

Particle Emissions from Ship Engines: Emission Properties and Transformation in the Marine Boundary Layer

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ABSTRACT: In the framework of a combined effort ship emission studies were conducted in 2004. Detailed aerosol microphysics and chemistry was measured in the raw exhaust gas of a single-cylinder test bed engine. The emission studies were complemented by airborne aerosol transformation studies in the marine boundary layer using the DLR aircraft Falcon 20 E-5. In this experiment a single plume of a large container ship was extensively investigated. Observations from emission studies and plume studies combined with a Gaussian plume dispersion model yield a consistent picture of particle transformation processes from emission from a ship engine to atmospheric processing in the marine boundary layer during plume expansion. The results are used for the determination of emission indices of particulate matter from ships and for the estimation of life times of ship exhaust particles in the marine boundary layer.

1 INTRODUCTION

Currently, gaseous and particulate matter emissions from ship engines are gaining increasing attention because of possible environmental and climate impacts (Eyring et al., 2005a, b). Emitted species can considerably influence the atmospheric composition and in particular the ozone chemistry in the troposphere (Endresen et al., 2003). As for any combustion source, ship engine exhaust also contains particulate matter. Ship engine exhaust particles are composed of combustion aerosol particles consisting of elemental and organic carbon, sulphate and ash (Petzold et al., 2004), and of volatile particles forming outside the combustion process in the expanding plume.

Elemental or black carbon (BC) is the most efficient particulate absorber of atmospheric solar radiation and has therefore a strong impact on the atmospheric radiation balance. Additionally, combustion particles can act as nuclei for the formation of cloud droplets and affect by that means the life cycle and radiative properties of marine stratus clouds at the top of the marine boundary layer (Durkee et al., 2000).

In particular the emission of particles and their fate in the marine environment are however widely unknown. Until today, observations reported mainly bulk aerosol properties like mass concentrations (Cooper, 2003), while detailed chemical analyses and aerosol microphysical data are missing.

In the framework of a combined effort, ship emission studies were conducted in 2004 as part of the European Integrated Project HERCULES (**H**igh **E**fficiency **R**&**D** on **C**ombustion with **U**ltra

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Low Emissions for Ships). Detailed aerosol microphysics and chemistry were measured in the exhaust gas of a single-cylinder test bed engine, which was operated at various load conditions, running on fuel with a sulphur content of 3.45 wt.-%.

The emission studies were complemented by airborne aerosol transformation studies in the marine boundary layer as part of the ICARTT-ITOP (**I**ntercontinental **T**ransport of **O**zone and **P**recursors) experiment in 2004. Research flights using the DLR aircraft Falcon 20 E-5 were conducted in the English Channel and in a single plume of a large container ship.

2 METHODS AND RESULTS

On board of the DLR research aircraft Falcon, a comprehensive set of instruments was operated for measuring aerosol microphysical properties of both the secondary volatile aerosol, the primary combustion aerosol and trace gases H_2O , NO , NO_x , O_3 , CO , CO_2 , and SO_2 . The excess CO_2 , or ΔCO_2 respectively, was calculated from the total CO_2 time series by subtracting the average background value from the full CO_2 signal. Since there is no additional source for CO_2 in the vicinity of the investigated ship plume, the value of ΔCO_2 is a very good indicator for combustion emissions. Using a calculated value of 40000 ppm CO_2 in the raw exhaust gas of the investigated vessel, the plume dilution can be determined from ΔCO_2 .

Figure 1 shows the flight track of the aircraft during the plume study in the exhaust of the container ship. The colour of the symbols represents the black carbon mass concentration in the plume. Close to the source, ΔCO_2 exceeded a value of 10 ppm, ranging up to > 100 ppm. Simultaneously, the BC mass concentration reached values of close to $10 \mu\text{g m}^{-3}$, while the Condensation Particle Counters (TSI 3760A) were above their upper detection limit of $20,000 \text{ cm}^{-3}$.

The plume encounters observed during the Single Plume Study are shown in Figure 2. The strength of the plume event was rated according to the CO_2 measured above the background signal (excess CO_2 , ΔCO_2).

The analysis of the measured DMA size distributions in the fresh plume yielded a count median diameter of the combustion particles of 70 nm. Comparable data from emission studies yield a count median diameter of 52 nm. For aged plumes the count median diameter of the combustion particle mode was found at approx. 100 nm. These values reflect a considerable size shift in the particle size distribution by coagulation during the plume expansion from exhaust conditions to an aged plume embedded in the marine boundary layer.

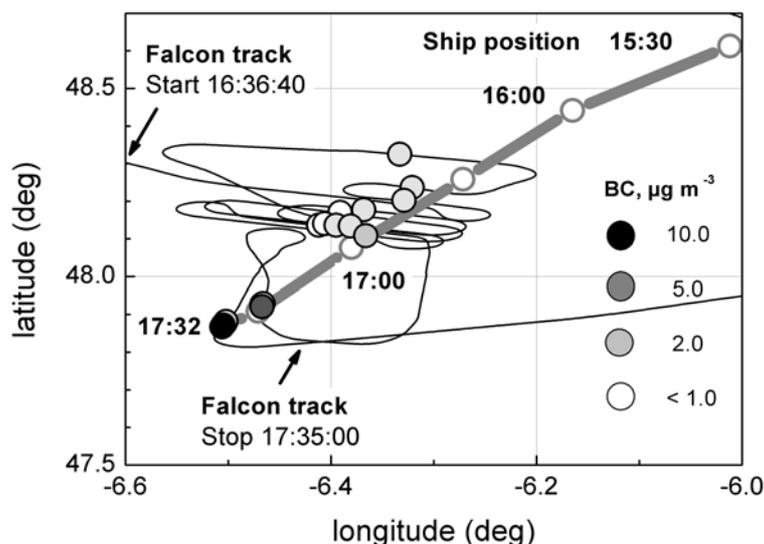


Figure 1. Tracks of the source ship and the research aircraft Falcon during the Single Plume Study; symbol colours represent Black Carbon mass concentrations in the plume.

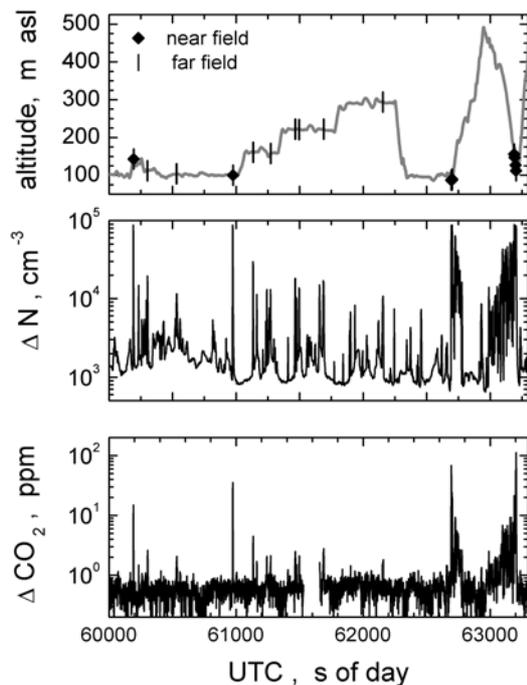


Figure 2. Time series of flight altitude in m above sea level (asl), excess CO_2 (ΔCO_2) and excess number concentration ΔN during the Single Plume Study.

As is shown in Figure 3a, the exhaust particle mode inside the ship plume exceeds the background aerosol in the size range from 20 to 200 nm by max. two orders of magnitude. In the size ranges below and above this range of particle diameters, no deviation from the background aerosol was found. The ship exhaust particle mode was still detectable in polluted air masses outside the single plume. This observation is in agreement with data reported by Osborne et al. (2001). For comparison, Figure 3b shows the size distribution measured on the test bed in the exhaust of an engine which operated at 100% load.

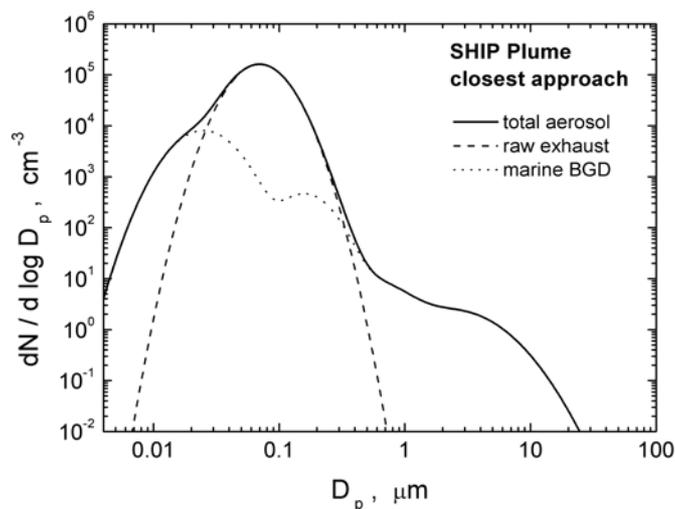


Figure 3a. Composite size distributions from data from DMA, PCASP 100X and FSSP 300 for a strong plume encounter and for a marine background case; the log-normal size distribution represents the exhaust particle mode.

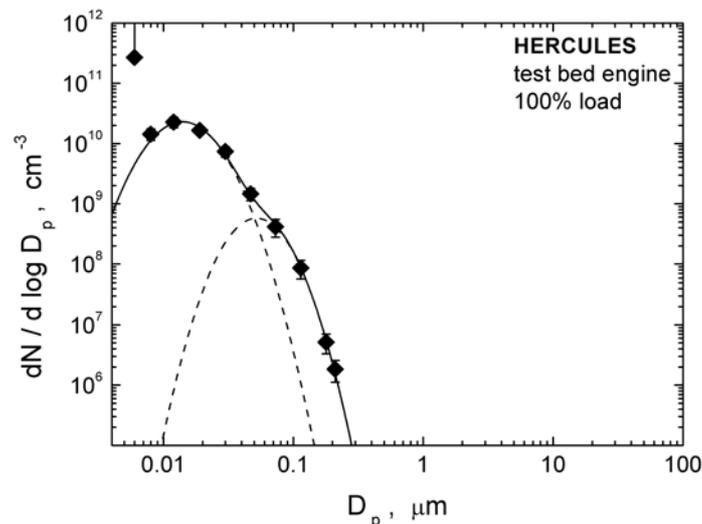


Figure 3b. Size distributions of raw particle emissions measured in the test bed studies.

The plume age during the Single Plume Study was calculated from trajectory analyses of the ship plume with respect to the aircraft flight track and from the geometrical distance from the probing aircraft to the source ship at the time of emission. The CO_2 data measured during the Single Plume Study were then used for the determination of the plume diffusion coefficients according to the plume dispersion model of Glasow et al. (2003). Glasow et al. report best estimates for horizontal and vertical diffusion coefficients of 0.75 and 0.6, while a plume model fitted to the CO_2 measurements yields values of 0.74 and 0.7. When using ΔCO_2 as an indicator for the plume age, plume observations extended from very young plumes in the Single Plume Study ($t \cong 60$ s) to well aged plumes in the English Channel ($t \cong 10,000$ s).

Comparing Figures 3a and 3b, a strong mode of particles with diameters around 10 nm is visible in the raw exhaust data (Figure 3b) while only very few particles are observed in the young plume (Figure 3a). A detailed analysis yielded a ratio $N(D > 4 \text{ nm}) / N(D > 10 \text{ nm})$ of 1.25 at plume ages of about 600 s while this ratio decreased to 1.0 ± 0.1 after about 100 s. Hence nucleation mode particles are expected to live no longer than about 1 h.

Assuming an estimated precision of 0.2 ppm for the determination of ΔCO_2 , the ΔCO_2 signature of a ship plume becomes indistinguishable from the CO_2 background after about 10 h. This value can be used as an average ship plume life time when plume dispersion is the only active dilution process. As soon as turbulent mixing comes into play the life time can be much shorter.

3 CONCLUSIONS

The adapted Glasow plume dispersion model in combination with the observations from emission studies and plume studies yields a consistent picture of particle transformation processes from emission from a ship engine to atmospheric processing in the marine boundary layer during plume expansion:

- Black carbon mass and ΔCO_2 are well correlated for emission and young plume conditions.
- Extensive transformation of particle size distribution properties from exhaust to aged plume observed: count median diameter of the size distributions shifts from 52 nm (raw exhaust) to 70 nm (young plume) and to approx. 100 nm (aged plume).
- Strong nucleation particle mode in raw exhaust; nucleation mode is almost completely depleted in the single plume far field (plume age $> 10^3$ s).
- High abundance of volatile Aitken mode particles in raw exhaust, increased fraction of volatile particles in ship plumes is still visible at plume ages $> 10^4$ s.
- Ship plume reaches top of MBL after approx. 1000 s, earliest onset of ship track effects.
- ΔCO_2 plume signature disappears latest after approx. 10 h.

A detailed analysis of the SHIP Plume Study is under way. Quantitative results on emission factors in terms of mass and number and particle life times will be reported soon in a publication in preparation for *Atmospheric Chemistry and Physics*.

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