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Characterization of fleet emissions from ships through multi-individual determination of size-resolved particle emissions in a coastal area

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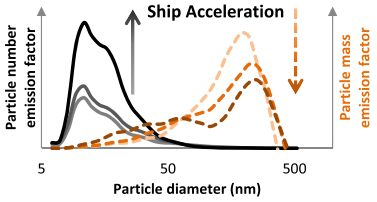
HIGHLIGHTS

- Evaluation of real-world dilution size-resolved emission factors from 154 ships.
- A non-volatile ~10 nm core is suggested to be formed by plume processes.
- Different characteristics of particle emissions for departure and arrival.

GRAPHICAL ABSTRACT



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ABSTRACT

Shipping is becoming a major source of particulate air pollution in coastal cities. Here we describe the use of a stationary measurement site to characterize nanoparticle emissions (5.6–560 nm) from a large ship fleet (154 ships) in a harbor region of an emission control area (ECA) under real-world dilution conditions. Emission factors (EFs) are described with respect to particle number (PN), mass (PM), size and volatility. Automatic Identification System data were used to obtain information on ship class, direction, speed and acceleration. Cargo and passenger ships had similar average EFs: 2.79 ± 0.19 vs. 2.35 ± 0.20 × 10^{16} # (kg fuel)$^{-1}$ and 2550 ± 170 vs. 2200 ± 130 mg (kg fuel)$^{-1}$ respectively. The number size distributions for cargo and passenger ships were unimodal, peaking at ~40 nm. Tug-boats and pilots emitted smaller particles with lower EF_{PN} and EF_{PM}. For emissions of non-volatile particles from cargo and passenger ships EF_{PM} increased with decreasing speed and acceleration while the EF_{PN} decreased. The size distributions of the non-volatile particles for all ships contained a large mode at ~10 nm. This peak is believed to be formed during plume aging. A detailed understanding of size-resolved particle emissions from individual ships will be important in designing appropriate emission regulations for coastal areas.

1. Introduction

Shipping has emerged as an important source of air pollution in coastal areas. Up to 70% of all shipping occurs within 400 km of a coastline, and emissions from ships may be transported several hundreds of kilometres inland (Fuglestvedt et al., 2009). Sulfate, black carbon (BC) and organics are important particulate constituents of ship exhaust emissions (Lack et al., 2009). The IMO (International Maritime Organization) recently revised their regulations to reduce emissions of NO_X and SO_2. However, no regulations regarding particles were introduced but are expected to indirectly be affected by reductions in NO_X emissions and fuel sulphur content (FSC). It has for the year 2012 been estimated that without control of FSC (a scenario with 2.7 wt% FSC), fine particles

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($PM_{2.5}$) would globally cause 87,000 premature deaths. However, if the FSC in coastal areas (i.e. areas within 370 km of land) were restricted to at most 0.5 wt%, the number of premature deaths would fall to 53,500 (Winebrake et al., 2009). For comparative purposes, since the 1st of July 2010 the FSC in emission control areas (ECAs) may not exceed 1 wt%.

Emission inventories are often based on emissions per ship class, which necessitates accurate measurements for each ship class in different environments. The characteristics of the ships and the typical driving pattern differentiate between these classes which also are reflected in their emission profiles (Hallquist et al., 2013; Petzold et al., 2010).

It is also important to develop methods for assessing the effects of introducing new emission control technologies. Selective Catalytic Reduction (SCR) is a technology that could reduce NO_X emissions by more than 90% but might also increase particle emissions by e.g. increasing the oxidation of organic carbon or SO_2 over the catalyst (Jayaram et al., 2011). However, high enough exhaust gas temperature, i.e. engine load, is needed for the SCR to work efficiently (Hallquist et al., 2013). Despite the increasing environmental impact of shipping, there is a lack of size-resolved data on particle emissions from ships. As summarised in a recent paper by Diesch et al. (2013), the currently available measurement data were primarily acquired during on-board or laboratory studies, along with a few chasing studies in which the investigators monitored ships' plumes using research ships or aircraft. In this work, we characterized ship emissions in terms of particle number (PN), particle mass (PM), size and volatility based on measurements of a large ship fleet in a major harbor region using a stationary measurement site (Jonsson et al., 2011). The effects of ship type and driving pattern on both intra- and inter-ship particle emission variability were investigated.

2. Experimental

The measurements were conducted between the 9th of July and the 23rd of August 2010 on an island ~150 m north of the main fairway towards the port of Gothenburg (N57.6849, E11.838) (Figure S1). The site lies within an ECA, hence the FSC was at this time limited to 1 wt%.

Air samples were extracted from a sample inlet that was mounted at a suitable height to capture plumes from passing ships (17 m above sea level). To characterize the particle emissions a method previously used for road vehicle emission measurements was used to derive number and mass emission factors (EF_{PN} and EF_{PM}) from the ship plume measurements, (Hak et al., 2009) Eq. (1).

$$EF_{part} = \frac{\Delta part}{\Delta gas} \times EF_{gas} \quad (1)$$

The Δpart and Δgas terms in Eq. (1) represent changes in the measured particle number, mass, or CO_2 concentration. The EFs were calculated by integrating the peak plume concentration after subtraction of the background concentration, defined as the average concentration during the 30 s immediately prior to plume detection. Elevated concentrations due to plumes lasted typically less than one minute but as long as only one plume was present the calculations were not sensitive to the length of the integration. Particle size-distributions in the range 5.6–560 nm were measured with an EEPS (Engine Exhaust Particle Sizer, Model 3090, TSI Inc.) and total number of particles with a CPC (Condensation Particle Counter, Model 3775, TSI Inc.). CO_2 levels were measured and used as 'tracer' of exhaust gas dilution using a non-dispersive infrared gas analyzer (LI-840, LI-COR Inc.). All instruments collected data with a time resolution of 1 Hz. Samples to be analyzed with the EEPS were first passed through a Thermodenuder (TD, Dekati) operating at 298 K or 573 K in order to determine the ratio of non-volatile particles to total particles in terms of both particle number (R_n) and particle mass (R_m).

Size-dependent particle losses within the TD were accounted for and the resulting corrected values were compared to those determined with the CPC. The EF_{PN} values obtained using these two methods were in very good agreement (slope 0.97 ± 0.05). The CPC was also used to determine the total particle number when the EEPS was being used to characterize non-volatile particle emissions. The particle mass emissions were derived by assuming spherical particles with unit density. The EF_{gas} used in Eq. (1) was 3.2 kg CO_2 (kg fuel)$^{-1}$ (Hallquist et al., 2013). Count median diameters (CMD) and geometric standard deviations (GSD) were calculated from the unimodal or bimodal size-distributions using least square fits, assuming a log-normal distribution.

Data were extracted based on wind sector (105–225° centred perpendicular to the fairway) and wind speed (above 2 m s^{-1}). An individual ship passage was identified as having occurred if the CO_2 increased with at least 2 ppm s^{-1} (the instrument noise was <1 ppm) and no additional plumes were detected within two minutes. For a few passages (6%), uncertainties in the measurement of larger particles (>300 nm) prevented the determination of EF_{PM}. This was attributed to either vessels with low mass emissions or plume dilution.

Automatic Identification System (AIS) data were used to determine the identity, speed, direction and position of the passing ships. An area was defined with two cross sections perpendicular to the fairway. AIS position and speed data for each ship passage were collected at both ends of this area to determine the rate of the passing ship's acceleration/deceleration. In conjunction with information on the current wind speed and direction, these position measurements were used to calculate the age of the ship's plume.

The ship fleet studied in this work was divided into categories based on the ships' AIS classifications. The "cargo ships" category included ships classified as cargo ships (CA), oil tankers (OIL), tankers (CIT), container ships (CON) and roro ships (ROU). The "passenger ships" category included the original AIS passenger ship classification (PA) but excluded a high speed passenger ship, which was placed in a separate category. Tug boats (TUG) and pilot tenders (PLT) were placed in unique categories. The category "others" includes 20 ships that did not fit into the major categories. The ships were further classified according to their engine speed in revolutions per minute (rpm) as either slow speed diesels (SSD; <240 rpm), medium speed diesels (MSD; 240–960 rpm), or high speed diesels (HSD; >960 rpm) with ranges of engine speed as defined by Kuiken (2008).

3. Results and discussion

Calculated EF_{PN} and EF_{PM} for 611 and 574 ship passages, respectively, were associated with 154 different ships that were identified by their IMO or MMSI numbers. The EF values for the 154 ships ranged from 0.14–8.63 × 10^{16} # (kg fuel)$^{-1}$ and 340–5600 mg (kg fuel)$^{-1}$ for total particle emissions and 0.11–4.11 × 10^{16} # (kg fuel)$^{-1}$ and 120–1550 mg (kg fuel)$^{-1}$ for non-volatile particle emissions. The average EF_{PN} was 2.52 ± 0.12 × 10^{16} # (kg fuel)$^{-1}$ which is in line with literature plume data, ranging from 0.2 to 6.2 × 10^{16} # (kg fuel)$^{-1}$ (Alfoldy et al., 2013; Beecken et al., 2014; Cappa et al., 2014; Chen et al., 2005; Diesch et al., 2013; Hobbs et al., 2000; Jonsson et al., 2011; Juwono et al., 2013; Lack et al., 2009; Murphy et al., 2009; Petzold et al., 2008; Pirjola et al., 2014; Sinha et al., 2003). The average EF_{PM} was 2060 ± 120 mg (kg fuel) $^{-1}$ to be compared with a range of 90–4900 mg (kg fuel)$^{-1}$ in recent studies (Cappa et al., 2014;