the monodisperse coagulation model was extended by considering the CMD, the geometric standard deviation (σ_g), and the average coagulation coefficient ($\underline{K}$).

2.6 Particle sampling on filters and chemical analysis

Particles were collected on 47-mm diameter quartz fiber filters (QMA 1851-047, Whatman, USA), after passing a 2.5-µm preimpactor (Fig. 1). The sampling flow and time of the filter sampling system were set to 10 L/min and 20 min for each condition, respectively. Immediately after the collection, the filters were stored at −20°C. Each filter was extracted three times with 2 mL of deionized water in an ultrasonic bath for 5 min, which resulted in a total extraction volume of 6 mL per filter sample. Extracts were filtered to remove particles, and for the determination of anions, ion chromatography was carried out according to DIN EN ISO 10304. It has to be mentioned that the used methodology (sampling and analysis) cannot distinguish sulfate from sulfite. Therefore, the results were reported as sulfate. In addition, thermal-optical carbon analysis (Desert Research Institute Model 2001A, Reno, NV, USA) was conducted to determine the concentration of elemental carbon (EC) in $PM_{2.5}$ at the 60 kW engine load using the Improve A protocol (Chow et al., 2007).

2.7 Open-loop scrubber

A downscaled research open-loop wet scrubber (EGCS30HB, SAACKE, Germany) was specially adapted to the 80 kW research ship engine of the University of Rostock. The scrubber was operated with Baltic Sea water with salinity of 10.5 ± 3.6 in practical salinity unit (PSU) and pH of approximately 8. The wet scrubber consisted of a quench with three sprays; wash tower with a total volume of 0.9 m^3, including a spray; filling body of a randomly structured package; and demister structure. During operation, the ratio of SO_2 to carbon dioxide (CO_2) of exhaust gas was held to a maximum of 4.3 SO_2 (ppm)/CO_2 (% v/v), which corresponds to the emission of fuel oil with 0.1% m/m of sulfur content (MEPC 259(58), 2015). Before the

exhaust gas was introduced into the wash tower, the exhaust gas temperature was reduced by adjusting the sprays inside the quench to optimize the solubility of SO_x in wash water (Bandyopadhyaya and Biswasa, 2006). The cooled exhaust gas was further guided into the wash tower equipped with a filling body package of metal saddle rings to offer a better mass transfer of SO_x from gas to liquid phase as well as to remove particles in the exhaust gas.

2.8 Wet electrostatic precipitator (WESP)

In addition to a scrubber, a second exhaust gas cleaning stage, a WESP (CAROLA, RVT Process Equipment GmbH, Germany), was connected after the scrubber and tested to remove particles further. A WESP is widely used for controlling particle emissions because of the high removal efficiency of particles with humid gases or sticky particles, especially in the nanometer range (Di Natale and Carotenuto, 2015). The CAROLA WESP consists of two sections, namely, an ionizing section and a grounded collection section. Briefly, the particle-laden gas is vertically guided to the ionizing section, in which the particles are charged by a corona discharge. Then, they are moved toward the collection section with an electric drift velocity proportional to the acquired charge (Bologa et al., 2009). For the cleaning of the electrodes in the ionizing section, dry air (30 m^3/h) is guided additionally into WESP, which dilutes the exhaust gas from the scrubber (<10% v/v). Because the cooled and humid gas from the scrubber outlet leads to the optimal working conditions of the WESP, the scrubber is an ideal exhaust gas preconditioner.

3. Results and Discussion

We investigated the emission of SO_2; NO_x as NO_2; PM in terms of particle number concentration, particle size distribution, and mass concentration of particles; eBC; and sulfate for different fuel types. The results are shown as emission factors (EFs), given as mass or number emissions per engine work output in g/kWh or mg/kWh and 1/kWh, respectively (Table 2).

3.1 Ship emissions by changing fuel type and using a sulfur scrubber

In Table 2, the EFs of SO_2 and NO_x as NO_2 are presented. High SO_2 EFs in untreated exhaust were observed from the high sulfur-containing HFOs, and the low amount of sulfur in the emissions from MGO was below the limit of quantification for the instrumentation used, which corresponded to <0.15 mg/kWh for 20 and 60 kW engine loads. The highest NO_x emission factor was observed using HFO A, and MGO and HFO B had similar NO_x EFs. According to the NO_x limits in MARPOL Annex VI, the NO_x emission factor should be below 10.19 g/kWh, which was not completely fulfilled in this case (Tier II and n = 1500 rpm). To comply with the IMO NO_x regulation, an after-treatment system, such as selective catalytic reduction (SCR), can be used in addition to a sulfur scrubber (MEPC 251(66), 2015). For both engine loads, the EFs of NO_x remained almost unchanged, and the SO_2 EFs were significantly reduced by the use of a wet scrubber. This significant reduction in SO_2 emissions using a sulfur scrubber is in line with previous studies (Fridell and Salo, 2016; Yang et al., 2021; Zhou et al., 2017). In addition to the gas emission, the total mass emission factor of particles is presented in Table 2. MGO demonstrated approximately 60 and 90 mg/kWh of total particle mass emission factor for 60 and 20 kW, respectively, and HFO A and HFO B showed 275 and 201 mg/kWh as well as 1960 and 1224 mg/kWh for 60 and 20 kW, respectively. The results are comparable to particle mass EFs measured for diesel oil and HFO with 1.3% m/m by Mueller et al. (2015), although the authors demonstrated the averaged emission factor from four different engine loads. The switch of fuel types from HFO to MGO resulted in at least 80% lower particle mass EFs in both operation modes (Table 2). This reduction is in agreement with previously reported studies showing a correlation

between FSC and the particle mass emission factor (Winnes and Fridell, 2009; Winnes et al., 2020). However, we also observed that the particle mass emission factor of HFO B with a higher FSC was lower than that of HFO A at a 60 kW engine load, indicating that a lower FSC does not necessarily imply lower particle emissions. To understand the composition of the particles, their sulfate fraction was measured by ion chromatography. The sulfate mass emission factor at the scrubber upstream takes up to 18% of the total mass emission factor HFO B at 60 kW. In addition to sulfate, the eBC fraction of the particles was investigated. The change in fuel type from HFO to MGO did not cause a reduction in eBC mass emission factor in the open-sea operation mode, although it was remarkably reduced in the maneuvering mode. Although MGO contained no detectable sulfur and therefore emitted less sulfate than HFOs, its eBC fraction was approximately half of the total mass emission factor of inhalable particles ($PM_{2.5}$) at a 60 kW engine load. A relatively high eBC fraction of the MGO PM was also reported during the combustion of MGO and distillate fuel (Moldanova et al., 2013; Mueller et al., 2015), and the formation of eBC is related to various parameters such as the chemical composition of the fuels and the combustion process (Lack et al., 2009; Sippula et al., 2014). A higher eBC mass emission factor of HFO A was also observed at an engine load of 20 kW, whereas the total particle mass emission factor and eBC emission factor of HFO B were more than 10-fold higher than that of MGO in this case (Table 3). In addition to eBC, the EC mass emission factor is given for a 60 kW engine load. The observed trend of the EC mass emission factor is consistent with the mass emission factor of eBC, although the measurement principles are fundamentally different. EC analysis was conducted from two filter samples at each condition, so only the maximum and minimum of the mass emission factor were given.

Downstream of the scrubber, we found reduced EFs for particle mass and eBC, and the sulfate mass emission factor was considerably increased for both HFOs (Table 2), which is contrary to the results of several other studies (Fridell and Salo, 2016; Winnes and Fridell, 2009; Winnes et al., 2020). Although the scrubber has the theoretical potential to remove particles, a significant reduction in the particle mass emission factor by a scrubber was not observed in this study. Because the particle mass is mainly determined by particles with diameters larger than 100 nm, i.e., accumulation and coarse mode particles (Walter, 2011), the measured particle mass EFs can vary strongly depending on each experiment's sampling method and the targeted particle size range. For inhalable particles ($PM_{2.5}$), which were investigated in this study, the general particle mass removal efficiency was reported to be as low as 5%, and it increased up to 35% under the same engine condition if the particle size of interest changed to PM_{10} (Zhou et al., 2017). In agreement with our results, Yang and colleagues observed that the $PM_{2.5}$ particle mass emission factor remains similar both up- and downstream of a scrubber at different engine loads (Yang et al., 2021).
Sulfate mass EFs downstream of the scrubber correlated with their FSC (Table 2). The relative sulfate fraction is remarkably increased by the scrubber up to ca. 15% and 50% of the total particle mass EF of HFO A and HFO B at a 60 kW engine load, respectively. A slight increase in sulfate fraction was also observed at a 20 kW engine load. The high mass EFs of sulfate on the filter can be attributed to the formation of the SO_2 exhaust gas, which is dissolved in the seawater in the scrubber, hydrated to sulfurous acid (H_2SO_3), and ionized to bisulfate (HSO_3^-) as well as sulfite (SO_3^{2-}). These compounds could be oxidized either by oxic seawater to sulfuric acid (H_2SO_4) in the scrubber or by air oxygen on filters to sulfate during sampling and storage (Karle and Turner, 2007). The possibility of the presence of sea salt (NaCl) was excluded because the amount of chloride ions (Cl^-) was below the detection limit of the ion chromatographic method. The reduction in the eBC emission factor by the scrubber was approximately 25% for MGO and HFO A and 55% for HFO B at 60 kW, and reductions of ca. 10% for MGO and ca. 30%

for HFO A and HFO B were found at 20 kW. The high reduction in eBC with HFO B for both engine loads seems to be caused by the higher FSC compared to the other fuel types that enhances the formation of hygroscopic particulate sulfate. During the combustion process, sulfur-containing particles are internally mixed with eBC, which can lead to higher scrubbing efficiency by wash water due to the enhanced hygroscopic growth of particles (Lack and Corbett, 2012). Consequently, the scrubber reduced the EFs of total particle mass and eBC, and the sulfate mass emission factor was increased for sulfur-containing fuels.

3.2 Alteration of particle number emission by a sulfur scrubber

To improve the understanding of the physical mechanisms in the scrubber, the particle number, mass EFs, and their size distributions in the size range of 14.1–713 nm were investigated (Fig. 2 and Fig. S2). Two distributions were demonstrated per panel for the scrubber upstream and downstream for both engine loads. In agreement with previous studies, very high particle number EFs were measured in the raw exhaust gas of both HFOs, approximately 15–20 times higher compared to MGO at 60 kW, which is partially attributed to the sulfuric/sulfurous acid particle formation (Moldanova et al., 2013; Sippula et al., 2014). The formation of those small particles results in the skewed size distribution of HFOs (Fig. 2), which is in line with previous experiments with the same engine (Mueller et al., 2015). An unstable operation of the motor with MGO at a 20 kW engine load resulted in a comparable particle number emission factor as HFOs and no reduction in PM from MGO by the sulfur scrubber (Table 3).

The distribution downstream of the scrubber exhibited a shift in size distribution to larger particles in the case of the HFOs in both operation modes, whereas the MGO size distribution was almost unchanged in the open-sea operation mode. The scrubber reduced the particle number emission factor for all fuel types. In the case of MGO, scrubber usage resulted in a 40% reduction in the particle number emission factor, and the particle number reduction of HFOs was 25% and 30% for HFO A and HFO B, respectively. In the maneuvering mode, particle number reduction of HFOs was 38% and 44% for HFO A and HFO B, respectively (Table 3). Interestingly, the particle number EFs were reduced to a higher extent for HFO B compared to HFO A in both operation modes. A remarkable part of this reduction was found in particle diameters below 100 nm, while the scrubber increased the number of particles with diameters larger than 100 nm. A reduction in particle number concentration from HFOs by the scrubber was already shown because of the particles which are formed during the combustion process in the nucleation and Aitken mode (Fridell and Salo, 2016). However, a shift in size distribution was only found in recently published studies by Kuittinen et al. (2021) andd Santos et al. (2022). Therefore, it is worth investigating to what extent a pure physical coagulation process could play a role in the observed change in particle size.

In Table 3, the observed particle number emission factor and CMD for all fuel types are compared with calculated values of the coagulation model. As aforementioned, the applied research ship motor was not running stable at an engine load of 20 kW for MGO. Therefore, the model prediction of the coagulation process barely met the experimental results. However, at an engine load of 60 kW, the model showed slightly changed number EFs and CMD, indicating that the coagulation played a negligible role due to the low initial particle number emission factor (Hinds, 2011). The deviation between the calculated and measured values of MGO in the open-sea operation mode is caused by the effect of the washout by the scrubber, which was observed in the size distribution with a minor change in the mode and a reduction in the particle number and mass EFs (Fig. 2).

Compared to MGO, a shift in size distribution of the HFOs in both operation modes was observed, which can partly be attributed to the particle-to-particle coagulation processes initiated by the high number emission factor, although the particles were partially removed by the scrubber (Table 2). On the one hand, the calculated particle number EFs for HFO A in the open-sea operation mode and HFO B in the maneuvering mode were lower than the measured emission factor. On the other hand, the particle number EFs for HFO A and HFO B were comparable with the measured value. However, the calculated CMD was smaller than the measured CMD for both HFOs as well as both operation modes, which could be caused by an additional coagulation process of particles with sulfur-containing particles in the scrubber. The enhanced fraction of the sulfur-containing particles resulted in a higher particle mass emission factor after the scrubber, although the mass emission factor of the primary particles such as eBC was reduced to a moderate extent by washout processes. According to the results of the coagulation model and its limitations, the mechanisms leading to a shift in the size distribution and a change in the mass emission factor of the HFOs after the scrubber can be seen as a mixture of the coagulation of both primary and sulfur-containing particles in the scrubber and its washout process. Therefore, the results of the coagulation model should be interpreted carefully to study an important physical mechanism in the scrubber.

In conclusion, the investigation of ship engine emissions with and without the application of a wet scrubber showed that the scrubber technology on its own only enables little or no precipitation of the inhalable PM fraction regarding particle number and mass EFs. This is in line with studies by Lehtoranta, Yang, and Zhou (Lehtoranta et al., 2019; Yang et al., 2021; Zhou et al., 2017). Clearly, particle size and its number concentration are not the only parameters leading to adverse health effects. Nevertheless, further reduction in the inhalable PM fractions from ship engines, which were identified as severe public health risk, should be considered one important measure (Corbett et al., 2007; Oeder et al., 2015; Sapcariu et al., 2016). In this context, a promising particle removing technology, the WESP, was also tested in this study as an additional exhaust gas cleaning device.

3.3 Particle removal using a wet electrostatic precipitator (WESP)

The potential benefits from a WESP device were explored as an additional abatement system downstream of the wet scrubber only with HFO B at an engine load of 60 kW due to the electrical malfunctions of the WESP itself. The total particle number emission factor was reduced by WESP up to 98%. The significant reduction in the particle number emission factor by WESP resulted in a remarkable reduction in the particle mass emission factor from 218 mg/kWh (downstream) to below the detection limit of the instrument (3 µg/m^3, 2 mg/kWh). In terms of mass collection efficiency, the results of this study correspond with those from the previous experiments by Bologa et al. (2009). As herein shown, the WESP approach demonstrates very good removal efficiencies and seems to be suited to precipitate PM from the wet exhaust gas downstream of the wet scrubber. Additional tests are needed as the WESP was operated with the maximum PM removal efficiency for only a limited timeframe of 30 min in our experiments. We recommend that this technology should be further optimized for marine applications. Thus, vessels using the scrubber technology to operate with HFOs can be equipped with an efficient technology to reduce the health impact of the shipping emissions.

4. Conclusions

In this study, gaseous and particulate emissions from a ship diesel engine, as well as their treatment using a scrubber and WESP, were investigated. The measurement of particulate matters without any abatement

systems showed a skewed size distribution toward a smaller particle size of HFOs with a higher total number and mass EFs compared to those of MGO. Downstream of the scrubber, the total particle mass decreased slightly for MGO and HFO A but not for HFO B, which can be attributed to the reduction in eBC and an increase in inorganic sulfate particle fractions. Therefore, we could elucidate that the utilization of the wet scrubber can lead to changes in the compositions of PM. The scrubber also affected the number size distribution by decreasing the number concentration of smaller particles with diameters below 100 nm and increasing the number concentration of bigger particles. This agrees with the results of a polydisperse coagulation model as the scrubber serves as a coagulation chamber by extending the residence time of particles. Therefore, the scrubber was not able to remove inhalable PM fractions regarding the number and mass EFs substantially, which have been identified as severe public health risk. To overcome the limitation of scrubbers related to particle emission, in our measurement, WESP was connected directly after the scrubber to attain a secondary reduction in PM from the engine. The combination of the scrubber and WESP removed particle emissions from the exhaust almost entirely.

This study could help in understanding the feasibility of scrubber usage regarding the reduction in particulate and gaseous emissions from ships. Moreover, it suggests options for potential enhancements of this technique to help policy makers update their regulations. Our study showed that the reduction in particulate emissions by wet scrubbers is linked to the fuel type, but it is not as efficient as intended by the coregulation of pollutants. Hence, a ship engine connected with only a scrubber cannot be a promising solution to comply with future regulations that will consider particulate emissions. To reduce the effects of PM related directly as well as indirectly to climate impacts and adverse health effects, the implementation of a filter system seems to be a promising approach.

401 # Tables

402 **Table 1.** Physico-chemical properties of applied fuels.

		MGO	HFO A	HFO B
Density at 15 °C	g/cm^3	0.835	1.01	0.990
Viscosity at 50 °C	mm^2/s	2.37	378.42	354.41
Sulfur	% (m/m)	0.001	1.02	2.16
Water	mg/kg	23	1031	586
Flash point	°C	72	147	135
Ash	% (m/m)	0.001	0.019	0.051
Heating value	MJ/kg	42.7	40.1	40.1
Ni	mg/kg	<1	34	60
V	mg/kg	<1	47	235
Fe	mg/kg	<1	20	51

403

404 **Table 2**. Results from the exhaust gas and particle measurements at 20 kW and 60 kW engine loads. Emission factors
405 of NO_x, SO_2, PM and eBC with standard deviations measured within 20 min, 20 min, 60 min, and 60 min, respectively.
406 Sulfate and EC mass emission factor (only for 60 kW) measured on filter within 20 min and presented as minimum
407 and maximum (n =2).

		MGO	HFO A	HFO B
Upstream			**20 kW**	
NO_x as NO_2	g/kWh	9.3 ± 0.6	12.6 ± 0.7	9.6 ± 0.6
SO_2	g/kWh	< LOQ*	5.0 ± 0.1	11.3 ± 0.3
PM mass	mg/kWh	96 ± 13	1960 ± 167	1224 ± 225
Sulfur as Sulfate	mg/kWh	< LOQ**	< LOQ** - 17	19 - 37
eBC	mg/kWh	26 ± 6	849 ± 20	361 ± 78
			60 kW	
NO_x as NO_2	g/kWh	8.0 ± 0.5	10.6 ± 0.6	8.1 ± 0.6
SO_2	g/kWh	< LOQ*	4.0 ± 0.1	13 ± 0.8
PM mass	mg/kWh	59 ± 3	275 ± 12	201 ± 8
Sulfur as Sulfate	mg/kWh	< LOQ***	< LOQ*** - 13	31 - 36
eBC	mg/kWh	36 ± 4	94 ± 12	36 ± 4

EC	mg/kWh	28 - 49	120 - 129	33 - 34
Downstream			**20 kW**	
NO_x as NO_2	g/kWh	8.7 ± 0.4	12.8 ± 0.6	9.4 ± 0.6
SO_2	g/kWh	< LOQ*	< LOQ*	< LOQ*
PM mass	mg/kWh	216 ± 27	1416 ± 327	1148 ± 465
Sulfur as Sulfate	mg/kWh	< LOQ**	< LOQ**	48 - 64
eBC	mg/kWh	23 ± 6	547 ± 11	238 ± 23
			60 kW	
NO_x as NO_2	g/kWh	7.7 ± 0.4	10.3 ± 0.4	8.0 ± 0.5
SO_2	g/kWh	< LOQ*	< LOQ*	< LOQ*
PM mass	mg/kWh	39 ± 1	201 ± 23	218 ± 13
Sulfur as sulfate	mg/kWh	< LOQ***	21 - 31	89 - 116
eBC	mg/kWh	26 ± 4	70 ± 18	16 ± 3
EC	mg/kWh	16 - 17	42 - 62	8 - 14

* LOQ-limit of quantification (0.15 mg/kWh), **LOQ (12 mg/kWh), *** LOQ (7.7 mg/kWh)

Table 3. Comparison of measured and estimated particle number emission factors regarding the coagulation process at 20 kW and 60 kW engine loads with standard deviations of particle number emission factors measured within 60 min.

		MGO	**HFO A**	**HFO B**
Number emission factor			**20 kW**	
Upstream measured	E14 1/kWh	1.28 ± 1.2	3.99 ± 0.3	6.38 ± 0.7
Downstream measured	E14 1/kWh	2.79 ± 0.7	2.45 ± 0.1	3.60 ± 0.7
Downstream calculated	E14 1/kWh	0.96	2.33	2.73
			60 kW	
Upstream measured	E14 1/kWh	0.39 ± 0.04	2.56 ± 0.06	4.23 ± 0.2
Downstream measured	E14 1/kWh	0.23 ± 0.001	1.93 ± 0.09	2.88 ± 0.3
Downstream calculated	E14 1/kWh	0.36	1.45	2.93
Count median diameter (CMD)			**20 kW**	
Upstream measured	nm	50	80	65
Downstream measured	nm	72	109	102

Downstream calculated	nm	55	96	86
			60 kW	
Upstream measured	nm	64	32	46
Downstream measured	nm	66	58	72
Downstream calculated	nm	65	39	52

Figures

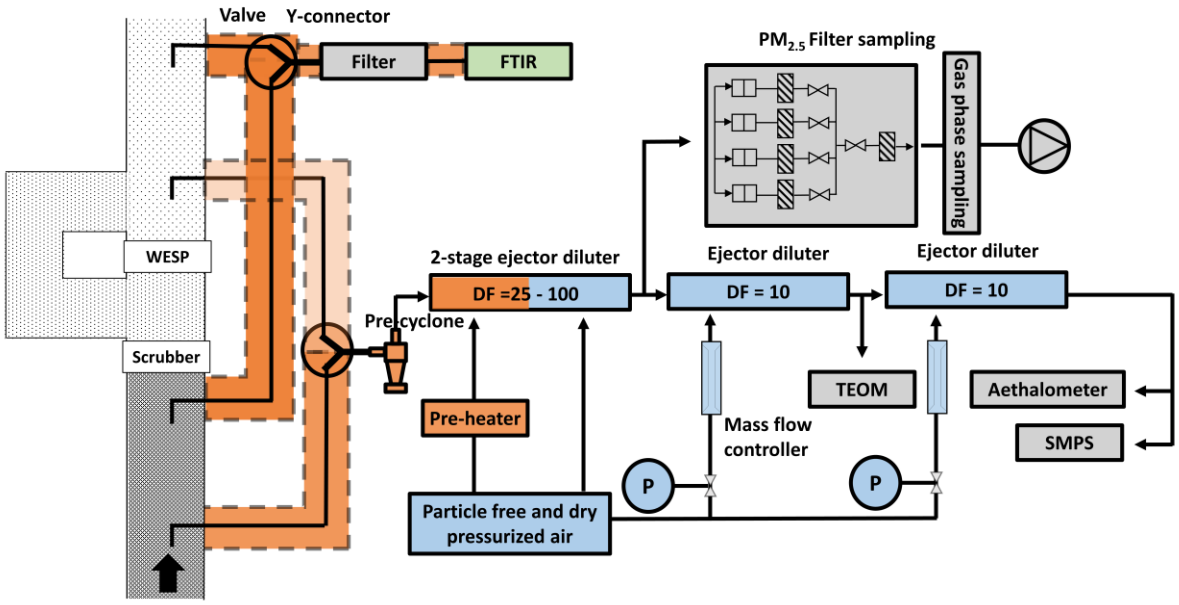
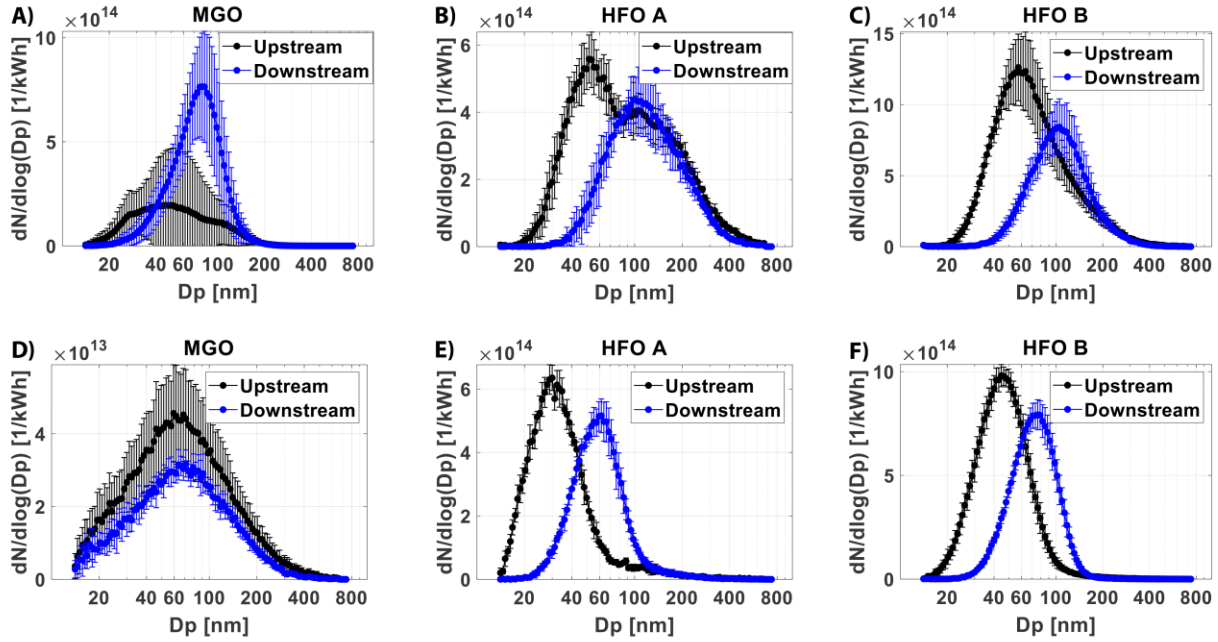


Fig. 1. Scheme for the sampling setup in the measurement held at the University of Rostock, Germany. TEOM- Tapered element oscillating microbalance, SMPS- Scanning mobility particle sizer, FTIR- Fourier transform infrared spectroscopy, WESP- wet-electrostatic precipitator, DF- Dilution Factor, P- Pressure regulator.



419

420 **Fig. 2**. Size distribution of particle number emission factors up- (black circles) and downstream (blue circles) of the
421 wet scrubber from fuel types A) MGO, B) HFO A, C) HFO B, D) MGO, E) HFO A, F) HFO B at 20 kW and 60 kW
422 of engine load, respectively.; Error bars represent the standard deviations of particle number emission factors measured
423 within 60 min.

424

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Declaration of competing interest

The authors declare that they have no competing interests.

Appendix A. Supplementary data

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