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Emissions of NO, NO_2, and PM from inland water transportation

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Abstract

Particulate matter (PM) and nitrogen oxides NO_x ($NO_x = NO_2 + NO$) are key species for urban air quality in Europe and are emitted by mobile sources. According to European recommendations, a significant fraction of road freight should be shifted to waterborne transport in the future. In order to better consider this changed emission pattern in future emission inventories, in the present study, inland water transport emissions of NO_x ($NO_x = NO_2 + NO$), CO_2 and PM were investigated under real world conditions at the river Rhine, Germany in 2013. An average NO_2/NO_x emission ratio of 0.08 ± 0.02 was obtained, which is indicative of ship diesel engines without after-treatment systems. For all measured motor ship types and operation conditions overall weighted average emission indices of $EI_{NO_x} = 54 \pm 4$ g/kg and a lower limit $EI_{PM_1} = \geq 2.0 \pm 0.3$ g/kg were obtained. EIs for NO_x and PM_1 were found to be in the range of 20–161 g kg^{-1} and ≥ 0.2–8.1 g kg^{-1}, respectively. A comparison with threshold values of national German guidelines shows that the NO_x emissions of all investigated motor ship types are above the threshold values, while the obtained lower limit PM_1 emissions just within. To reduce NO_x emissions to acceptable values, implementation of after-treatment systems is recommended.

Keys-words: Air pollution, city air quality, emission factors, guidelines, inland water transport.

1 Introduction

Particulate matter (PM) and nitrogen dioxide (NO_2) are key species for urban air quality in Europe. Whereas the exceedence of PM limiting values has attracted

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considerable public attention during the last decade, NO_2 is a topical problem, which became mature through the introduction of new European limiting values in January 2010.

The reduction of nitrogen oxide (NO_x = NO + NO_2) emissions has been historically one of the key objectives for improving air quality in Europe. NO_x emissions have started to decrease considerably since the mid eighties of the last century in many European areas. However, emissions from mobile sources are still important contributors to air pollution, in particular for NO_x. Together with NO_x, non-methane volatile organic compounds (NMVOCs) undergo photochemical reactions producing secondary pollutants such as ozone (O_3), peroxyacetyl nitrate (PAN) and others (Chameides et al. 1997, Atkinson 2000).

According to the European Commission's White Paper (2011), 30% of road freight transported over more than 300 km distance should shift to other transport modes such as waterborne or rail transport by 2030, and more than 50% by 2050 (European Commission 2011). Accordingly, such a shift will result in an increase of emissions from inland water transportation in the next years.

Today in Germany the contribution of inland navigation to the total freight traffic is about 12% (BDA 2015a). In the Rhine corridor the contribution is 16-18%, respectively (BDA 2015b). With respect to the goods categories ”coal, crude oil and petroleum gas“, ”ores, industrial rocks and minerals, other mining products“ and ”coking plant and petroleum products“ inland water navigation is the most important transportation mode. In comparison to road transport, inland navigation has a contribution of 72% for these goods categories and 52% for container transport. Inland water navigation is a competitive alternative to road and rail transport, because the energy consumption per km and ton of transported goods is only approximately 17% of road and 50% of rail transport (ECT 2015). As a consequence of the lower energy consumption, inland water transportation emits significantly less CO_2 and, therefore, has a direct impact on climate change.

In the EU-15 the emission of NO_x, VOC, PM and CO from road and rail transport decreased from 1990 to 2000, whereas emissions from inland navigation remained more or less constant and emissions from sea transport slightly increased (Trends 2003). However, in the Netherlands a slight reduction in inland shipping emissions were observed in the same time period when modern engines were introduced in the fleet (Adviesdienst Vereer en Vervoer 2003).

It has been also conclusively demonstrated that the fuel has an important impact on the emissions. Using liquid natural gas (LNG) as fuel for inland water vessels leads to substantial emission reductions, i.e. 75% for NO_x, 97% for PM and 10% for CO_2 (Van der Werf 2013).

The emissions from inland water transportation have been regulated by several national and international guidelines. In 2005 the German national guideline “Binnenschiffabgasverordnung, BinSchAbgasV” was implemented for national water ways, defining engine dependent emission indices, i.e. emitted mass of pollutant per kg burnt fuel, for NO_x and PM of EI_{NO_x}: 30 - 42 g/kg and EI_{PM}: 1.2 –

2.4 g/kg, respectively (BinSchAbgasV9 2005). In 2011 an international guideline for the Rhine river "RhineSchUO" was implemented with engine dependent EI_{NO_x}: 28 - 36 g/kg and an EI_{PM}: 0.9 – 3.1 g/kg (RheinSchUO 2011). In addition, for river-sea-ships the MARPOL guideline (International Convention for the Prevention of Pollution from Ships) (IMO 2012) has to be applied. For example, for marine diesel engine with a medium-speed of 720 min^{-1} NO_x-emission indices of 58 g/kg since 2000 (Tier I), 56 g/kg since 2011 (Tier II) and 11 g/kg since 2016 (Tier III) have been introduced.

The correct determination of emission indices (EI) is prerequisite for establishing and developing emission inventories (VBD 2001, Klimont et al. 2002, Browning and Bailey 2006, Rohacs and Simongati 2007, TNO 2008, CBS 2009, UBA 2013). Up to now, several studies have been published in which NO, NO_2, SO_2 and PM emissions from sea ships (Sinha et al. 2003, Chen et al. 2005, Eyring et al. 2005, Petzold et al. 2008, Moldanova et al. 2009, Murphy et al. 2009, Schrooten et al. 2009, Williams et al. 2009, Eyring et al. 2010, Beecken et al. 2014, Jonsson et al. 2011, Lack et al. 2011, Alfödy et al. 2013) and, in particular, from sea ferries (Cooper et al. 1996, 1999, Copper 2001, 2003, Copper and Ekström 2005, Tzannatos 2010, Pirjola et al. 2014) were investigated. Motor test bed studies can also be used for the determination of EIs from single ship's engines (Petzold et al. 2008). However, up to now only three studies have reported on inland water transportation emissions (Trozzi and Vaccaro 1998, Kesgin and Vardar 2001, Schweighofer, and Blaauw 2009, Van der Gon and Hulskotte 2010)

In the present study, inland water transport emissions were investigated under real world conditions at the riverside of the river Rhine in Germany during a field campaign from February 20, to February 22, 2013.

2 Description of the Experimental Procedures

Measurement site

The measurement campaign was carried out at the river Rhine in Germany close to the "Wunderland Kalkar" at Rhine kilometre 843. Figure 1 shows a map of the measurement site. During the campaign emissions from both, upstream and downstream driving inland ships were studied. The sampling point was located 50 m downwind from the river bank.

It is reasonable to assume that the engines of the ships passing the sampling site, were under warm operation conditions.

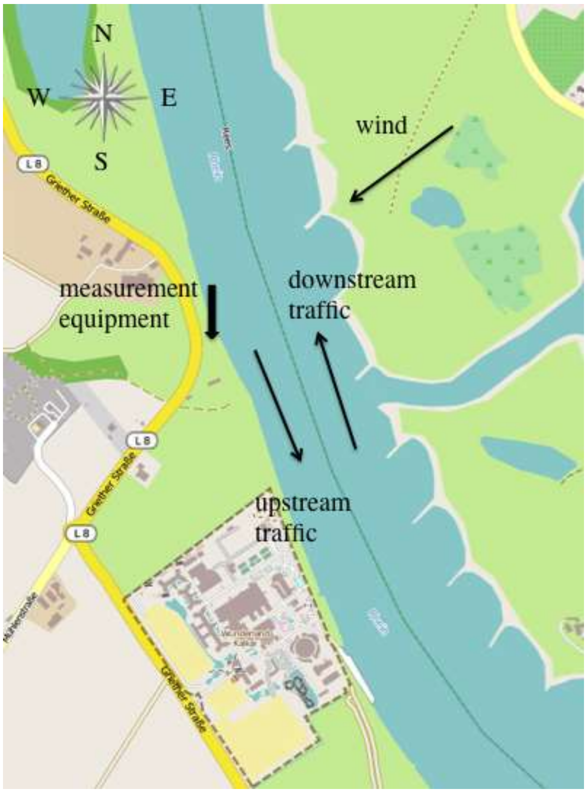

Figure 1: Location of the measurement site at Rhine kilometre 843.
(This map is made available under the Open Database License: http://opendatacommons.org/licenses/odbl/1.0/. Any rights in individual contents of the database are licensed under the Database Contents License: http://opendatacommons.org/licenses/dbcl/1.0/ - See more at: http://opendatacommons.org/licenses/odbl/#sthash.hMw4LgYT.dpuf).

Analytical Equipment

The analytical equipment used was installed in a mobile van with an external power supply. NO and NO_2 were measured on-line with a commercial NO_x chemiluminescence analyzer (Environnemental, AC 31M with molybdenum converter). The time resolution was 10 s and the detection limit, which was calculated from the variation of the zero signal was 2 ppbV for NO and 3 ppbv for NO_2. The NO channel of instrument was directly calibrated by diluted standard NO calibration mixtures (Messer, stated accuracy 5 %). The NO_2 channel was calibrated by using a NO titration unit (Environnemental, GPT). NO_2 was produced by the reaction of NO with O_3 in a flow reactor leading to the quantitative conversion of the calibrated NO ($\Delta NO = \Delta NO_2$).

Ozone (O_3) was measured on-line with a commercial O_3 monitor (Environnemental, O3 41M with UV absorption). The time resolution was 10 s and the detection limit, which was calculated from the variation of zero measurements, was 1 ppbv. O_3 was calibrated by using an O_3 calibration unit

(Environnemental, K-O_3, accuracy 10 %). O_3 was produced by the photolysis of synthetic air in a flow reactor leading to the quantitative formation of O_3.

Carbon dioxide (CO_2) was measured on-line with a commercial CO_2 monitor (LICOR 7100 with IR absorption). The time resolution was 1 s and the detection limit, which was calculated from the variation of background concentrations, was 0.5 ppmv. CO_2 was directly calibrated by diluted standard CO_2 calibration mixtures (Messer, stated accuracy 2 %).

PM was measured by an optical particle counter (OPC) (Grimm Aerosol Technik GmbH, DustMonitor EDM 107). The OPC counts particles in a size range from 0.25 and 32 µm in 31 size-channels. The time resolution was 6 s and the detection limit 0.1 µg/m^3. However, the instrument only provided the concentrations of the fractions PM_1, $PM_{2.5}$ and PM_{10}.

Meteorological parameters, such as temperature, pressure, relative humidity and wind speed were also measured. In addition to the measurement of compounds in the ambient air, the number and types of ships passing the measurement site were counted.

3 Results and discussion

Inland water transportation emissions

NO, NO_2, O_3, CO_2, PM_1 and PM_{10} concentrations, wind speed and wind direction at the measurement site as well as movements of the ships were measured. During the campaign more than 170 emission peaks from motor ships were observed. From these peaks almost 140 could be attributed to single ships and were analyzed accordingly. Figure 2 shows as an example the temporal variation of NO, NO_2, O_3 and CO_2 mixing ratios at the measurement site on February 20, 2013 from 11:30 to 14:00. The perfect correlation between NO and NO_2 with CO_2 confirms that these compounds were emitted from the same source, i.e. the engine exhaust. The anti-correlation between NO_2 and O_3 provides information about NO_x chemistry in the ship exhaust plumes, i.e. the formation of NO_2 by the titration reaction of NO with O_3.

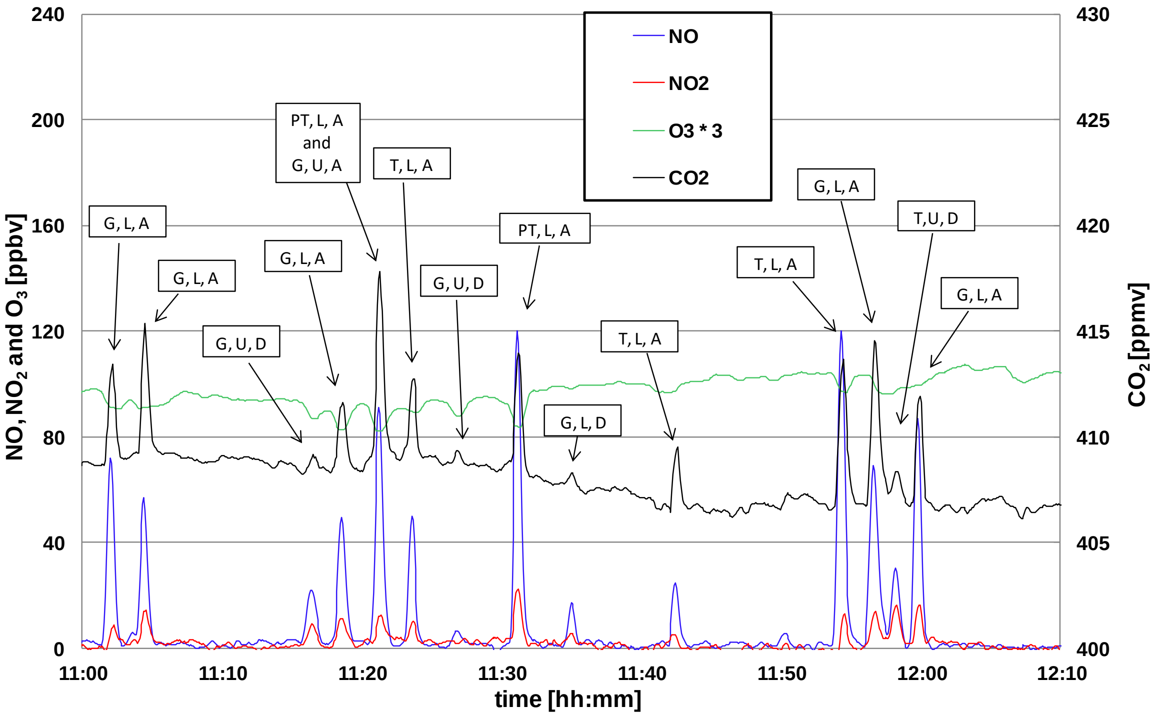

Figure 2: Temporal variation of the NO, NO_2, O_3 and CO_2 concentration at the measurement site on February 20, 2013 from 11:30 to 14:00 from different ship types (G=goods ship, T=petroleum tanker, PT=push tow) and at different operation parameters (L=loaded, U=unloaded, A=upstream and D=downstream).

NO_2/NO_x emission ratio

In order to obtain information about the ships engine types and to estimate the impact of ship emissions on the ozone formation the NO_2/NO_x ratio in the exhaust plume is an important parameter. It is well known that diesel engines without after-treatment systems show NO_2/NO_x ratios of 0.10 – 0.12 for road traffic (Kurtenbach et al. 2001, Kousoulidou et al. 2008, Carslaw and Rhys-Tyler 2013) and (0.14 ± 0.04) for navigation (Cooper 2001, Grice et al. 2009). In contrast, the NO_2/NO_x ratio from road traffic diesel engines with after-treatment systems such as oxidation catalyst or PM filter systems are in the range of 0.25 – 0.30. The NO_2/NO_x emission ratio from navigation diesel engines with selective catalytic NO_x reduction systems (SCR) is (0.009 ± 0.003) (Cooper 2001).

In order to obtain the correct NO_2/NO_x emission ratio from the measurements it is important to distinguish between primarily emitted NO_2 and NO_2, which is being formed by the reaction of NO with ozone in the exhaust plume. The correct NO_2/NO_x ratio is obtained by plotting O_x, which is the sum of NO_2 and O_3 versus the measured NO_x concentration as shown in Figure 3 (Clapp and Jenkin 2001). The NO_2/NO_x emission ratio and the local O_3 background mixing ratio are obtained from the slope and intercept of the regression line, respectively. From the data shown in Fig. 3 a NO_2/NO_x emission ratio of (0.08 ± 0.02) and a local ozone background volume mixing ratio of (23 ± 2) ppbv were obtained. The obtained NO_2/NO_x ratio indicate that the ships passing the measurement site were equipped with conventional diesel engines without exhaust after-treatment.

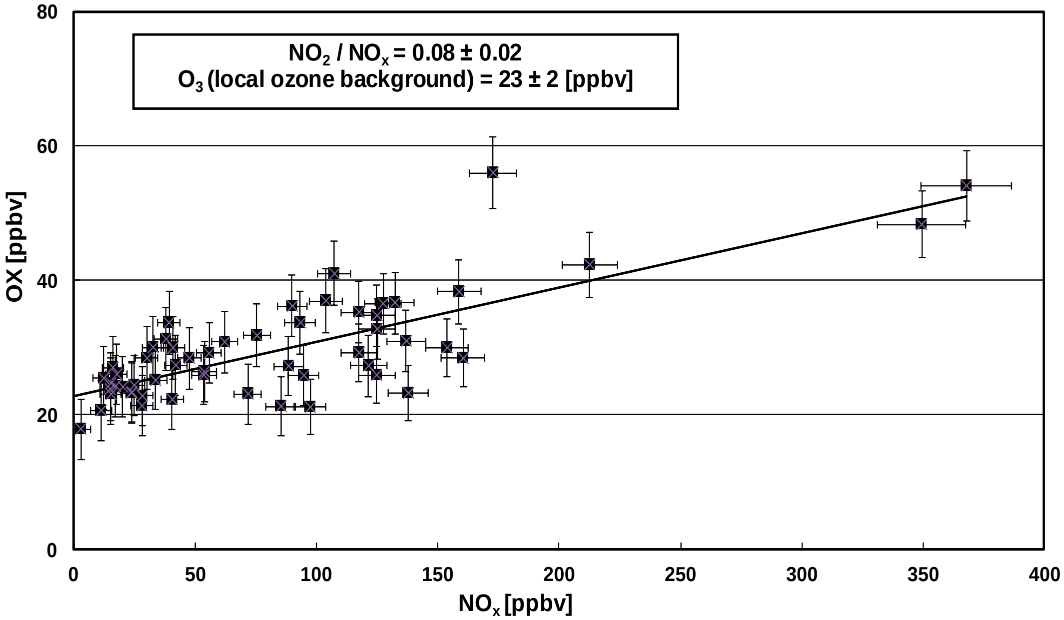

Figure 3: Plot of O_x vs. NO_x

PM_1 and PM_{10} emissions

Figure 4 shows the temporal variation of CO_2, PM_{10} and PM_1 concentrations at the measurement site on February 20, 2013 from 11:50 to 12:10. Some PM_1 peaks are well correlated with those of CO_2 mixing ratios, therefore, with ship plumes. In contrast, some PM_{10} peaks showed no correlation with ship emissions. This indicates that the main PM emissions from ships diesel engines are in the PM_1 range. This result is in good agreement with other studies e.g. from the US-EPA (1996), Petzold et al. (2008), Beecken et al. (2014), Pirjola et al. (2014) and Westerlund et al. (2015). Therefore, in the present study particle ship emissions are defined as PM_1. According to Westerlund et al. (2015) the maximum in the particle number size distribution was observed at about 10 nm and the maximum particle mass distribiution at 250 nm. Therefore the used optical particle counter (OPC) detect only a lower limit of the emitted particle mass.

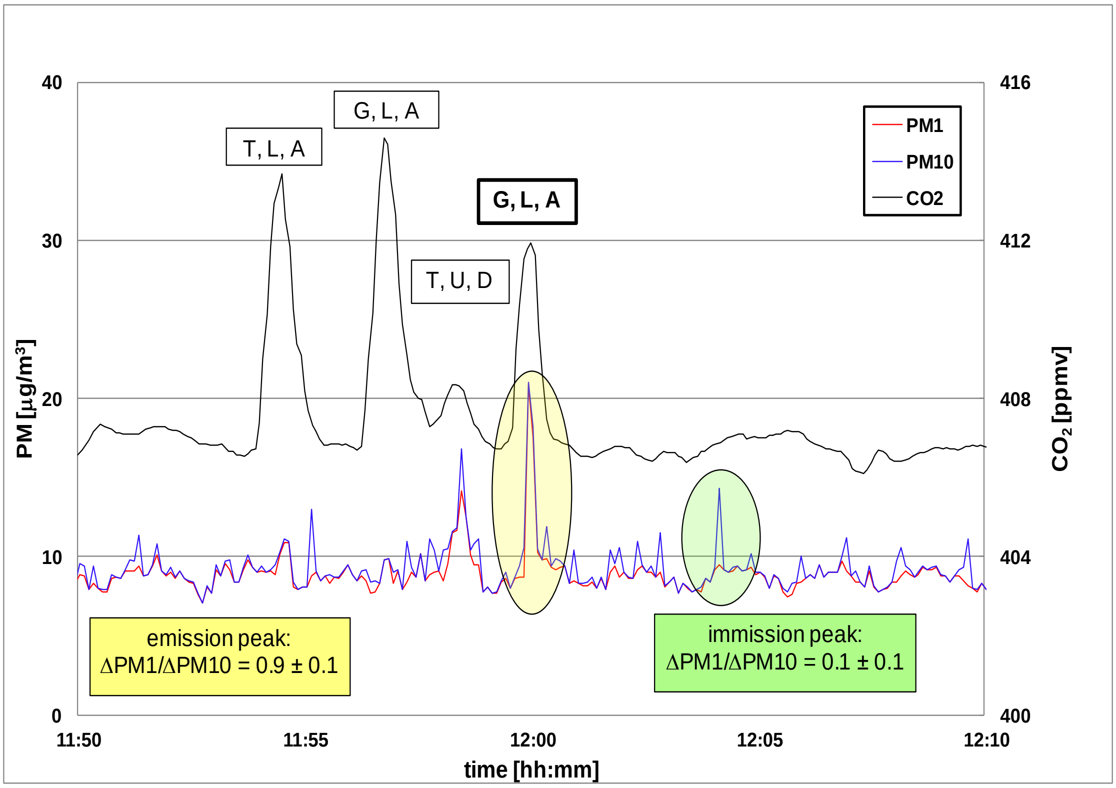

Figure 4: Temporal variation of CO_2, PM_{10} and PM_1 at the measurement site on February 20, 2013 from 11:50 to 12:10 for different ship types (G=goods ship, T=petroleum tanker) and different operation parameters (L=loaded, U=unloaded, A=upstream and D=downstream).

Emission indices

From the measurement data, emission indices (EIs) for NO_x (NO calculated as NO_2) and PM_1 (unit: mass per kg burnt fuel) were calculated. In Figure 5 the integrated emission peak (peak area) for NO, NO_2, CO_2 and PM_1 as ΔNO, ΔNO_2, ΔCO_2 and ΔPM_1 are shown as an example for a single motor ship. If one assumes that the increase of NO, NO_2, PM_1 and CO_2 in the plume is proportional to the emission strength of the ship engine, an emission ratio to CO_2, e.g. $\Delta NO_x/\Delta CO_2$, can be easily calculated (Petzold et al. 2008). In addition the ΔNO, ΔNO_2, ΔCO_2 and ΔPM_1 were also calculated by the difference between background and plume mixing ratios (Schlager et al. 2008) and considering the precision errors of the background data of typically ±1.7 ppbv, ±3.6 ppbv, ±1.4 ppbv, ±0.7 ppmv and ±2 $\mu g/m^3$ for NO, NO_2, O_3, CO_2 and PM_1, respectively.

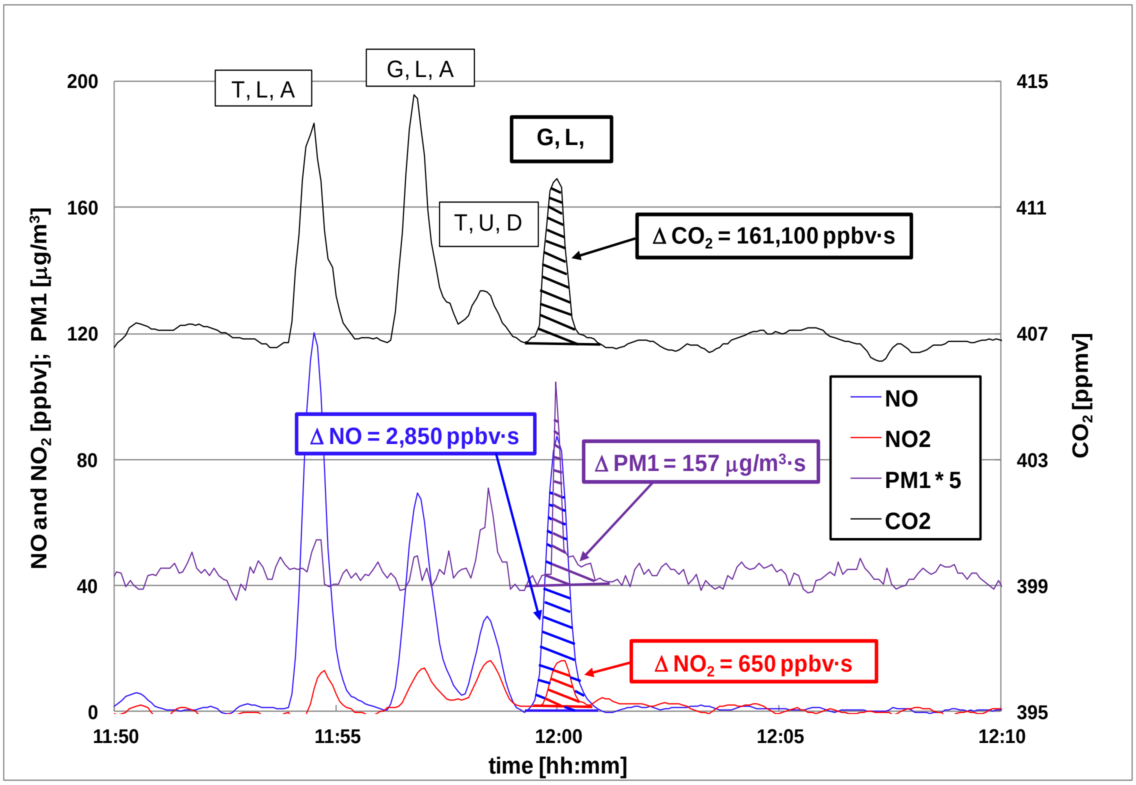

Figure 5: Temporal variation of the NO, NO_2, CO_2 and PM_1 concentration and the integrated emission peaks as ΔNO, ΔNO_2, ΔCO_2 and ΔPM_1 peak area at the measurement site on February 20, 2013 from 11:50 to 12:10 for a goods ship (G) under loaded (L) and upstream (A) conditions.

Both approaches were used to calculate the emission indices and were in good agreement, in general better than ± 6 %. Caused by the slightly different time responses of the instruments, finally the integrated peaks results were specified. Elementary analysis of a typical ship diesel fuel yielded: 86 wt% carbon and 14 wt% hydrogen (Cooper 2001). From the wt% carbon and under the assumption that all fuel is burnt to the final end product CO_2 an emission index EI (CO_2) of 3,150 g CO_2 per kg burnt fuel was calculated and further used to calculate the corresponding emission index (EI) for the ship engines. The emission index (EI) is calculated by the following equation (Petzold et al. 2008):

$$EI(X) = EI(CO_2) \times \frac{M(X)}{M(CO_2)} \times \frac{\Delta(X)}{\Delta(CO_2)}$$

where *M* denotes the molecular weight and *Δ* the peak area, mixing ratios, column densities, etc. of the species. M (CO_2) with 44g/mol and M (NO_x) with 46 g/mol, NO_x as NO_2 were used for the subsequent calculations.

Figure 6 shows as an example the emission index for NO_x (as NO_2) (EI_{NO_x}) of single motor ships [goods] and the weighted average EI_{NO_x} for different operation parameters, i.e. L=loaded, U=unloaded, A=upstream and D=downstream.

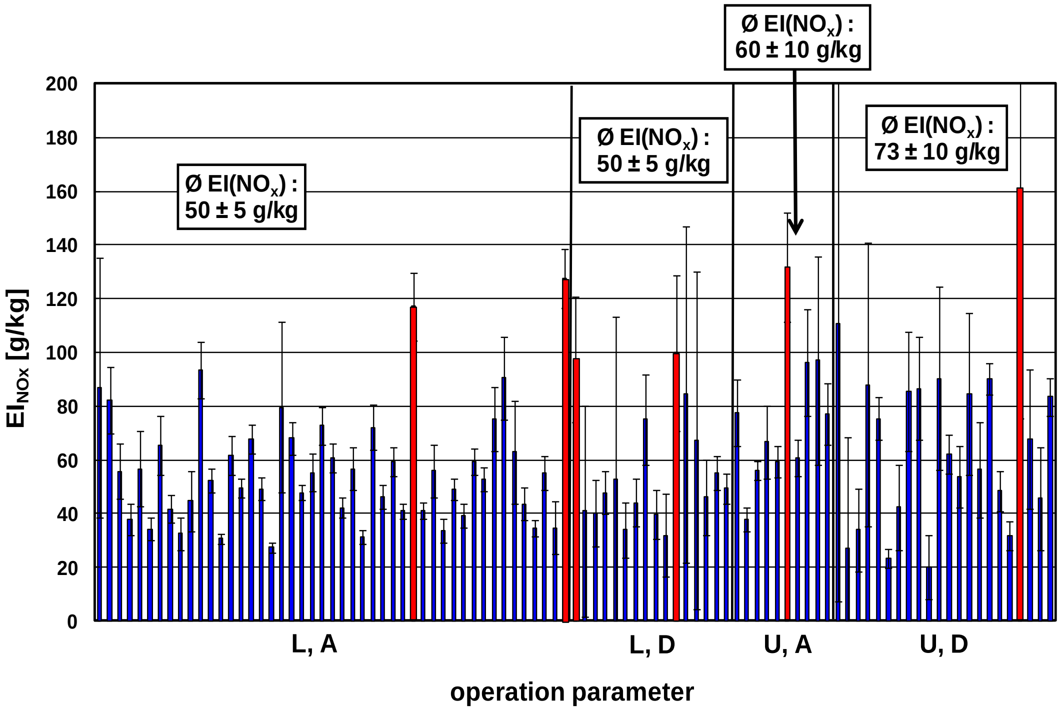

Figure 6: EI_{NO_x} (as NO_2) of single motor ships [goods] and the weighted average value of EI_{NO_x} for different operation parameters, L=loaded, U=unloaded, A=upstream and D=downstream. Red bars show outliers (4σ limit) and were not taken into account in the calculation of the weighted average value.

Figure 7 shows as an example the obtained lower limit PM_1 emission index (EI_{PM_1}) for single motor ships [goods] and the weighted average EI_{PM_1} for different operation parameters, i.e. L=loaded, U=unloaded, A=upstream and D=downstream. Red bars show outliers (4σ limit) and were not taken into account in the calculation of the weighted average value. Values are lower limits because of the detection range of the OPC system.

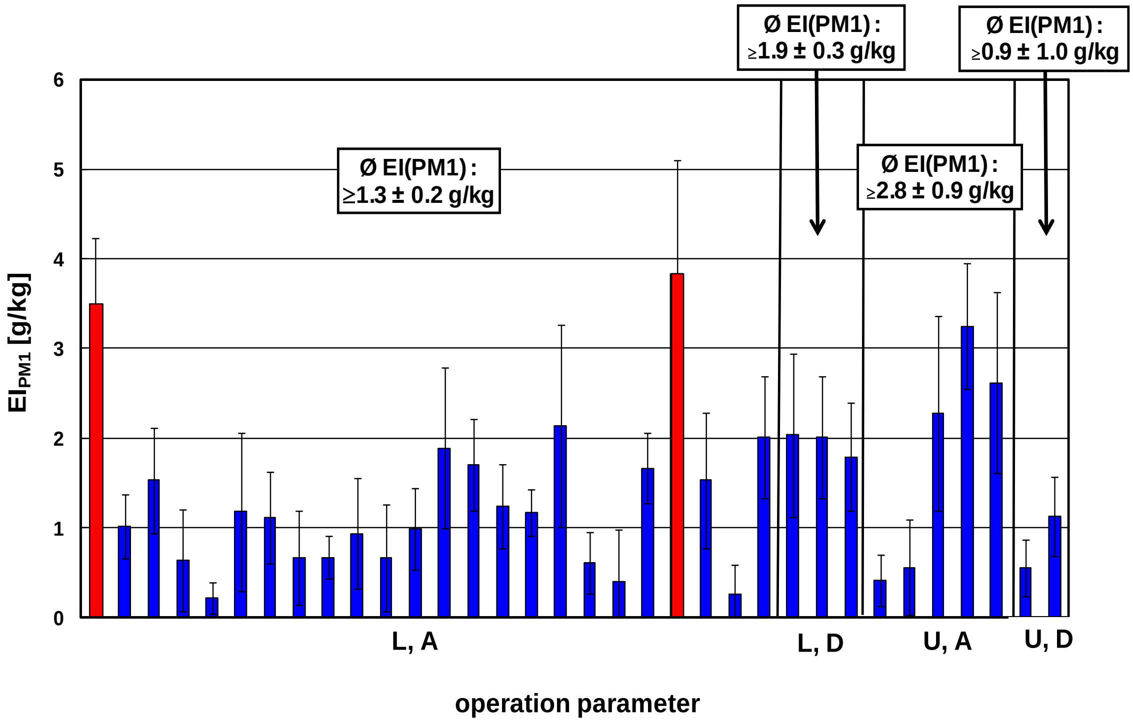

Figure 7: Lower limit EI_{PM_1} of single motor ships [goods] and the weighted average EI_{PM_1} for different operation parameters, L=loaded, U=unloaded, A=upstream and D=downstream.

Although Fig. 6 and 7 show a large variation of the EIs for NO_x and PM_1, the average data exhibit that the EI_{NO_x} are almost independent of engine operation parameters within the given error limits. The same was found for tankers and push tows, see weighted average emission index figures 8 and 9.

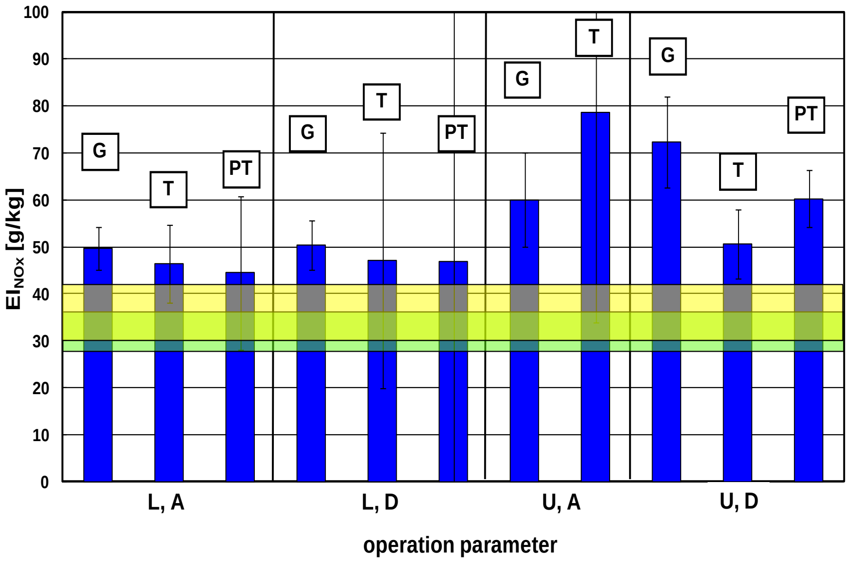

Figure 8: Weighted average emission index for NO_x (EI_{NO_x}) for different motor ship types (G=goods, T=tanker and PT=push tow) at different operation parameters, (L=loaded, U=unloaded, A=upstream and D=downstream) in comparison with German guide lines (BinSchAbgasV 2005 [yellow] and RheinSchUO 2011 [green])

Figure 8 exhibits that the NO_x emission indices of all motor ship types investigated are above the engine rotation speed dependent limit values of the German guide lines, which are 29 – 37 g/kg for the RheinSchUO and 36 – 46 g/kg for the BinSchAbgasV guide lines.

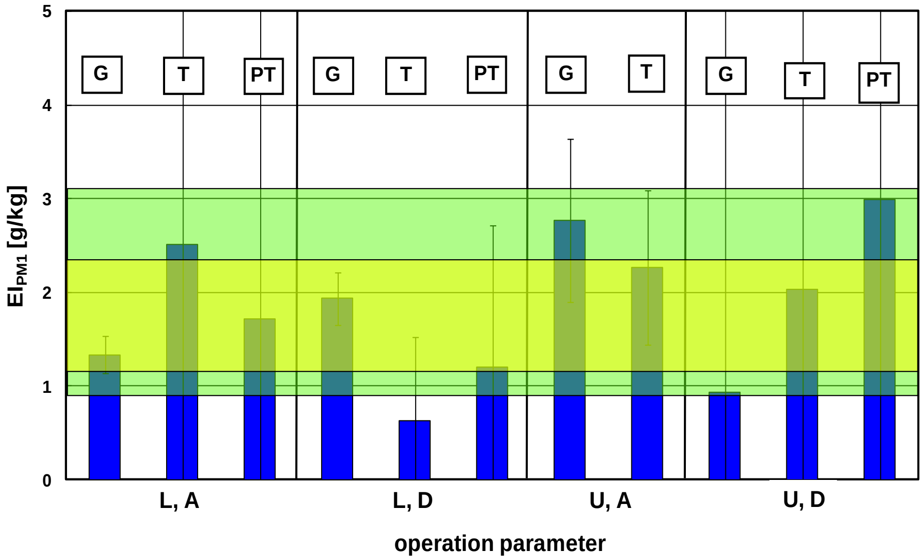

Figure 9: Weighted average lower limit emission index for PM_1 (EI_{PM_1}) for different motor ship types (G=goods, T=tanker and PT=push tow) at different operation parameters, (L=loaded, U=unloaded, A=upstream and D=downstream) in comparison with German guide lines (BinSchAbgasV 2005 [yellow] and RheinSchUO 2011 [green]).

Figure 9 exhibits that the obtained lower limit PM_1 emissions values for almost all motor ship types are just within the limit values of the German guide lines, which are 0.9 – 3.1 g/kg for the RheinSchUO and 1.2 – 2.4 g/kg for the BinSchAbgasV guide lines depending on the engine rotation speed.

For comparison with literature data, uncertainty(2σ)-weighted averaged EI_{NO_x} and EI_{PM_1} were calculated for all motor ship types and operation condition investigated. An EI_{NO_x} of 52 ± 3 g/kg and a lower limit EI_{PM_1} of ≥ 1.9 ± 0.3 g/kg were obtained. Minimum and maximum EIs for NO_x and PM_1 were found to be in the range of 20 – 161 g/kg and ≥ 0.2 – 8.1 g/kg, respectively. Table 1 show the emission indices NO_x and PM_1 in g/kg fuel calculated from the measured values in comparison with different literature data. Errors were calculated using error propagation for the different measured compounds.

Between 1998 and 2013 only a few studies reported EI_{NO_x} and EI_{PM_1} from inland water navigation (Trozzi and Vaccaro 1998, Kesgin and Vardar 2001, Schweighofer, and Blaauw 2009, Van der Gon and Hulskotte 2010) in the range 39 – 57 g/kg and 0.7 – 1.9 g/kg, respectively, see table 1. The uncertainty(2σ)-weighted averaged EI_{NO_x} and EI_{PM_1} were 48 ± 4 g/kg and EI_{PM1} 1.3 ± 0.2 g/kg, which are in good agreement with the present study.

Emission indices for NO_x and PM_1 from inland water navigation have been used in emission inventories by Klimont et al. (2002), Rohacs and Simongati (2007), TNO (2008), CBS (2009) and UBA (2013). The authors reported EI_{NO_x} and EI_{PM_1} in the range 46 – 51 g/kg and 1.5 – 4.0 g/kg, respectively (see table 1). From these data uncertainty(2σ)-weighted average values for EI_{NO_x} of 48 ± 2 g/kg and EI_{PM_1} 2.7 ± 1.2 g/kg were derived, which are in a good agreement with the present study.

In order to comply with the limit values of the current RheinSchUO guide line for inland water navigation for NO_x with 29 – 37 g/kg a further significant reduction of the NO_x emission is necessary. This can be achieved e.g. by using exhaust after-treatment systems, whose functional capability have been demonstrated in recent studies (Cooper 2001, Schweighofer and Blaauw 2009, BMVBS 2012, Future Carrier 2012, Hallquist et al. 2013, Pirjola et al. 2014). For example, the European project “The cleanest ship” (Schweighofer and Blaauw 2009) shows that NO_x and PM emission of a ship diesel engine equipped with an SCR (selective catalytic reduction) system and particle filter can be reduced to 4 g/kg and 0.02 g/kg, respectively.

Table 1: Emission indices NO_x and PM_1 in g/kg fuel calculated from the measured values in comparison with different literature data.

Reference	**Location**	**Sampling period**	**EI_{NO_x} [g/kg]**	**EI_{PM_1} [g/kg]**	**Ship types**
A) field measurements (inland, engine without after-treatment system)					
This study	Germany, Rhine (inland)	2013	54 ± 4	≥ 2.0 ± 0.3	different
Kesgin and Vardar (2001)	Turkey; Bosporus (inland)	1998	57	1.2	domestric passenger ships (a)
Trozzi and Vaccaro (1998)	Italy, Tyrrhenian Sea (inland)	1998	51	1.2	domestric passenger ships (a)
Van der Gon and Hulskotte (2010)	Netherlands (inland)	2010	45	1.9	different
Schweighofer and Blaauw (2009)	inland	2009	39	0.73	research vessel (b)
B) field measurements (inland, engine with after-treatment system)					
BMVBS (2012)	inland	2011	n.d.	0.08 – 0.48	research vessel
Futura Carrier (2010)	inland	2009	n.d.	0.29 ± 0.01	research vessel
Schweighofer and Blaauw (2009)	inland	2009	11 - 39	0.02	research vessel (c)
C) inventories					
Rohacs and Simongati (2007)	Average EU (inland)	2007	47	3.2	inventory
TNO (2008), CBS (2009)	Netherlands (inland)	2008-2009	46	1.9	inventory
Klimont et al. (2002)	RAINS, EU (inland)	2002	51	4.0	inventory
UBA (2013)	TREMOD, Germany (Inland)	2013	49 ± 6	1.5 ± 0.2	inventory

Remarks: n. d. no data, a) domestic passenger ships with diesel engine (medium-speed), b) without after-treatment system, c) with after-treatment system

4 Summary and Conclusion

The present study has shown that the measurement site at the Rhine river provided representative real world emission data from inland navigation. Emissions of NO, NO_2, CO_2, and particulate matter from a large number of individual ships were monitored and analyzed.

Particulate emissions measured in the ship plumes were dominated by PM1. An average NO_2/NO_x emission ratio of 0.08 ± 0.02 was obtained, which is typical for ship diesel engines without after-treatment systems such as oxidation catalysts or PM filter systems.

The emission indices for NO_x (EI_{NO_x}) and PM_1 (EI_{PM_1}) determined for different motor ship types (cargo, petroleum tanker and push tow) and for different operation parameters (L=loaded, U=unloaded, A=upstream and D=downstream) exhibited a large variation and were almost independent of the ship types and operation parameters. For the motor ship types and operation conditions investigated a weighted average EI_{NO_x} of 54 ± 4 g/kg and lower limit EI_{PM1} of ≥ 2.0 ± 0.3 g/kg was obtained with minimum and maximum values ranging from 20 – 161 g/kg for NO_X and ≥ 0.2 – 8.1 g/kg for PM_1, respectively. The EI_{NO_x} and EI_{PM_1} from the present study are in a good agreement with literature data.

The comparison of emission indices for NO_x and PM_1 with limit values of the German Guidelines (BinSchAbgasV 2005, RheinSchUO 2011) showed that NO_x emissions of all motor ship types investigated were above the limit values whereas the obtained lower limit PM_1 emissions for almost all motor ship types were just within the limit values.

In order to meet the limit values for NO_x und PM, in particular the NO_X emissions have to be reduced significantly, e.g. by the introduction of specific exhaust after-treatment systems, some of which have been proven to be very effective.

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