Atmos. Meas. Tech. Discuss., 5, C4130–C4141, 2013
www.atmos-meas-tech-discuss.net/5/C4130/2013/

Atmospheric Measurement Techniques Discussions

Interactive comment on "Measurements of air pollution emission factors for marine transportation" *by* B. Alföldy et al.

B. Alföldy et al.

balint.z.alfoldy@gmail.com

Received and published: 24 April 2013

The authors thank the Referee his/her efforts to correct the mistakes and the critical points and new ideas for increasing the scientific level of the paper. Detailed responses to the Referee can be found below.

"...very few details are given about the campaign. I was not able to ïňĄnd the start date or end date in the text, and an overview of meteorological conditions is entirely lacking."

The required details for the campaign have been added. We have summarises the meteorological conditions in a new figure (Fig. 6) and added a new phrase on Pg 8929 about the dates and a new chapter (2.2.) on 8930-8931 about meteorological

conditions and sampling strategy of the campaign.

"Then, the sampling locations (of the instruments) were changed in order to 'catch the plume' of the ship given the wind direction. The authors need to explain their measurement design in terms of distance to the ships, the time the plume travelled towards their instrumentation, and the possible effects of dilution because of non-linear chemistry occurring in the plume. As demonstrated by aircraft measurements that repeatedlyampled ship exhaust plumes –and conïňĄrmed by model studies– the (instantaneous) lifetime of SO2 and NOx within an exhaust plume is very long in the ïňĄrst stages after emission, because OH is strongly depleted within the plume. At a later stage, when the plume dilutes (depending on the stability boundary layer), the instantaneous lifetime of SO2 and NOx is reduced to below background-levels (e.g. Chen et al., 2005). In other words, the exact concentration of SO2, NO2, and NO measured depends very much on the time spent in the plume and on the diffusive character of the plume. These effects inïňĆuence the ratio of SO2 or NOx to CO2 (which is chemically inert in the plume) and therefore the emission factors. I don't think the authors have taken this into account (and I did not see how the brief discussion of Figure 15 helped). I think the authors should revise their manuscript and take these effects into account when interpreting the observed concentrations as representative for a certain ship type/engine/r.p.m."

The Referee mentions that the chemical conversion of SO2 and NOX in the plume may have an impact on the calculated emission factors for SO2 and NOX. We agree that this issue should be discussed in the paper and have thus added a paragraph on Pg 8936. The residence time of the plume in the atmosphere before reaching the measurement points has been calculated from the measured distances from the ships to the sampling points and the measured wind speeds; the average is 101 seconds and the maximum lies below 15 minutes. We added a new figure about the time distribution of plume age (Fig. 5), and a new paragraph (Pg 8930) with a related figure (Fig. 4) where we describe how the plume age was determined. The text on Pg 8936 has been completed with the following paragraph:

»The potential influence of chemistry on the measurements can be estimated based on the work by Chen et al. (2005), who studied the conversion of NOx and SO2 in a ship plume during daytime on the 8th of May, 100 km off the California coast and found a significantly reduced atmospheric lifetime of NOX, which was found to be as low as approximately 1.8 hours. The lifetime of SO2 is longer and is mainly depending on heterogeneous reactions that are largely independent from plume chemistry. Taking into account the different geographical location and time of the year we consider the NOX lifetime measured in the Californian experiment to be an upper limit to that of the experiment in Rotterdam. Further, it must be taken into consideration that the NOX lifetime in the initial phase of the plume development is likely to be relatively long due to depletion of OH. Assuming a NOX lifetime of 1.8 hours a 101 seconds plume residence time will lead to an underestimation of the NOX emission factor by less than 1% which we consider to be negligible. If the residence time is 15 minutes the underestimation will be 13%.«

“Section 2.2 reads rather like a company’s speciïňĄcation of the instrument, than as a scientiïňĄc account of measurement approach and associated uncertainties. The authors should explain better how the measurements have been taken, and what the relevant error characteristics are. It seems as if the SO2 instrument measures the ïňĆuorescent radiation of the ambient air, simply waiting for the plume to hit the instrument, but what is the spatial footprint of such a measurement? What process generates the ïňĆuorescence and why is that speciïňĄc to SO2 in the air? Stating the instrument accuracy as 10% is not informative enough of the error characteristics of such an instrument. How have similar instruments performed against independent measurements? How does the instrument function in the dynamical range of concentrations encountered in the Rotterdam study? What is the detection limit, relevant to determine the background concentrations?”

We do not find necessary to modify this section, since no any special instrument was applied during the measurements. The principals of the applied gas analysers are

broadly known, or can be reached in other sources. We think that there is no sense to analyse the measurement errors more detailed, since four independent measurements were taken. Each instrument was calibrated and verified prior the campaign. We did an intercomparison between the groups on the first day, and we repeated it twice during the campaign. We gave the mean and the standard deviation of the calculated emission factors originated from the four independent measurements, and as we found the resultant error is more than the double of the measurement uncertainty of the individual instruments. Errors mostly generated by the net area calculation of the emission peaks and the baseline consideration of CO2. We think that the given SD values for SO2 and NOX EFs (23%, 26%) coming from the comparison of the four independent measurement represent enough the reliability of the EF data. We also analysed the SD values for SO2 and NOX EFs of returning ships (30%, 34 %) that could be considered as an upper limit of the uncertainty of the measurement. The question of the detection limit of the analysers is not relevant here, since background concentrations were measured by ambient gas analysers.

“For NOx, it is well known that the use of molybdenum converters can lead to substantial high biases in the reported NO2 concentrations because also species such as HNO3 and PAN are converted on the molybdenum surface. Although I think the fresh NOx will dominate over these potential interferences when sampling the plume, the background values may well be susceptible to these errors. Since the background is so important when determining the EFs later, such effects should receive much more attention in the paper. In short, the paper needs to improve by discussing the error budget for the different measurements not as factory speciïňĄcations, but as relevant for Rotterdam conditions.”

We do not debate the possible interference of HNO3 and PAN with NOX measurement, especially in case of the background measurement. Unfortunately we cannot correct these effects due to the lack of HNO3 and PAN concentrations. However, since the plume NO2 concentration is much higher than the background, errors of NO2 baseline

concentration slightly modified the calculated EF. The EF calculation was much more sensitive for the CO2 baseline consideration due to the lower plume-to-background ratio of CO2. This latter is dominating in the uncertainty of EF calculation, within the mentioned interference is negligible. To make this situation clearer, the following sentences have been added to Section 3 :

» It should be mentioned that the main uncertainty of EF calculation come from the way of CO2 baseline consideration. Calculation of the net peak area for CO2 was very sensitive for the baseline due to its high background value.«

SPECIFIC COMMENTS

“The title is too generic. The paper is actually about determining SO2, NOx, and PM emission factors based on measurements in September 2009 in Rotterdam, The Netherlands. Unless the authors can ïňĄrmly prove that their results have universal validity, the title should reïňĆect the speciïňĄcs.”

The title has been specified.

“P8927, L28-29: MARPOL Annex VI is mentioned twice in the same context. Please rephrase.”

Done.

“P8928, L15-20: please provide some more detail on how the Tier I, II, and III will reduce the NOx emissions from ship engines.”

Detailed information about the technical aspects and expected impacts of the IMO regulations on NOx emissions can be found in a study published by IMO (2009): MEPC 59/INF.10, ‘Prevention of air pollution from ships’, IMO, 2009. A reduction of 12-14% of NOx emissions per tonne fuel for ships regulated by Tier is expected; this would cause a total reduction of NOX emissions from ships of about 6% compared to an unregulated situation. This reference has been added.

“P8929, L1: “statistically representative studies”. The authors should deïňĄne what in their eyes is a ‘statistically representative’ or ‘signiïňĄcant’ ïňĆeet.”

“Statistically representative sample or fleet” provide explanatory statistical distributions. We obtained quasi Gaussian distributions, which means that the fleet was statistically representative. We could also calculate p value for the points plotted in Fig. 13.

“P8929, L22: I think the authors should provide the start and end date of the campaign.”

Done.

“P8933, Eq. (1): please elaborate a bit more on the deïňĄnition of the EF. In any case you must be pretty certain that 87% of the ship fuel consists of carbon.”

See the reference (Cooper et al., 2005).

“P8935, Eq. (3): why isn’t the NO concentration weighted with its molecular mass of 30 but instead with 46 which holds for NO2? Does this have to do with expressing it as “NO2 equivalent”? If so, why would one want to do that?”

Yes, we gave NO2 equivalent to be consistent with EDGAR.

“P8939, L13: “are obtained with ïňĄlter sampling”. The authors should explain what they mean here.”

The sentence has been rephrased.

“P8940, L1-10: when comparing to previous campaigns, also the conditions and samples of these campaigns and how they relate to this particular campaign in Rotterdam need to be discussed. Now it seems as if all ship measurements are directly comparable, whereas I’m pretty sure that (meteorological, (photo)chemical, measurement geometry, ïňĆeet sample) conditions were pretty different.”

The following paragraphs have been added on Pg 8939-8940:

»Apart from the test rig measurements of Petzold et al., all other literature values in

Fig. 13 have been obtained by air borne measurements of emissions from ships sailing on the open sea. Petzold, 2008; Murphy, 2009 measure emissions from a container ship, and also Msc Giovanna, observed by Sinha et al., is a container ship; all of these vessels are using marine fuel oil. The data from Hobbs (2000) are an average of three container ships and three bulk carriers using marine fuel oil and from a navy ship using a distilled fuel. Also the tanker 'Royal Sphere', observed by Sinha (2003) uses a distilled fuel. The ships that create the plume observed by Chen (2005) were only partially identified. The information available about these ships and their operational conditions do not offer any obvious explanation of the differences that are observed in Fig. 13 concerning the relation between particle and SO2 emission rates. Petzold (2008) finds that coagulation has an important influence on the particle number concentration in the initial phase of the plume; he observes a decrease around 50% of the apparent particle emission factor in 10 minutes. It seems that the influence of coagulation could well explain the fact that some of the points reported in the literature lie well below those found in the present study where the plume has a relatively short residence time before encountering the measurement point. Another parameter that may have an influence on the observed number concentrations is the lower limit of the particle diameter that can be detected by the measurement devices. We do not know the size distribution of the particles measured in the present study. Based on the several observed particle size distributions of ship plumes in ambient air (i.e. having been subject to hygroscopic growth) published by Hobbs (2000), Petzold (2008) and Murphy (2009), it can be concluded that the part below 10 nm is typically small but not always negligible. In fact, Murphy (2009) reports evidence of a significant contribution of particles in the range between 3 and 10 nm diameter. One may speculate that the very high ratio of particle number concentrations to SO2 emission factor in the plume of 'Royal Sphere' may be due to an important contribution of ultrafine particles. The close to linear relationship found in Fig 13 for ships using fuels spanning a wide range of sulphur contents is potential useful for predicting particle emissions from ships. However it will need to be confirmed by other studies of particle factor emissions of ships under different opera-

tional conditions for the ship but with a similar (short) residence time of the plume in air.«

"P8941, L5: fourth instead of 'forth'."

Done.

"P8941, L24-27: the authors should also indicate whether the study by Lack et al. (2009) was also concerned with a SECA area, and if their results (mostly in open US waters) can be compared to the regulated Rotterdam harbour measurements."

We added points in the low sulphur regime in Fig. 14. The nice agreement with Lack et al.'s results indicates slight dependence on the area of the study (open sea vs. harbour area). Same was concluded in the text:

» This agreement is found in spite of the fact that that the observations by Lack et al. were made in a non-SECA area and on the open sea.«

"P8942, L21-23: the NOx EF is quantitatively similar to the EDGAR numbers, but what about the fact that Rotterdam is a SECA area, and the EDGAR EFs hold for all possible (including non-SECA and open sea) conditions?"

NOX has nothing to do with SECA.

"P8943, L11-13: what is the explanation for the difference in NOx EFs for ships built before and after 2000? Has combustion temperature been lowered?"

We are not experts in diesel engine technology so in the paper we would prefer not to go into details with these questions in the paper. The issue is discussed in the above mentioned IMO report. NOX emissions may be reduced either by improving energy efficiency (because the limit is in g kWh-1, obviously in ways that do not lead to an increase in NOX generation) or by applying abatement techniques such as injection of water, recirculation of exhaust gases or use of catalytic converters. We don't know if any of the applied techniques cause a decrease in the temperature of the combustion

chamber.

Interactive comment on Atmos. Meas. Tech. Discuss., 5, 8925, 2012.

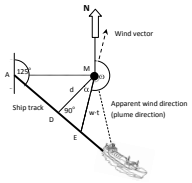

Fig. 1. Figure 4

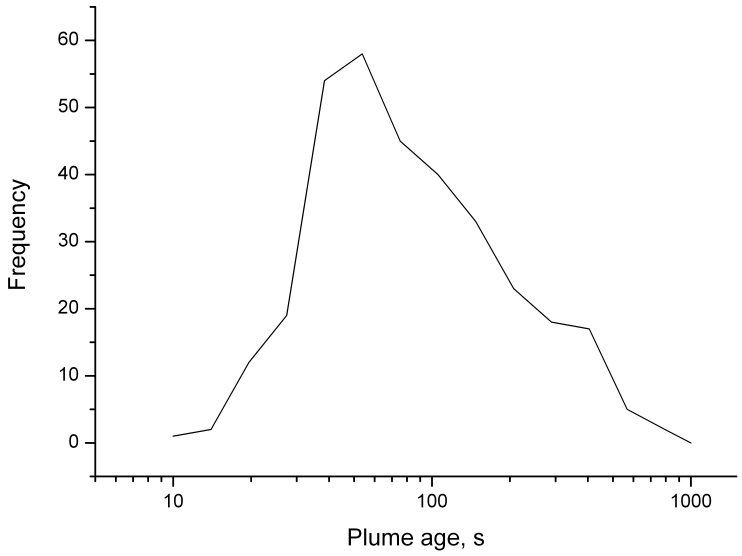

Fig. 2. Figure 5

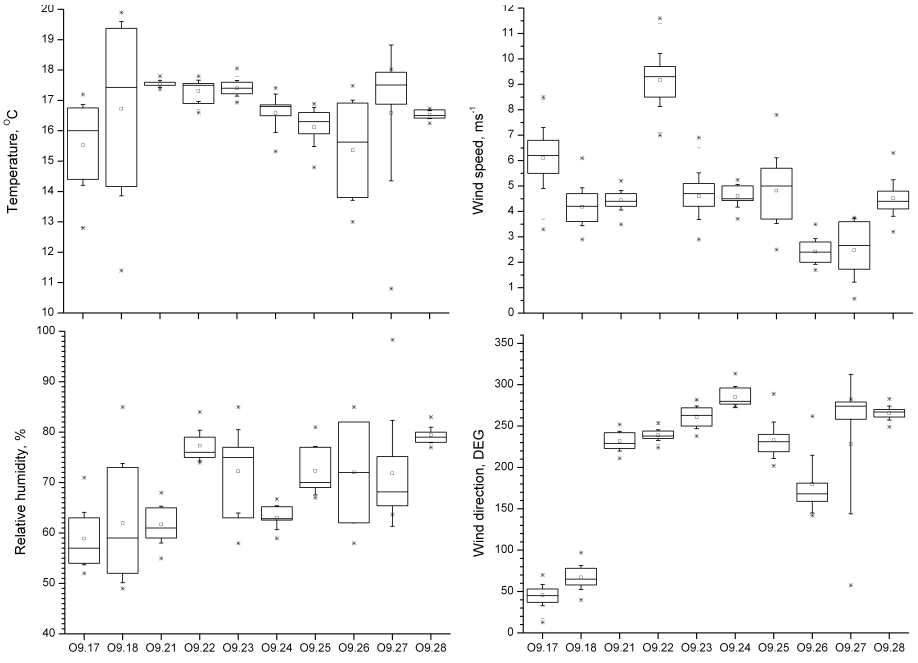

Fig. 3. Figure 6