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ORIGINAL ARTICLE

Limited efficiency of wet scrubbers in reducing the environmental impact of ship-emitted particles

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Abstract

Sulfur dioxide pollution by ship emissions can be efficiently decreased by using exhaust gas scrubbers, yet particles can pass through the scrubber and be released into the atmosphere. Here, we studied the impact of using a wet scrubber on the composition of particle emissions, by single-particle analysis. At low engine loads, results show no significant changes in particle composition of metals, salts, and polycyclic aromatic hydrocarbons (PAH). At high engine loads, the scrubber reduced soot and PAH signatures about fourfold. Particles passing through the scrubber undergo minimal chemical changes, except for sulfate uptake. The cleaning effect of wet scrubbers is attributed to the removal of water-soluble gas-phase compounds, diffusion-dominated uptake of ultrafine particles, and wet deposition of coarse particles. The scrubber has little effect on reducing the health and environmental impacts of the remaining particles that pass through it. These emitted particles, primarily in the 60–200 nm size range, constitute a significant portion of the inhalable particle mass and have the potential for long-range transport.

Keywords Ship emissions · Air pollution · Scrubber · Polycyclic aromatic hydrocarbons · Exhaust cleaning · Inhalable particles

Introduction

The shipping industry is a major source of global air pollution [1, 31]. To mitigate its environmental and health impacts, the International Maritime Organization has introduced a global regulation, limiting the fuel sulfur content to a maximum of 0.5% (w/w). In Sulfur Emission Control Areas, such as the North Sea, Baltic Sea, and coastal waters along the U.S. coast, the limit is set at 0.1% (w/w). As an alternative to cleaner fuels, ships are permitted to use cheaper high-sulfur bunker fuels, provided that exhaust scrubbers are used to remove the SOx from emissions. Wet scrubbers can impact the environment when wash water is discharged during so-called open-loop operation [9, 32]. In addition to the efficient removal of SOx, which also reduces secondary sulfate aerosol formation, the air quality benefits include partial scrubbing of water-soluble organic gases, such as carbonyls, thereby reducing the ozone-forming potential of the emissions [7].

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The reduction in particulate emissions depends on fuel, engine, and scrubber parameters, and consequently, it varies in the literature [33, 35] with the majority of the studies indicating only little to moderate effects of the scrubber on particle number and mass emission factors [11, 13]. Changes in the particle size distribution are primarily linked to the diffusion-driven uptake of ultrafine particles, wet deposition of coarse particles, and variations in soot morphology [13, 15]. Additionally, new particle formation and particle growth through the conversion of SO_2 to sulfate play a significant role [14].

Although the chemical composition is a crucial factor in determining the particle's environmental and health impacts [23], the effect of scrubbers on this composition remains poorly studied. Possible scrubber-induced changes in the bulk chemical composition can result, e.g., from the size-selective removal of particles, but also from hygroscopic growth of particles with water-soluble components [14].

However, while data on the scrubber's effect on particulate matter bulk composition are already limited, even less is known at the single-particle level. It has been shown that the mixing state of particles significantly affects their environmental impact [25]. For example, the distribution of components across individual particles plays a role in determining the cloud condensation nuclei potential of the particle ensemble [26, 36]. In terms of biological effects, the concentration of pollutants on individual particles is important, as is the particle's acidity, which influences the bioavailability of transition metals [8].

In addition to changes in bulk composition caused by the scrubber, any alterations in the distribution of pollutants within the aerosol ensemble can impact the emissions' environmental effects, including long-range transport potential, cloud condensation behavior, and biological effects. In this study, we investigate the effect of a wet scrubber on the aerosol composition from a research ship engine at the single-particle level. The single-particle mass spectrometer is sensitive to particles larger than approximately 100 nm [19], which typically constitute the majority of the lung-deposited particle mass [12], also for ship emissions [18] and are capable of long-range transport [29]. Using recently developed technologies, the instrument detects key aerosol components relevant to environmental and health effects, including transition metals [20] and carcinogenic polycyclic aromatic hydrocarbons (PAHs) [2, 17, 27]. The single-particle mass spectra of PAHs can provide indications of the particle source, such as the dominant presence of alkylated phenanthrenes in ship emission particles [4]. In contrast to analyses of the aerosol's bulk composition, single-particle analysis is capable of revealing changes in the distribution of pollutants across the aerosol ensemble. This allows for a deeper understanding of the removal mechanisms in the scrubber and provides an estimate of the potential environmental and health effects of the remaining particles.

Experimental

A single-cylinder, four-stroke, 80-kW research ship engine with common rail injection was operated using heavy fuel oil with 2.4% (m/m) fuel sulfur content, and, for run-in and comparison, marine gas oil with 0.001% fuel sulfur content. With a large displacement of 3.18 L and the capability to run on all marine fuels, this engine is a well-established model for ship propulsion systems [30]. The two investigated loads of 60 kW (75%) and 20 kW (25%) are representative of cruising and maneuvering conditions, respectively. Measurements began after a 20-min stabilization phase to avoid instabilities in the emissions that often occur during the start-up and warm-up phases. A downscaled research wet scrubber (SAACKE, Germany) with a total wash tower volume of 0.9 m^3 was specifically designed and adapted for the 80-kW research ship engine. Due to facility constraints, only open-loop operation was possible. Consequently, Baltic Sea water with a salinity of 10.5 ± 3.6 practical salinity unit and a pH of approximately 8 was used. The aerosol was sampled at 200 °C before the scrubber and at 60 °C downstream the scrubber to avoid condensation, diluted by a factor of 1:100, dried (Model MD-700-12S-1, Perma Pure LLC, U.S.) and guided to the single-particle mass spectrometer. For engine, fuel, and sampling details, see [11] and [3]. In the single-particle mass spectrometer (PhotonLIZA, Photonion GmbH, Germany), individual particles are optically detected, sized and exposed to laser pulses for ion formation. The bipolar time-of-flight mass analyzer detects both anions and cations, providing a chemical profile for individual particles [19, 27]. The instrument utilizes a novel laser excitation scheme, addressing refractory and inorganic components via laser desorption/ionization (LDI) and PAHs via laser desorption/resonance-enhanced multiphoton ionization (LD/REMPI). For details, we refer to [27]. Single-particle mass spectrometry data do not provide exact mass concentration values but instead offers chemical insights at the single-particle level, which enables comparative experiments and identifies characteristic chemical patterns.

Results and discussion

Sum signals and mass spectral signatures

Figure 1 shows the sum mass spectra of each 2000 particles sampled before and after the scrubber for 20 kW load (Fig. 1a) and 60 kW load (Fig. 1c), respectively. The spectra from laser desorption/ionization (LDI, blue) reveal the

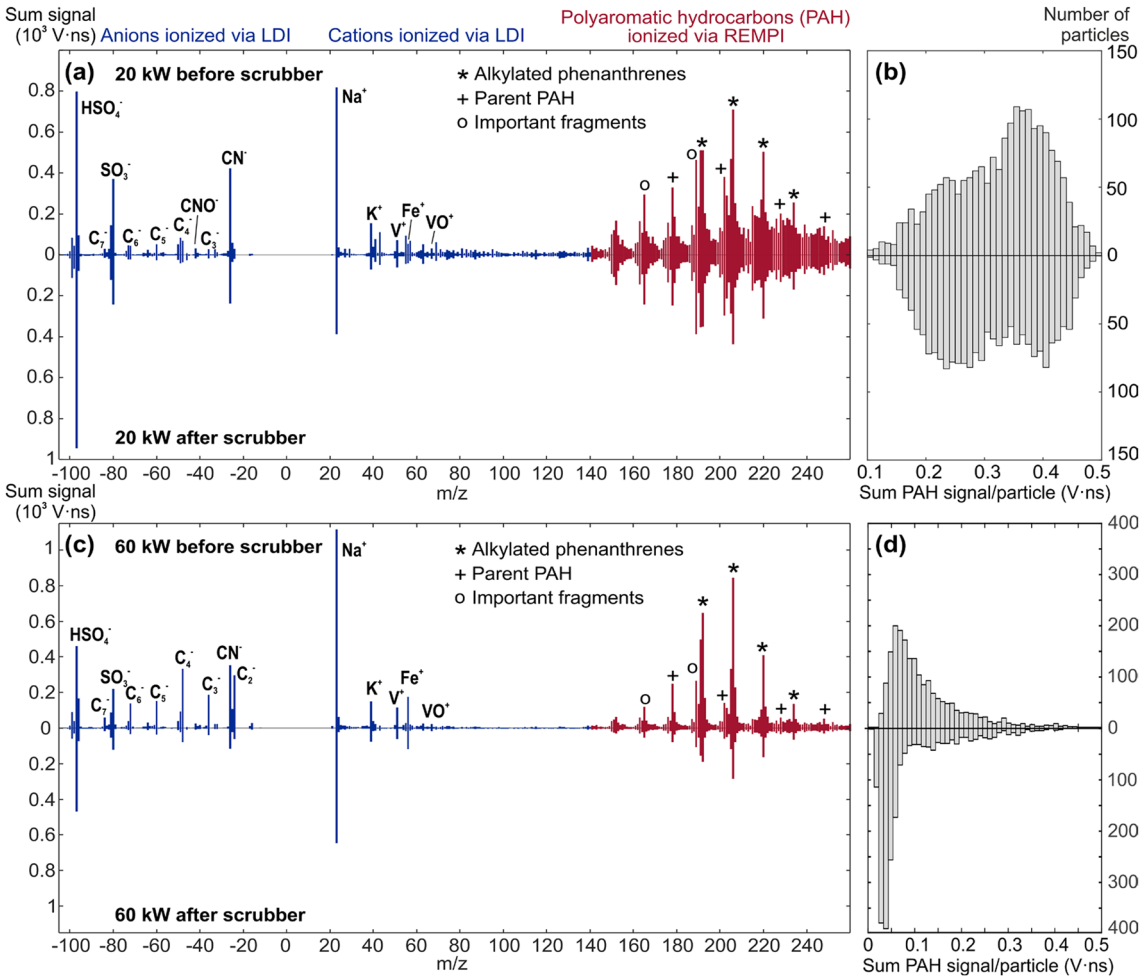

Fig. 1 **a** The summed mass spectra comparison for an equal number of particles (n = 2,000) from heavy fuel oil combustion, measured before (top) and after (bottom) the scrubber at low engine loads, shows no discernible changes in particle composition for either inorganics (ionized via laser desorption/ionization, LDI) or polycyclic aromatic hydrocarbons (PAHs, ionized via resonance-enhanced multiphoton ionization, REMPI). **b** Likewise, the single-particle distribution of total PAH signals remains unchanged after the scrubber. **c-d** At high engine load, the emissions are more soot-dominated. The scrubber reduces predominantly soot and PAH signals, however, without changing the PAH pattern

dominant particle components: sulfate, alkali metals, soot, organic carbon fragments and the typical ship emission markers vanadium and iron [5, 10, 16, 34]. The iron signal is enhanced by resonant laser desorption/ionization [20]. The PAH signatures obtained through LD/REMPI ionization (in red) can serve as an indicator of the particle source [22]. In ship emissions, PAHs primarily originate from unburnt fuel and can therefore be used as a fuel marker, independent of transition metals [3]. The PAH pattern in Fig. 1(a) and (c) is dominated by alkylated phenanthrenes, with its maximum at m/z = 206, which has been linked to heavy fuel oil emissions [4]. Comparing the mass spectra at 20 kW and 60 kW loads reveals stronger contributions of organic carbon and PAHs at 20 kW, while soot signatures are more prominent at 60 kW due to more complete combustion [14]. For comparison, the results for marine gas oil are presented in Supplementary Material (Fig. S1). Size distributions are shown in Fig. S2.

At a 20 kW load, the mass spectra before (Fig. 1(a), up) and after (Fig. 1(a), down) the scrubber are highly comparable. Since the same number of particles is analyzed before and after the scrubber, the mass spectra reveal changes in aerosol composition but not the reduction in particle number concentrations (25% at 20 kW and 38% at 60 kW load; for details, see [11]). Therefore, a slight signal reduction across the entire spectrum after the scrubber may be attributed to instrument performance. The HSO_4^- peak shows a small increase, but due to saturation effects in this mass channel, the SO_3^- signal serves as a more reliable indicator of sulfate content, and it is also reduced. The spectra show no signals of water-soluble organic acids, which can be detected in negative mode with single-particle mass spectrometers

in cases such as aged biomass burning aerosols [37]. The water-insoluble fraction of organic carbon can be assessed through the PAHs. The histogram on the right illustrates the PAH mixing state through the distribution of the summed PAH signals across the particles. Both the distribution and the PAH pattern remain nearly unaffected by the scrubber.

For the elemental carbon (soot)-dominated emissions at a 60 kW engine load (Fig. 1(c)), the scrubber has limited impact on the mass spectra, showing a marked reduction in soot signatures while sulfate signals persist. This aligns with aerosol bulk measurements from the same experiment, which show a reduction in elemental carbon due to the scrubber, while sulfate levels increase [11]. SO_2 is dissolved in the wash water, followed by its conversion to sulfuric acid and ultimately secondary sulfate in the particle phase [6]. The decrease in elemental carbon can be attributed to more effective removal of smaller particles through diffusion-driven coagulation [11, 24], as these smaller particles are often soot particles (see Supplementary Information, Fig. S2(d) and [33]). Alternatively, improved scrubbing efficiency for elemental carbon has been linked to increased soot particle growth from the formation of hygroscopic sulfate on these particles [14]. Nonetheless, this would lead to an increase in elemental carbon signatures in the spectra, given that the instrument is more sensitive to larger particles, which is not observed in this case.

The mass spectral pattern of the PAHs remains unchanged, supporting their effectiveness as fuel marker [4]. However, the histogram in Fig. 1(d) shows a decrease in total PAH signals after the scrubber at higher engine loads, consistent with previously reported PAH reductions in scrubbers [13]. Both the stability of the marker signatures as well as the reduction in PAH levels are confirmed by offline measurements (see Supplementary Material, Fig. S3). The close association between PAHs and elemental carbon indicates that the removal mechanisms for PAHs are likely the same as those for soot.

In summary, for the organic carbon-dominated, relatively large particles emitted at low loads, no substantial changes are observed for the chemical aerosol composition when comparing the same number of particles before and after the scrubber. For the elemental carbon-dominated emissions at high engine load, soot and PAHs are more efficiently removed than other aerosol components, likely due to the smaller particle size of the soot particles [33].

Single-particle analysis

While the summed mass spectra show only little to moderate effects from the scrubber, potential changes in the mixing state and distribution of particle components within the ensemble could alter the environmental effects of aerosol emissions [25] and influence marker-based approaches for source apportionment [4]. In order to analyze the variance of the aerosol composition on a single-particle level, an exploratory principle component analysis was conducted on the mass spectra from LDI (inorganics) and REMPI (PAHs), respectively (see Fig. 2). Data from marine gas oil combustion, sampled before the scrubber, were included for comparison (shown in gray). For both ionization techniques, the first two principal components explained about 70% of the total variance, and thus it creates a representative description of the dataset and variation of mass spectral signatures for different emission scenarios in only two dimensions.

In the scores from the first principal component, heavy fuel oil emissions are well separated from marine gas oil emissions regardless the engine load or sulfur scrubber operation, so single-particle mass spectrometry with LDI at 248 nm is still able to differentiate between these two fuels, mainly by different signatures of Na, Ca, V, Fe and sulfate. The effect of wet scrubber operation on the particle composition exceeds the variation of the engine load by increasing sulfate-related relative ion signals ($m/z = -97$) from hydrolysis-driven gas-to-particle conversion of SO_2. However, the ellipsoids representing the 68% confidence interval of the scores still overlap, highlighting the similarity in particle composition beyond sulfate. In the PAH mass spectra, the impact of wet scrubber operation on heavy fuel oil emission composition is less pronounced, as the distribution of polyaromatic hydrocarbons remains unaffected, and variations in emission composition are typical for all heavy fuel oil combustion emissions.

Moreover, engine load has a greater impact on aromatic emissions from marine gas oil compared to heavy fuel oil, significantly increasing the abundance of alkylated phenanthrenes [3]. However, despite their closer resemblance to heavy fuel oil particle emissions, polycyclic aromatic emissions from marine gas oil combustion at a 20 kW load can still be differentiated from heavy fuel oil emissions using PAH-sensitive single-particle mass spectrometry.

Conclusion

Our single-particle analysis indicates that changes in the chemical composition of particles passing through the scrubber are primarily linked to sulfate uptake. Moreover, our findings suggest that variations in the overall aerosol composition can be attributed to the size-dependent nature of the removal effects: Ultrafine particles with a diameter lower than 100 nm are efficiently removed through diffusion-driven coagulation. As these particles primarily consist of elemental carbon and PAHs, their emissions are notably reduced, especially under high-load conditions during steaming. At low loads, when the aerosol contains high amounts of organics, and for larger particles, no significant

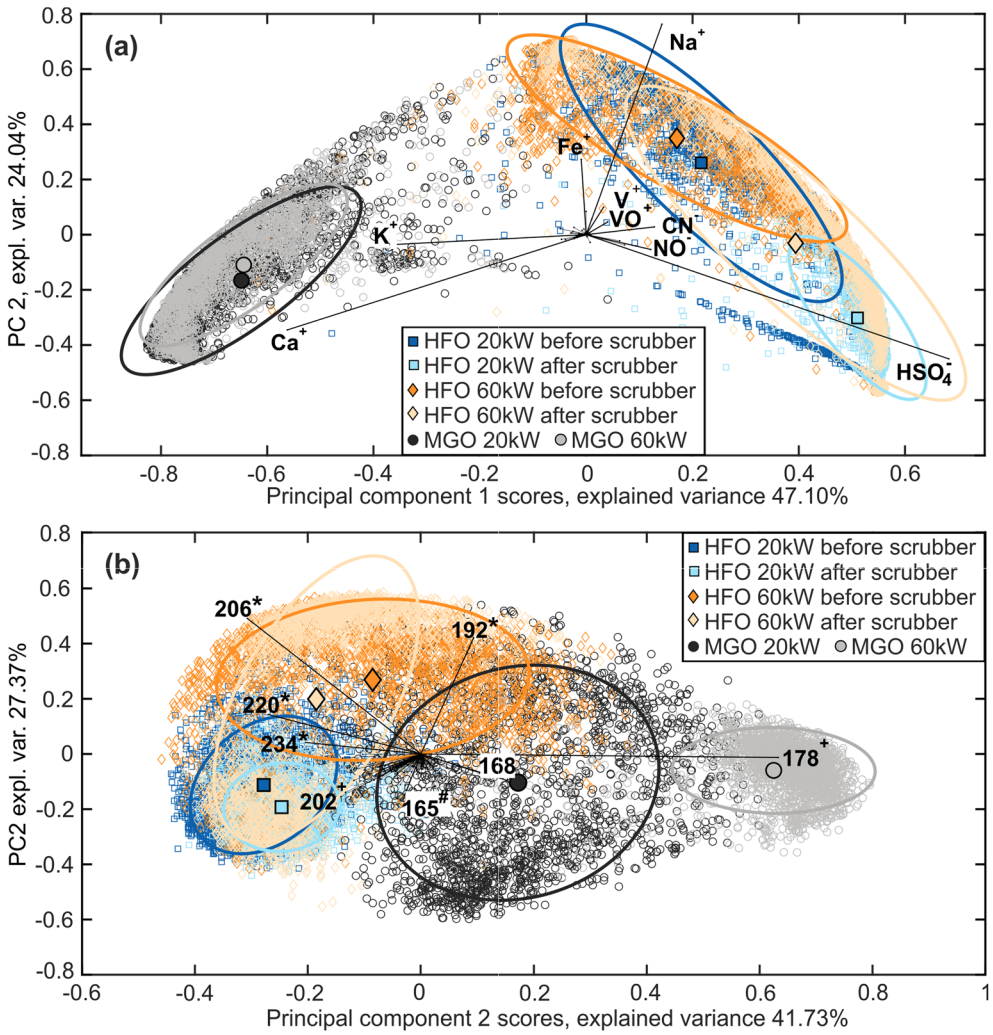

Fig. 2 **a** Principal component analysis of single-particle mass spectra from laser desorption/ionization for 20 kW and 60 kW engine loads, measured before and after the scrubber during operation with heavy fuel oil (HFO). Eigenvector lengths represent the contribution of signals at different m/z ratios to the mass spectral differences. The ellipses depict the 68% confidence interval for the principal component scores of each cluster. For comparison, results from marine gas oil (MGO) combustion, sampled before the scrubber, are shown in gray. The fuel clusters are distinctly separated, with Na, Ca, sulfate, and transition metal signals contributing most to this separation. The only clear effect of the scrubber when comparing the same number of particles is an increase in sulfate signals, due to the gas-to-particle conversion of SO_2. **b** For the polycyclic aromatic hydrocarbons (PAH), small load-dependent effects are observed, but no significant impact from the scrubber is detected. The fuel-specific PAH signatures remain distinguishable. m/z channels: * alkylated Phenanthrenes, $^{+}$ parent PAHs, $^{\#}$ fragment. For each fuel and load condition, n = 2,000 particles were analyzed before and after the scrubber, respectively

reduction in emissions was observed, and the scrubber had no noticeable effect on the aerosol's single-particle composition. However, this size fraction is particularly significant as it often contributes the most to inhalable particulate matter mass and is transported over long distances. Furthermore, since harbors—where ships operate at low engine loads—are located in densely populated coastal regions, exposure to these emissions is particularly high.

The novel ionization technology provides single-particle data on key health-relevant components—metals, soot, PAHs, and salts—but does not capture detailed organic composition beyond aromatic compounds. Schneider et al. [28] reported a scrubber-induced reduction in sulfur-containing water-soluble organics in the high molecular mass range and the formation of new elemental compositions from reactive gas chemistry. Additionally, future studies should focus on full-scale scrubber installations on large ships with two-stroke engines.

Another aspect concerns the environmental monitoring of ship emissions. Our study demonstrates that neither traditional ship emission markers (V, Fe), nor novel PAH-based approaches are influenced by scrubber operation. In fact, ships equipped with scrubbers have already been

detected by their metal emissions over distances of several kilometers [21].

Our findings suggest that using heavy fuel oil with scrubbers in low-emission zones, as an alternative to compliant low-sulfur fuels, may pose an increased health risk due to the insufficient capture of potentially harmful compounds like heavy metals and PAHs.

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Data availability The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Competing Interests The authors have no relevant financial or non-financial interests to disclose.

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