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Measurements of abatement of particles and exhaust gases in a marine gas scrubber

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Abstract

Measurements of exhaust gases from a marine engine equipped with an open-loop wet scrubber using seawater for sulphur dioxide (SO_2) abatement are reported. The scrubber reduces the SO_2 emissions effectively to levels corresponding to $<0.1\%$ S in the fuel (the level that applies to sulphur emission control areas from 2015). The scrubber also reduces the emissions of particulate matter by mass by 75%. The impact on the number of particles is studied for the total particle number and the solid fraction. The total number of particles is reduced by about 92% and the solid fraction by 48% in the scrubber indicating most effective abatement of volatile particles. Also, the content of polycyclic aromatic compounds in the exhaust is significantly reduced by the scrubber. The captured SO_2 gives a low pH and high sulphate content in the scrubber water. The reduction in particulate matter is of the same order as what is obtained with a fuel switch from heavy fuel oil to marine gas oil.

Keywords

Seawater scrubber, particulate matter, SO_2, polycyclic aromatic compounds, black carbon

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Introduction

From 2015, the maximum allowed sulphur content in marine fuels in sulphur emission control areas (SECAs) is 0.1% by weight, and from 2020, it is 0.5% for the rest of the world as stipulated by the International Maritime Organization (IMO) in the International Convention for the Prevention of Pollution from Ships (MARPOL), Annex VI, regulation 14.[1] These regulations will most likely result in a switch from heavy fuel oils (HFO) to distilled products such as marine gas oil (MGO) and possibly other fuels such as liquefied natural gas (LNG) or methanol. However, there is also according to MARPOL Annex VI, regulation 4,[1] the possibility to use exhaust abatement equipment that 'are at least as effective in terms of emissions reductions' in combination with high-sulphur HFO.

The after-treatment technologies that are available are dry or wet scrubbing of sulphur dioxide (SO_2) in the exhaust gas. In dry scrubbing, the exhaust gas is brought in contact with granules of calcium hydroxide producing gypsum. Wet scrubbing can be done with seawater (open-loop scrubbing) where the natural alkalinity is used in order to capture SO_2. The scrubber water is then returned to sea, sometimes after treatment to remove solid particles. The trapping of SO_2 makes the scrubbing water acidic and it will be neutralised by the natural alkalinity of seawater, either onboard or in the sea outside the ship.[2] Wet scrubbing can also be done with closed-loop systems using fresh water and the addition of sodium hydroxide to provide the alkalinity. Such system emits smaller amounts of treated water with sulphates into the sea and a sludge is formed that should be deposited on land. The ability of a wet scrubber to capture SO_2 has been demonstrated in a number of trials.[3–6] Furthermore, seawater scrubbers have been studied in laboratory experiments[7] and theoretical assessments.[8] The experimental and model data show that seawater scrubbers are capable of effectively capturing SO_2 in the exhaust gas, with an efficiency depending on the alkalinity, and that the scrubber

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water will be acidic. According to the IMO regulations, the emissions of SO_2 need to be reduced to levels corresponding to what is obtained following the regulations on fuel sulphur content (SFC).[1]

The background to the limits on SFC decided by the IMO was in part problems with acidification of water and land, but a main reason was to reduce the contribution from shipping to levels of particulate matter.[9] It has been established that there is a connection between the emissions of particulate mass (PM) and SFC[10] which is due mainly to the formation of secondary sulphate particles but also due to the change in fuel quality – from HFO to MGO – that follows from the regulations. Thus, the sulphur fuel content is used as a proxy for PM emissions eliminating the need for specific regulations and monitoring of PM emissions as is done, for example, for truck engines and inland waterway engines. However, it should be noted that there is no direct proportionality when other particle types than sulphates are considered, and further research in this field is required. A switch from HFO with a high sulphur content (2%–3%) to MGO with about 0.1% S results in reduction in the PM emissions of about 75%.[10] It should be noted that there are different methods to measure PM emissions, both regarding the dilution and the instrumentation used, giving uncertainties in the reduction. Trials with wet scrubbers have given somewhat different results on the PM reduction capability, from negative values,[11] most likely associated with difficulties in probing PM in the humid and cool exhaust after the scrubber, to about 80%.[3] Furthermore, it is also of interest to analyse the abatement when it comes to number of particles (PNs) and particle fractions such as black carbon (BC). Reduction in the number concentration of 10%–80% has been reported.[12]

The guidelines decided by the IMO regarding abatement equipment also state certain criteria for the scrubber water that is returned to sea. These criteria should make certain that harmful substances are not emitted into the environment and comprise limits for pH, polycyclic aromatic compounds (PAHs), turbidity and nitrate content.[1]

In this study, data from onboard measurements for exhaust abatement and wash water properties are reported for an open-loop wet scrubber. The focus is on analysing the abatement of particles and characterisation of particles before and after the scrubber.

Methods

The measurements took place onboard the 8420 dwt roll-on roll-off vessel Ficaria Seaways where a scrubber system (Alfa Laval PureSOx) had been installed on the two-stroke 21.06-MW main engine. The scrubber system can be switched between open and closed loop, but only the open (seawater) mode was used during the measurements reported here. The SO_2 is scrubbed in two stages, first in a jet scrubber where the exhaust is also cooled, and then the main scrubbing is done in an absorption tower. A main pump feeds water into the jet, the absorber stage and a demister. The water is then returned to the sea. The absorption tower is 4.6 m in diameter, 10 m high and weighs 32 ton (including water). More details on the system can be found in Hansen[3]. The measurements were done in February 2013 during a journey on the North Sea for an engine power of around 10,700 kW giving an exhaust flow of about 130,000 N m^3/h. The scrubber used about 500 m^3/h of seawater at these conditions. The fuel used was an HFO with about 2.3% S. Fuel analysis was obtained from an accredited laboratory, and the results from this analysis are displayed in Table 1. Detailed time-resolved (every 30 s) information was obtained from the ship regarding engine power, water flow, fuel flow and specific fuel consumption.

Table 1. Results from the fuel analysis.

Viscosity at 50 °C	mm^2/s	386
Density at 15 °C	g/cm^3	0.9864
C	wt%	86.4
H	wt%	10.6
S	wt%	2.3
O	wt%	<0.5
Calorimetric heat capacity at constant volume	MJ/kg	42.83
Effective heat capacity at constant pressure	MJ/kg	40.57

An overview of the sampling setup is given in Figure 1. Gas sampling first took place using a sampling port after the scrubber and then using a port in the engine room for probing the gas before the scrubber. Thus, the exhaust gases before and after the scrubber were not probed simultaneously. However, the conditions and the engine power were similar throughout the measurements. Scrubber water was sampled using a tap for wash water and seawater was collected using a water line in the engine room.

Sampling of gases was done with a probe positioned inside the exhaust tube. Continuous measurements were done for nitrogen oxides (NO_X) (Horiba PG-250 chemiluminescence instrument), SO_2 (Horiba PG-250 non-dispersive infrared (NDIR)), oxygen (O_2) (Horiba PG-250 galvanic cell), carbon monoxide (CO) (Horiba PG-250 NDIR), carbon dioxide (CO_2) (Horiba PG-250 NDIR) and total hydrocarbons (THCs) (Bernath Atomic BA 3006 flame ionisation detection (FID)). Particle sampling was done using an isokinetic heated probe kept at 250 °C. The exhaust gas was diluted between 64 and 109 times using a Dekati FPS-4000 dilution system. The dilution ratio was monitored by measuring NO_X in the diluted gas (instrument Thermo Scientific 42i) and relating to the concentration in the raw gas. Particle number and size distribution were monitored continuously using two instruments: the electrometer-based Engine Exhaust Particle Sizer

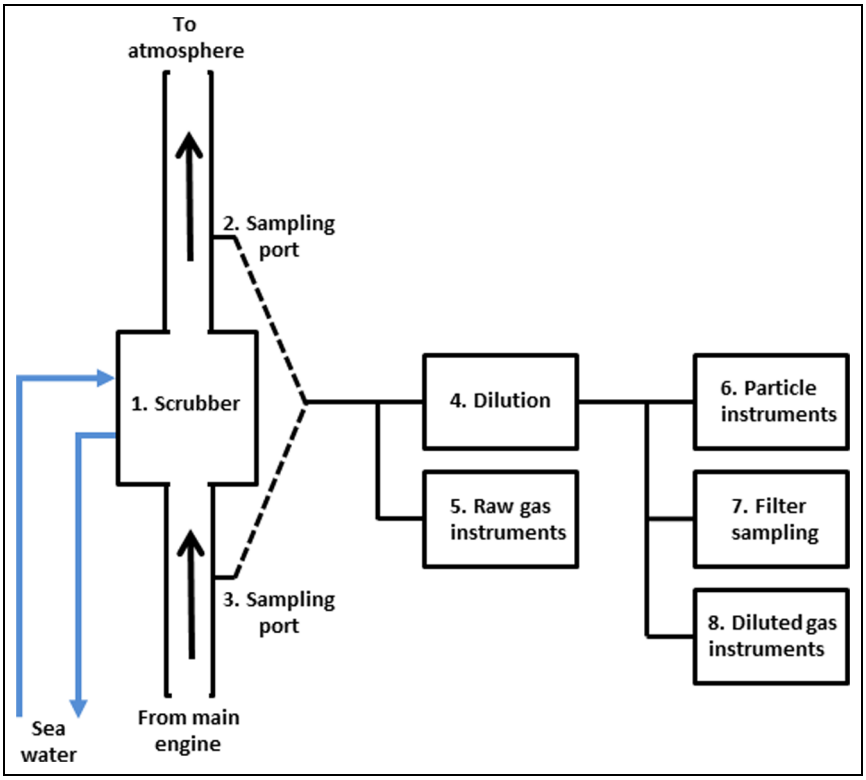

Figure 1. A schematic illustrating the sampling setup and main instrumentation used (1: the open-loop seawater scrubber; 2: sampling port after scrubber; 3: sampling port before scrubber; 4: dilution of raw exhaust gas; 5: raw exhaust gas instruments; 6: particle instruments; 7: filter sampling; 8: diluted gas instrument).

(EEPS Model 3090; TSI Inc.) with a size range of 5.6–560 nm and a time resolution of 0.1 s; and an Optical Particle Sizer (OPS 3030; TSI Inc.) with a size range of 0.3–10 μm. Before the particle instruments, the sample flow optionally passed through a thermodenuder (TD; Dekati ELA 423) kept at 300 °C to measure the ratio of volatile to non-volatile particles. These measurements were corrected for size-dependent particle number losses, for example, thermophoresis and diffusion according to the manufacturer's instructions. Filter samples were used to quantify the total suspended particulates (TSPs) and were taken in the diluted gas for gravimetric analysis and analysis of BC, elementary carbon (EC) and organic carbon (OC) content.

The analysis of EC/OC is based on a thermal optical method described earlier.[13] A 1-cm^2 piece of the sample taken on quartz fibre filters was used for the measurement. OC is removed from the filter in the temperature range of 200 °C–650 °C in a non-oxidising carrier gas (helium). EC is then removed in the temperature range of 500 °C–850 °C making use of a mixture of helium and oxygen. The originated CO_2 is then converted to methane and detected by FID. For analyses of BC, a soot scan transmissometer was used (see Moldanová[13] for details). PM-bound PAHs were sampled on Teflon Mitex membrane filters. In series with the filter-holders were coupled glass columns with XAD2 absorbent, which sampled the gas-phase PAHs. The PAH content was analysed using high-performance liquid chromatography (HPLC) with fluorescence detection.

One measurement of the sulphur trioxide (SO_3) concentration in the hot exhaust after the scrubber was done. The sampling was done by pumping a flow of exhaust gas through a column with sodium chloride salt placed inside the hot exhaust manifold.[14] In this sampling, the SO_3 reacts to form sulphates while SO_2 passes unaffected. The salt columns were later analysed for sulphates with ion chromatography.

The temperature and humidity of the inlet air were monitored. The exhaust temperature in the hot exhaust before the scrubber was around 254 °C. The water samples were tested for pH, nitrite/nitrate and sulphate content using ion chromatography.

The uncertainties given for particle mass are the statistical 95% confidence intervals from analysis of several filters. The uncertainties for particle number are the 95% confidence interval from the fluctuations in the signal during stable conditions. The uncertainties for gas concentrations, PAH and ion content in water are 95% confidence interval as given in the documentation for the accreditation of the analysis methods. Uncertainties are not given for BC, EC and OC due to too few samples. These uncertainties are likely at least as large as for particle mass.

Results

The results for the exhaust gas and PM measurement before and after the scrubber are shown in Table 2 as concentrations and calculated as emission factors, given both as mass of emission per engine work output (g/kW h) and as mass of emission per mass of fuel consumed (g/kg fuel). The NO_X emission factors are corrected for ambient temperature and humidity as described in the IMO NO_X Technical Code.[15] There is a significant decrease in the SO_2 concentration showing that the scrubber is working as expected. The emission factor for SO_2 after the scrubber corresponds to what would be obtained with a fuel containing 0.03% S without a scrubber. The NO_X emissions are somewhat reduced by the scrubber. The emission of THC from the engine is low and no significant influence from the scrubber can be observed. The measured concentration of CO was higher after the scrubber than before and the reason for this is not clear.

The emission factors for PM are also given in Table 2 as an average for three and four filters sampled before and after the scrubber, respectively. The effective abatement for TSP in this scrubber is about 75%. The analysis of BC, EC and OC shows that the scrubber abates soot and organic particles at the same order of magnitude as for TSP.

The results from the PAH analyses are shown in Table 3. There is a significant reduction in gas-phase PAH after the scrubber.

The results from the measurements with the EEPS of size distribution before and after the scrubber of the emitted particles are shown in Figure 2(a) and (b).

Table 2. Results from the exhaust gas measurements.

	Before scrubber			After scrubber		
	ppmv	g/kW h	g/kg fuel	ppmv	g/kW h	g/kg fuel
NO_Xcorr	829 (41)[a]	15.7 (0.8)	91.4 (5.1)	627 (31)[a]	13.8 (0.7)	80.1 (4.6)
HC	4 (2)	0.03 (0.01)	0.15 (0.07)	3 (2)	0.02 (0.01)	0.13 (0.08)
CO	102 (7)	1.31 (0.09)	7.62 (0.53)	264 (18)	3.93 (0.28)	22.9 (1.6)
SO_2	374 (26)	11.0 (0.8)	63.9 (4.5)	3.1 (0.2)	0.11 (0.01)	0.61 (0.04)
SO_3	n.d.	n.d.	n.d.	38 (n.d.)[b]	1.0 (n.d.)	5.9 (n.d.)
PM (TSP)	83 (11)[c]	0.89 (0.12)	5.2 (0.7)	19 (16)[c]	0.22 (0.18)	1.3 (1.1)
BC	1.0 (n.d.)[c]	0.011 (n.d.)	0.064 (n.d.)	0.10 (n.d.)[c]	0.0012 (n.d.)	0.0069 (n.d.)
EC	1.7 (n.d.)[c]	0.018 (n.d.)	0.11 (n.d.)	0.30 (n.d.)[c]	0.0034 (n.d.)	0.019 (n.d.)
OC	10 (n.d.)[c]	0.12 (n.d.)	0.67 (n.d.)	2.9 (n.d.)[c]	0.032 (n.d.)	0.19 (n.d.)

HC: hydrocarbon; PM: particulate mass; TSP: total suspended particulate; BC: black carbon; EC: elementary carbon; OC: organic carbon; n.d.: not determined.
Uncertainties in brackets.
[a]Uncorrected value.
[b]mg S/Nm^3.
[c]mg/Nm^3.

Table 3. Results from PAH analysis.

	Before scrubber		After scrubber		Uncertainty (%)
	Filter sample μg/kg fuel	Gas sample μg/kg fuel	Filter sample μg/kg fuel	Gas sample μg/kg fuel	
Naphthalene	0.8	862	0.2	10	40
Acenaphthene	4.3	119	< 0.08	2.0	40
Fluorene	0.6	140	0.3	3.7	40
Phenanthrene	0.3	464	0.2	67	30
Anthracene	0.1	23	0.008	3.7	40
Fluoranthene	0.01	4.5	< 0.04	8.6	20
Pyrene	0.8	62	0.09	31	20
Benzo(ah)anthracene	<0.6	1.6	0.7	8.6	40
Chrysene	<0.6	<0.8	0.3	4.4	30
Benzo(b)fluoranthene	4.4	<1.0	0.5	0.2	30
Benzo(k)fluoranthene	5.2	<0.6	0.2	0.03	20
Benzo(a)pyrene	4.8	<0.8	0.2	<0.03	20
Dibenz(ah)anthracene	27	<0.8	<0.02	<0.03	30
Benzo(ghi)perylene	5.1	<1.4	<0.04	<0.2	40
Indeno(1,2,3)pyrene	16	<2.7	0.03	<0.06	20
Sum analysed PAH	66	1657	3.0	138	

PAH: polycyclic aromatic compound.

Table 4. Measured total particle number concentration.

	Before scrubber			After scrubber		
	#/cm^3	#/kW h	#/kg fuel	#/cm^3	#/kW h	#/kg fuel
PN	7.76×10^8 (2.1×10^8)	8.74×10^{15} (2.3×10^{15})	5.08×10^{16} (1.4×10^{16})	7.20×10^7 (1.6×10^6)	8.08×10^{14} (1.8×10^{13})	4.70×10^{15} (1.0×10^{14})
PN solid	1.40×10^8 (1.1×10^7)	1.36×10^{15} (1.1×10^{14})	7.93×10^{15} (6.1×10^{14})	6.29×10^7 (8.0×10^5)	7.07×10^{14} (9.0×10^{12})	4.11×10^{15} (5.2×10^{13})

PN: number of particle.
'PN solid' refers to measurements using the thermodenuder. Uncertainties in brackets.

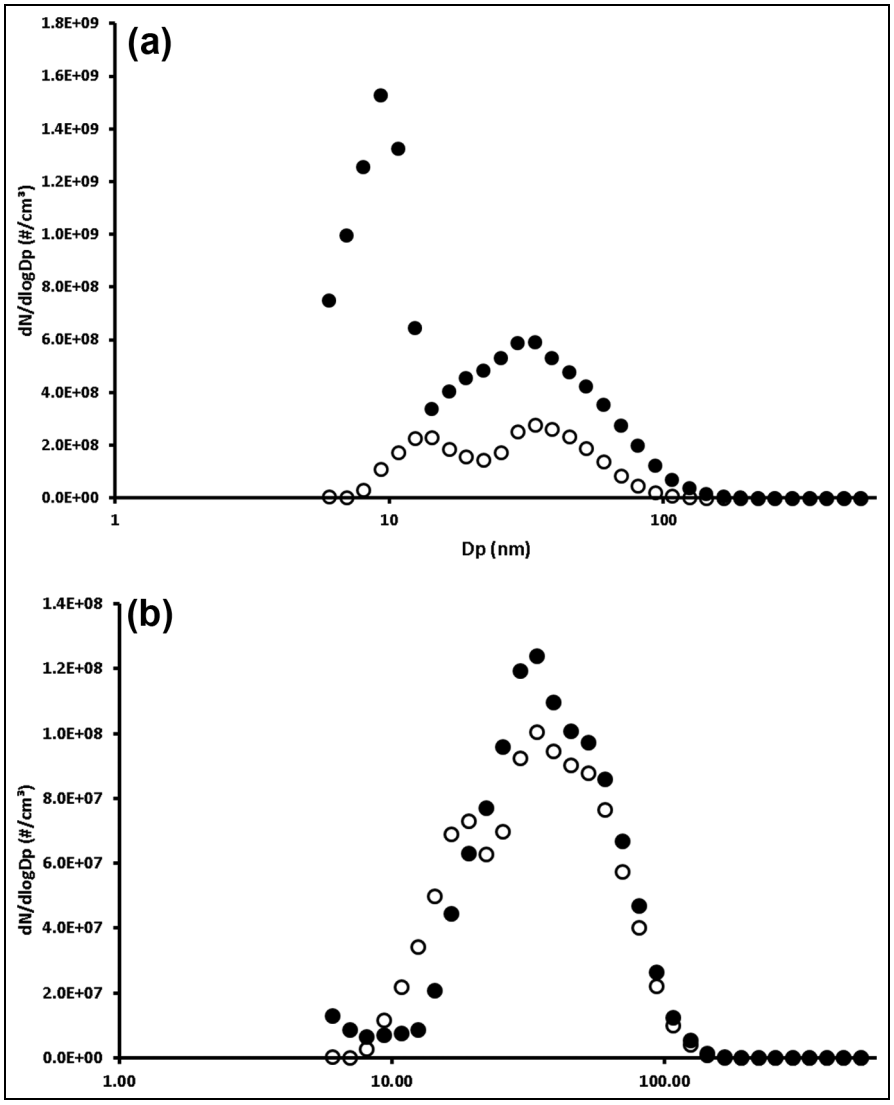

Figure 2. Particle number concentrations (a) before and (b) after scrubber, with (open circles) and without (closed circles) thermodenuder.

There is a significant reduction in particle concentration for all measured sizes after the scrubber compared with before the scrubber. The use of the TD reduces all particle sizes in the unabated exhaust. Most notably, the peak at around 10 nm is significantly reduced showing that this peak contains volatile particles such as sulphates and hydrocarbons that are removed by the TD. For the exhaust gas after the scrubber, the TD has little influence on the PN concentration showing that most of the volatile particles are removed from the exhaust by the scrubber. The measured emission factors for total PNs are given in Table 4.

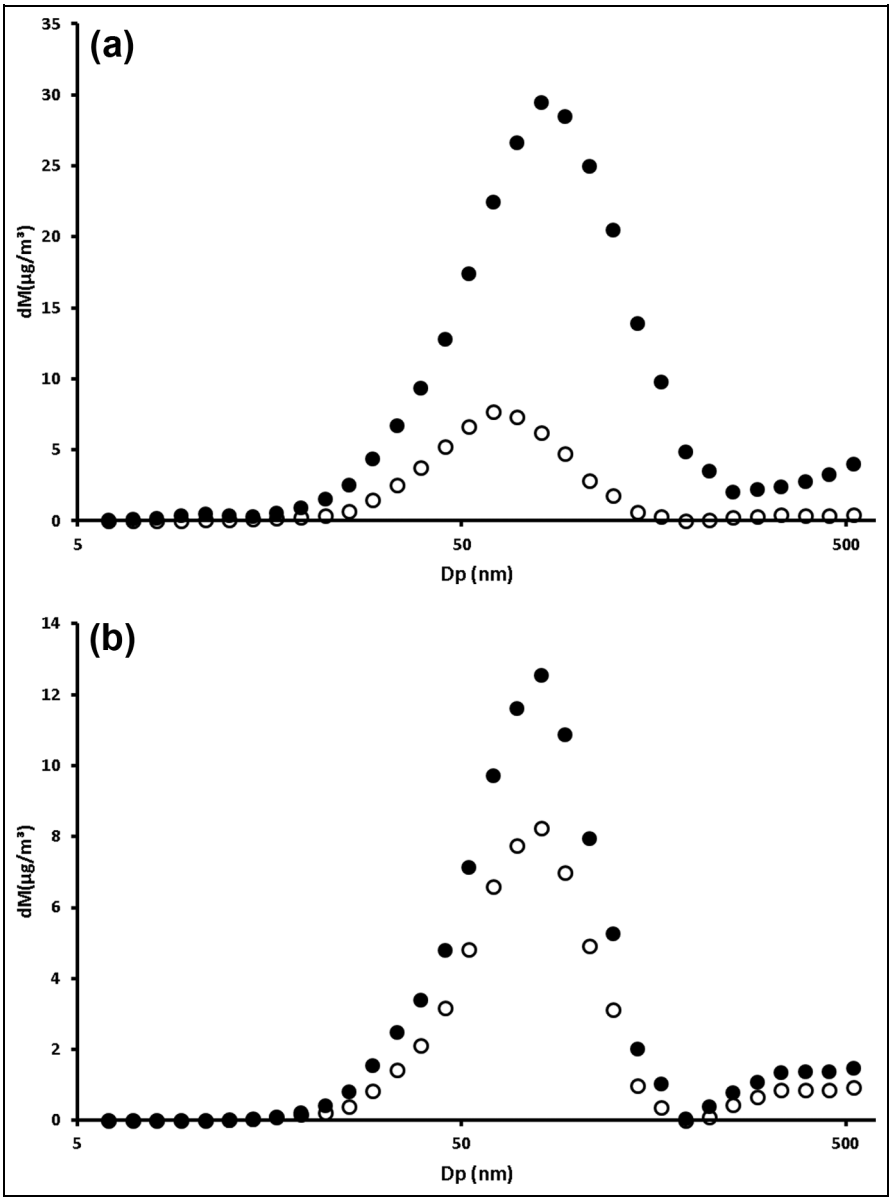

Figure 3. Particle mass concentrations (a) before and (b) after scrubber, with (open circles) and without (closed circles) thermodenuder.

In Figure 3(a) and (b), the data from Figure 2 are shown as mass distribution calculated assuming spherical particles and unit density. The distributions show maxima at 60–80 nm and it can be noted in Figure 3(b) that the peak at around 10 nm in Figure 2(a) contributes little to the mass.

The emission factors for PM, PN and SO_2 before and after the scrubber are compared with a typical MGO in Figure 4.

Figure 5 shows the size distribution of particles measured with OPS before and after the scrubber. There are very few particles > 500 nm and a reduction in the particle concentration can be observed also for the size range of 300–600 nm.

The results of the analyses of the scrubber water are presented in Table 5 where the content in seawater has been subtracted. The nitrite content was below the detection limit and the pH was 3.44.

Discussion

The emissions from the engine studied here can be considered as typical for a two-stroke marine engine running on HFO. The NO_X emission was 15.7 g/kW h, which is below Tier 1 limit of 17.0 g/kW h for slow-speed engines. The emission of PM as TSP was found to be 0.89 g/kW h. This is in range with earlier studies but is somewhat on the low side.[10,13,16] Also, the emission factors for BC, EC and OC are in the same range as reported earlier.[14,17] The total emission of PNs was 8.74×10^{15}/kWh without the TD. This is in agreement with earlier onboard measurements[13,18] and also with

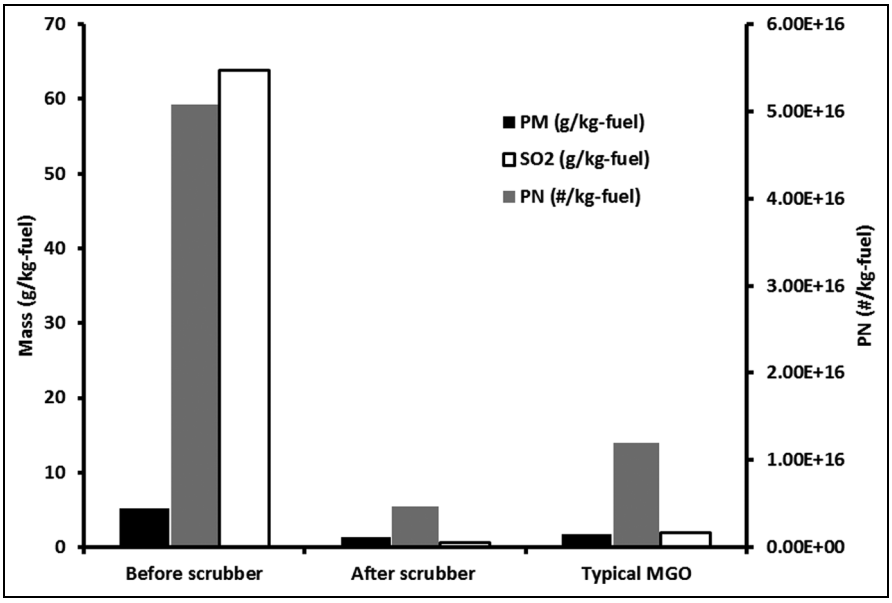

Figure 4. Emission factors for PM, PN and SO_2 in comparison with an MGO.
PM: particulate mass; PN: number of particle; MGO: marine gas oil.

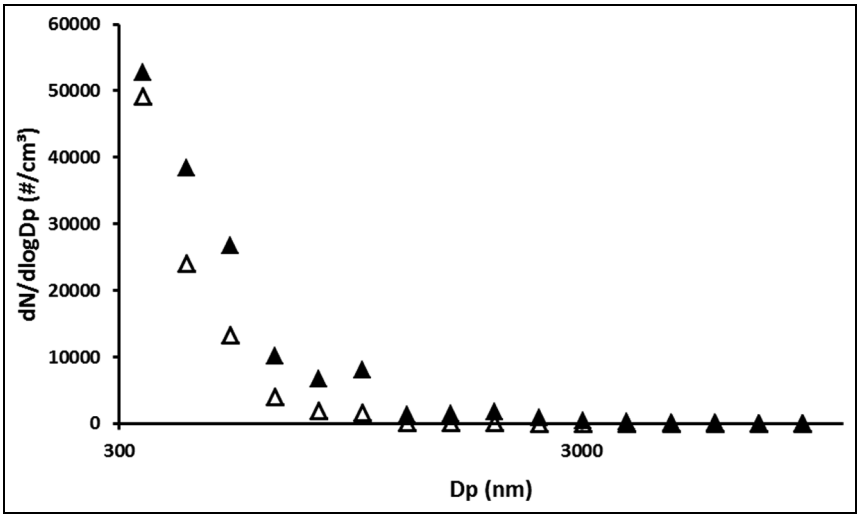

Figure 5. Aerosol and number distribution between 0.3 and 10 μm as measured with optical particle sizer (OPS), before (closed symbols) and after (open symbols) the scrubber.

Table 5. Results from analysis of scrubber water (content in seawater has been subtracted).

	Contents in scrubber water	
	g/kW h	g/kg fuel
NO_3	1.8 (0.09)	11 (0.5)
SO_4	29 (1.2)	170 (7)
TOC	0.07 (0.008)	0.4 (0.04)

TOC: total organic carbon.
Uncertainties in brackets.

plume measurements.[19–21] The emission factor for PN when using the TD was 1.36×10^{15}/kWh; thus the solid fraction represents about 16% of the PNs. This value is lower than the value reported by Jonsson et al.[19] performing plume measurements within an SECA that limits the SFC to 1%. Considered the high SFC in the fuel used here (2.3%), a higher fraction of volatile particles is expected. Furthermore, the size distribution shows a peak around 10 nm with mainly volatile particles and another peak at around 30 nm. The emissions of PAH are much larger to the gas phase than as particle-bound

PAHs. The total PAH emitted is here measured to be around 1.7 mg/kg fuel, which is of the same order of magnitude as has been reported earlier for HFO.[22]

The studied scrubber can effectively reduce the emissions to air of SO_2 to values that correspond to sulphur content significantly lower than for MGO. At an exhaust gas-to-water flow ratio of about 260 (corresponding to about 47 m^3 of seawater per MWh), a 99% reduction in SO_2 was measured. At a similar gas-to-water ratio of about 60%, SO_2 removal efficiency was reported by Caiazzo et al.[7] for laboratory experiments. A typical water flow rate of 45 m^3/MWh has been reported. Thus, the system fulfils the IMO regulations for SECAs after 2015. This opens for the possibility to use this system in combination with high sulphur HFO, which may be a less costly solution compared with switching to MGO at least for ships that frequently sail within SECAs. There is a discrepancy between the measured SO_2 (before the scrubber) and the SFC from the analysis with the measured SO_2 being 39% higher. Looking at the balance of S (using the measured SO_2 value), there are 32 g/kg fuel (as SO_2) accounted for before the scrubber and 60 g/kg fuel after the scrubber. Of the latter, 95.5% is in the form of sulphate in the scrubber water, 4% as gas-phase SO_3 and only 0.5% as gas-phase SO_2.

The emission of particulate matter is significantly reduced by the scrubber. About 75% of the total PM is captured by the scrubber and the obtained corresponding fractions for BC, EC and OC are 89%, 81% and 73%, respectively. The total PNs are reduced by 92% and the solid fraction (as measured using the TD) is reduced by 48%. The emission factor after the scrubber can be compared to what is obtained when MGO is used in a marine engine without any exhaust abatement, which typically is around 0.3–0.4 g/kWh.[10] Thus, the value obtained here of 0.22 g/kWh is somewhat lower showing that this scrubber may reduce PM levels to what is obtained from a fuel switch from HFO to MGO. The reduction in OC, BC and EC follows the change for TSP. It is difficult to assess the reduction that can be obtained from a fuel switch since the emissions of at least BC and EC also will depend on engine characteristics and engine maintenance. The reduction in the scrubber observed here is somewhat higher than what has been reported[13] as the difference between HFO and MGO. The number concentration is difficult to compare due to the uncertainties, but it is reasonable that there is a larger reduction than for mass for both a scrubber and a switch to MGO since sulphate particles are found in the ultrafine range.

When it comes to total mass of particles, the measurements reported here support that the scrubber can reduce the emissions at least to values that would result from a switch from HFO to MGO. However, when looking at the solid particles by number, the fraction captured in the scrubber is rather low. This is in somewhat contradiction to the results for BC and EC where a large fraction was captured. However, the latter is presented as mass which may explain the discrepancy. Thus, it is important to pay further attention to the capacity of scrubbers to reduce solid particles. It can also be noted that the recent regulations regarding emissions of PN by road vehicles use a method where only the solid particles are measured.

The data in Table 2 indicate that the scrubber captures some of the NO_X in the exhaust. When looking at the N-balance, 28 g/kg fuel is accounted for before the scrubber (all as NO and NO_2 in gas phase) and 27 g/kg fuel after the scrubber (91% in gas phase and 9% as nitrate in the scrubber water). Thus, these data indicate that the scrubber system complies with the wash water criteria from IMO for nitrates, which state that less than 12% of the NO_X may be trapped in the scrubber water. The origin of this criterion is a worry to further increase the load of nutrients to the sea. It should be emphasised that the measurements of NO_X before and after the scrubber were not done simultaneously in this study, which imposes an uncertainty on the results.

Other wash water criteria concern pH and PAH content. The pH measured here was 3.44, clearly lower than the criteria of 6.5. To reach a pH of 6.5, the scrubber water would need to be diluted in large volumes of seawater onboard the ship before being returned to sea. The PAH criteria state that the concentration of phenanthrene in the wash water must be less than 50 μg/L. The IMO criterion corresponds to about 2.25 mg/kW h of phenanthrene. This can be compared to the *total* mass of PAH in the raw exhaust measured in this study of 1.7 mg/kW h.

Finally, it should be noted that the measurement results presented here have inherent uncertainties, especially when it comes to PN and PAH concentrations. Before conclusions can be made about the possibility of scrubbers to abate particulate matter, further studies with other engines are needed.

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Declaration of conflicting interests

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