Particle size distribution in ship emissions

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Abstract

There is not much data available regarding particle emissions from ships. In this study the size distribution of particles in ship exhaust was studied using e.g., an impactor. Large emissions and a strong dependence of the sulfur content in the fuel were observed. High amounts of relatively large particles were observed. These are attributed to reentrained soot particles from walls in the engine systems. A strong variation between different ships was observed for the particle size distribution and for the dependence on engine load. The particle emissions were found to be reduced to about half, over the whole size-range, by a SCR system.

1. Introduction

There is a growing concern about the health effects caused by particulate matter (PM) in the air (Forsberg et al., 2005). In many large European cities the levels of PM frequently exceed upcoming air quality limits. It has been estimated that PM cause 100 000 deaths annually in Europe alone (CAFE, 2005). Theses particles can be from natural sources such as soil or sea-salt or may be generated from industry, the energy sector, and, most importantly, the transport sector. In the latter there is a strong focus on road transport. Diesel engines produce large amounts of particles dominated by sizes < 50 nm. These particles are formed from soot (pure carbon), sulfates and volatile hydrocarbon compounds from unburnt fuel and lubricant oil. Further, a large amount of non-engine particles are associated with traffic. These are formed from wear of road and vehicle parts, and from resuspension of dust from the roadside. The latter type of particles dominates if one considers the mass of emitted particles (Omstedt et al., 2005). Also particle emissions from railway traffic, both from diesel engines and wear of brakes, wheels etc., and from airplanes, have received some attention recently.

However, one means of transportation that produces large amounts of particles is shipping. There has been a large focus on emissions of nitrogen oxides and sulfur oxides but fact is that particle emissions from ships may be an even larger health problem. There are a few reasons why these emissions are significant. Many ships use old engines or old engine technology. There has been a strong progress in reducing emissions from truck engines during recent years but this has not yet given a breakthrough for larger engines. Secondly, the fuel quality used for most ships is very poor. Residual oil usually contains very high levels of sulfur, up to 4.5%, to be compared with 10 ppm S for the diesel used for road traffic in most European countries. This leads to the formation of large amounts of sulfate particles. Further, residual oil contains large amounts of polyaromatic hydrocarbons. There is a known

connection between these and the formation of soot particles within the engine. A third factor is the lack of aftertreatment for ship engines. One way to illustrate this is to look at the emission factor. According to Corbett and Koehler (2003) and Eyring et al. (2005), the PM emission factor for ship engines is around 1.25 g/kWh. This can be compared with the regulated value for a modern truck with Euro 5 standards which is 0.02 g/kWh. The total worldwide emission of PM from shipping has been estimated and was found to have increased from 0.39 to 1.67 Tg annually between 1950 and 2001 (Eyring et al., 2005). For comparison, the number for road traffic was about 2.1 Tg in 2001.

The particles emitted from a diesel engine are usually divided into three modes. These are called nucleation mode (10 - 100 nm), accumulation mode (0.1 - 1 μm) and coarse mode (1-10 μm). The nucleation mode is usually dominated by volatile compounds, such as sulfate and unburnt hydrocarbon, which form particles as the exhaust is cooled and diluted, but also contains ash and soot particles. However, most soot particles agglomerate and are usually found in the accumulation mode while the larger particles can be break-up of larger soot particles from the engine. Further, many particles grow as, e. g., volatile compounds are adsorbed onto soot particles forming a complex mixture of particulate matter.

There are not many studies where the size distributions of particles emitted from ship engines are presented. This has probably in part to do with difficulties in performing accurate measurements. Further, there is even less information available regarding how the operating conditions influence the emission of particles and if the size-distribution varies. The operating conditions then include engine load, fuel quality and the use of aftertreatment like catalysts for NO_X reduction.

Cooper (2003) presented PM size distributions from measurements on two ships at berth, measured using either SEM analysis of filters, or a cascade impactor. A relatively large amount of particles were found when considering the mass of particles, compared to what has been observed from heavy-duty truck engines. Sinha et al. (2003) measured concentration of pollutants in a plume with instruments placed in an airplane. They report particle size distribution for accumulation mode particles and observed more particles in the range 0.1 - 0.3 μm for a ship using residual oil compare with a ship using marine diesel.

In this study we report measurements of particle size distributions from three different ships. The measurements where made for different engines and during different operating conditions. We also investigated how an SCR system influences the particle emissions. The particle sizing experiments were performed with a cascade impactor placed in the hot gas inside the exhaust tube.

2. Experimental methods

Measurements were performed during three different campaigns onboard three ships and altogether nine engines. Descriptions of the ships and engines can be found in Table 1. The ships were chosen to give a varying range of conditions and measurements were performed during normal operation. Ship A is a multipurpose ferry operating in Kattegat and the Baltic Sea. For this ship, measurements were made

on one main engine and one auxiliary engine. Measurements were made at different loads and both during manoeuvring and at sea. Ship B is a Ro-Ro ship sailing on the North Sea. Measurements were done on one main engine and one auxiliary engine. The latter was equipped with a SCR catalyst and samples were taken both in front of and behind the catalyst. Ship C is a container ship where measurements were done mainly on the North Sea. However, some measurements were also made during low speed operation on a river. Measurements were made on one main engine and one auxiliary engine. The sampling was in all cases done via three holes in the exhaust manifold. These holes were placed high in the three ships, close to the funnel.

The main purpose of this study is to provide data on the size distribution of particles in the hot diesel exhaust. For this purpose a cascade impactor (Andersen Mark III Particle Sizing Stack Sampler) with eight stages was used. The impactor stages have cut-offs between about 10 and 0.2 μm. The impactor was placed inside the exhaust pipes of the ships, close to centre, during sampling. The nozzle had a diameter of 6.35 mm and the flow through it was around 20 l/min. The filters were analysed gravimetrically and the size distributions were obtained with the use of the software WINCDRS. The filters were kept in closed cylinders and their mass was measured before and after the campaigns.

The total particle mass was measured using glass fibre filters and a partial flow dilution system with CO_2 measurement to determine the dilution factor. The filters were analysed gravimetrically. During the sampling with the impactor a number of other gas constitutes were measured. Nitrogen oxides (Horiba PG-250 chemiluminescence instrument), carbon monoxide and carbon dioxide (Horiba PG-250 NDIR), oxygen (Horiba PG-250 galvanic cell), total hydrocarbon (Bernath Atomic BA 3006 FID), and in some cases sulfur dioxide (Horiba PG-250 NDIR) were measured. The temperature and humidity were monitored as well. Engine data were obtained from the ships' instrumentation. Further details on the measurement methology can be found in Cooper (2003). Fuel samples were taken from the three ships and analysed later.

The sampling with the impactor normally took around 60 minutes. For most measurements steady conditions regarding engine load was kept throughout this period. However, for some occasions measurements were done at varying conditions in order to get an indication of the emissions during manoeuvring and while leaving a port. To measure total particle concentration normally took around 20 minutes. Also this was usually done at steady conditions but sometimes during manoeuvring.

3. Results and discussion

The resulting oil analyses are presented in Table 1. For Ship A different fuels were used for the main engines and the auxiliary engines. Both of these fuels had relatively low sulfur content. For ships B and C similar residual oil with high sulfur content was used.

The size distribution for particles in the hot exhaust was measured at a total of 22 occasions. Figure 1 shows examples of size distributions resulting from analysis of the data. Shown are the mass distributions from four measurements from ship B taken

for different conditions and one example of number distribution (data corresponding to curve 2). The first two curves (1 and 2) are for the main engine. For curve 2, taken at a high load of 91% one can clearly distinguish one large peak at around 5 μm, one peak at around 0.5 μm and a third between 0.1 and 0.2 μm. These peaks correspond to the well-known different modes in the size distribution of particles emitted from a diesel engine. It should be pointed out that the sampling was done at high temperature (186 - 375 ºC) and one can expect that the size-distribution will change as more particles are formed during cooling and also some particles will grow due to condensation of hydrocarbons and sulfates as the temperature of the exhaust decreases. The number distribution is dominated by particles in the nucleation mode.

Curve 1 in Fig.1 shows the size distribution of particles from the main engine during the departure from port. The power from the engine varied significantly during the approximately one hour that the sampling lasted. The purpose was here to get an indication of emissions during manoeuvring close to harbours where the emissions are likely to influence a relatively large number of people. It is interesting to note that the large fraction is significantly smaller in this measurement than for the measurement taken at sea (curve 2 in Fig. 1). This fraction may contain contribution from break-up of soot from the combustion chamber and ash.

In Fig.1 size distributions of particles emitted from one auxiliary engine are also shown. Curve 3 shows the distribution measured before the SCR system and curve 4 the corresponding result after the SCR. In general the two curves are fairly similar but the overall particle emissions was somewhat reduced by the catalyst. This is likely due to combustion of soot particles that takes place over the catalyst.

The corresponding results from the other measurements show similar pattern. However the magnitude of the course fraction was in general much lower for ship C.

Figure 2 shows the accumulated mass from three measurements on the main engines of the three ships. One can see the different modes of PM sizes. For ship C there is a much smaller contribution from particles in the course mode compare to what is the case for ship B.

Table 2 shows a summary of data from the 22 measurements of particle size distributions on the three ships. The temperatures in the last column are the temperatures of the exhaust where the particles were collected with the impactor in order to analyse the size distributions. The total PM (TSP) was measured after dilution at below 52ºC as described above. The other components were measure at ambient temperature.

The measured emissions of NO_X are between 10 and 20 g/kWh with the exception of the engine with SCR where it is an order of magnitude lower. These numbers are normal and no clear trends can be observed regarding a dependence of load. Also the CO and HC values can be considered normal.

The total particle emissions are as mentioned measured using filters and a flow dilution system at < 52 ºC. The measurements will thus include secondary particles as sulfates and hydrocarbons. For ship A, high emissions of PM were observed for the first measurement, taken while the ship was leaving the coast and with varying load.

For the AE on ship A, higher PM emissions were measured at low load than at high load. Ship B used a fuel with higher sulfur content than ship A, and thus the PM emissions are generally higher. The main engine shows the highest emissions for high load. When measured before the SCR system, the exhaust from the AE showed higher emissions for the low load case. After the SCR system, the PM as measured had dropped by more than a factor of two. For ship C, running on a fuel with 2.4% S, both the ME and the AE showed higher PM emissions at high load than at low load. Looking at ships B and C, both using fuel with relatively high sulfur content, the values for the PM emission factor varies between 0.75 and 2.91 g/kWh (not taking the effect of the SCR system into account). The effect of load varies between the engines. For the AE on ships A and B, the PM emissions increase with decreasing load while the opposite was found for ship C. For the MEs there were generally increased PM emissions with increasing load. There is a clear effect of the SCR system on particle emissions probably related to the oxidation of soot over the catalyst. The values found here are in the range of what is previously reported (Corbett and Koehler, 2003, Eyring et al., 2005). Cooper (2003) observed a large variation in emissions from AEs and a strong dependence of sulfur content in the fuel.

The use of PM_{10}, $PM_{2.5}$ and PM_1 is motivated by the regulations for air quality that often target these parameters. However, these parameters may give scattering results with respect to the present measurements. It can be seen in Fig.1 that the peak for the course particles in the measurements show its highest value at values between 2 and 8 μm and that there is a large part at 10 μm. Further, the exact position of the peak varies to some extent between the measurements. This has the consequence that the value at a particle size of 10 μm also varies to a fairly large extent between the measurements making the values of PM_{10} scatter. The same problem is also obvious for $PM_{2.5}$. However, this is an inherent weakness in the definition of these parameters that apply to many measurements of emissions as well as air quality. The average emission factors obtained here for measurements at elevated temperature are 0.38, 0.24 and 0.19 g/kWh for PM_{10}, $PM_{2.5}$ and PM_1, respectively.

There is not much data available about particle emissions from ships, especially not when it comes to size distributions and speciation. The data presented here give some information about the size distribution of particles in the hot exhaust. Since the temperature where the particles were collected was between 200 and 350ºC it is not likely that particles have grown in size through accumulation. Further, it is not expected that there is a contribution from sulfate particles or from volatile organic compounds to the measured particles. However, the amount of nanoparticles can start to increase around 200ºC as reported by Matter et al. (1999). This is not likely to significantly influence the mass distribution, and methods more sensitive to nanoparticles would be needed to study these in detail.

The distribution of particles in the different size modes as measured here differ between the different engines and also depends on driving conditions (i.e., load). These types of variations were also reported by Lyyränen et al. (1999) from laboratory experiments. For most measurements reported here the coarse mode is large when comparing to distributions often reported for smaller diesel engines for trucks etc. These particles are often attributed to accumulation mode particles that have been deposited on different surfaces where they grow and are later reentrained as larger particles (Lyyränen et al., 1999, Kittelson, 1998). The contribution of liquid

particles is probably negligible in these measurements due to the high temperatures. There may however be contributions of ash to the coarse mode. There is most likely a relation between these large particles and the high content of aromatics in the residual fuel which is known to contribute to particle formation (Lee et al., 1998, Kweon et al., 2003). The reentrainment can be expected to be a random procedure and also influenced by the history of the engine as well as by the operating conditions. The reason why ship C in this study in several measurements showed relatively low amounts of course particles could thus be attributed to several factors. One important factor is certainly the fuel quality.

The importance of shipping to local air quality problems is getting increasing attention. The most significant concern may be the levels of particulate matter in cities. For port cities, the contribution from shipping may be considerable. This study shows that large amounts of particles are emitted from ship engines and that the amount and the size distribution both varies significantly between engines and with fuel parameters and operating conditions. Further studies are needed to evaluate, e.g., the importance of engine start-ups etc. In order to control, these emissions it is important to improve fuel quality (reduce sulfur and aromatics) and to introduce emission cleaning e.g., with filters. Further, since the emissions vary significantly between ships, it is important to assess the PM emissions on an individual basis.

4. Conclusions

Particle emissions from three different ships were studied. Large emissions and a strong dependence of the sulfur content in the fuel were observed. The particle sizing measurements revealed large amounts of relatively large particles that are attributed to reentrained soot particles from walls in the engine systems. The size distribution was found to vary between the ships. Further, the dependence on particle emissions on engine load was found to vary between the ships. Particle emissions were also measured before and after a SCR system. It was found that the emissions were reduced to about half over the whole size-range by the SCR system. These results imply that emissions from ships have a large influence on air quality. This is especially true for the air in cities with large ports but the particles sizes observed here also indicate that ther particles emitted from ships can be transported over wide ranges. Further, the size distributions of emitted particles have implications on the influence on atmospheric chemistry by shipping.

5. Acknowledgements

This work was financed by the Swedish Maritime Administration and the Swedish Environmental Protection Agency. Assistance and hospitality from the crew onboard the measurement ships are gratefully acknowledged.

6. References

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Captions

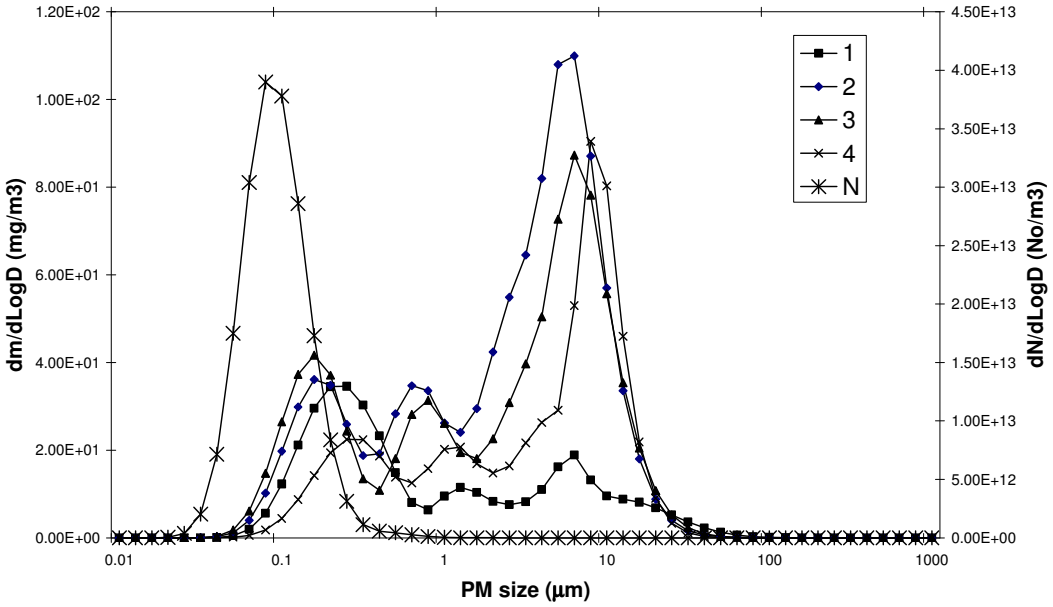
Figure 1
The size distribution from four measurements on Ship B. Curves 1-4 shows the mass distribution while curve N shows the number distribution.

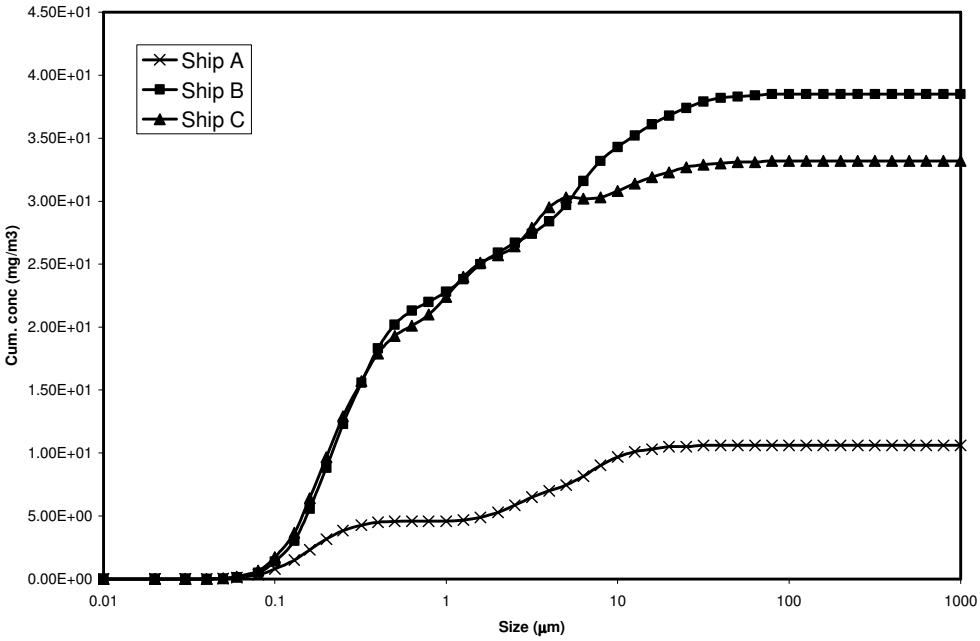
Figure 2
Acckumulated concentrations vs. particle size from measurements from the three ships' main engines.

Table 1
Data for the measurement ships.

Table 2
Results from the oil analysis.

Table 3
Summary of the data from the measurements.


1.20E+02
1.00E+02
8.00E+01
6.00E+01
4.00E+01
2.00E+01
0.00E+00
dm/dLogD (mg/m3)
4.50E+13
4.00E+13
3.50E+13
3.00E+13
2.50E+13
2.00E+13
1.50E+13
1.00E+13
5.00E+12
0.00E+00
dN/dLogD (No/m3)
0.01
0.1
1
10
100
1000
PM size (µm)
1
2
3
4
N


Ship A
Ship B
Ship C
4.50E+01
4.00E+01
3.50E+01
3.00E+01
2.50E+01
2.00E+01
1.50E+01
1.00E+01
5.00E+00
0.00E+00
Cum. conc (mg/m3)
0.01
0.1
1
10
100
1000
Size (µm)

	Ship A	Ship B	Ship C
Type	Multi-purpose ferry	Ro-Ro	Container
Main engines	4x30617 kW	1x20070 kW	1x17500 kW
Auxiliary engines	5x1200kW	4x1720 kW	2*2250 kW
Aftertreatment	-	SCR on AE	-

	Ship A, AE	Ship A, ME	Ship B	Ship C
Density at 15ºC, g cm^{-3}	0.922	0.942	0.991	0.991
Net calorific value, MJ kg^{-1}	44.60	44.27	42.40	42.36
Viscosity at 50ºC, mm^2 s^{-1}	78.3	332	413	375
Carbon, wt%	87.6	87.0	87.1	86.9
Hydrogen, wt%	12.5	12.0	10.4	10.3
Nitrogen, wt%	0.24	0.30	0.47	0.39
Sulfur, wt%	0.33	0.49	2.2	2.4
Oxygen, wt%	<0.3	<0.3	<0.3	<0.3

Ship	Engine	Load	Fuel-S	NO_X	HC	CO	CO_2	TSP	PM_{10}	$PM_{2.5}$	PM_1	Temp
		%	%	g/kWh	g/kWh	g/kWh	g/kWh	g/kWh	g/kWh	g/kWh	g/kWh	ºC
A	ME	var	0.49	12.66	-	3.03	636	2.99	0.20	0.15	0.13	315
A	ME	41	0.49	13.16	-	2.40	634	0.33	0.041	0.023	0.022	350
A	ME	41	0.49	13.15	-	2.26	634	0.27	0.06	0.036	0.028	350
A	AE	48	0.33	13.26	-	2.09	639	0.53	0.18	0.16	0.14	350
A	AE	29	0.33	9.34	-	5.26	640	0.70	0.59	0.48	0.40	290
A	AE	73	0.33	10.5		2.24	638	0.40	0.19	0.13	0.10	375
B	ME	var	2.2	13.6	0.15	4.29	750	1.53	0.43	0.33	0.28	200
B	ME	91	2.2	16.22	0.03	0.66	668	2.12	0.97	0.45	0.31	220
B	ME	72	2.2	14.78	0.12	0.57	666	1.34	0.94	0.44	0.31	220
B	AE before SCR	46	2.2	11.96	0.26	0.66	810	1.51	0.42	0.33	0.29	328
B	AE before SCR	24	2.2	13.1	0.32	1.12	911	2.91	0.80	0.45	0.33	328
B	AE before SCR	48	2.2	10.27	0.31	0.55	779	1.11	0.68	0.35	0.27	320
B	AE after SCR	41	2.2	1.53	-	1.14	789	0.67	0.49	0.23	0.15	330
B	AE after cat	41	2.2	1.53	-	1.14	789	0.82	0.39	0.18	0.13	330
B	AE after cat	24	2.2	2.27	-	1.69	884	0.79	0.48	0.25	0.19	330
C	ME	51	2.4	19.94	0.37	0.35	634	1.61	0.25	0.22	0.19	186
C	ME	76	2.4	15.85	0.34	0.40	634	2.63	0.19	0.19	0.16	285
C	ME	80	2.4	13.75	0.01	0.37	634	2.48	0.27	0.22	0.2	285
C	ME	31	2.4	20.06	0.45	0.44	634	0.93	0.22	0.19	0.16	330
C	AE	40	2.4	11.71	0.44	0.82	634	1.11	0.20	0.17	0.14	275
C	AE	40	2.4	11.77	0.09	0.93	634	0.86	0.21	0.18	0.16	300
C	AE	22	2.4	12.12	0.58	1.20	634	0.75	0.25	0.22	0.19	300