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

Anonymous Referee #3

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The paper by Alfoldy and co-autors reports on the determination of emission factors for pollutants generated by ship engines in the Rotterdam harbour. The measurement of pollutant concentrations in ship plumes in Rotterdam, a SECA area, and a densely populated region, is very welcome. Some of the results are potentially really interesting, for instance Fig. 10-11 could be very useful to the ship emission community as these allow to quickly derive particle emissions from SO2 emission factors. However, such relationships stand or fall with their general validity, and I have some serious concerns about that, as indicated below. The paper claims that sulphur content of the fuel can be derived from simultaneous CO2 and SO2 measurements, and could be a tool for port authorities to check the sulphur limit compliance of the ships.

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In spite of these interesting and relevant aspects, I have serious concerns with the interpretation of the measurements taken at three locations in the Rotterdam harbour. First of all, very few details are given about the campaign. I was not able to find the start date or end date in the text, and an overview of meteorological conditions is entirely lacking.

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 repeatedly sampled ship exhaust plumes -and confirmed by model studies- the (instantaneous) lifetime of SO2 and NOx within an exhaust plume is very long in the first 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 influence 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.

Section 2.2 reads rather like a company's specification of the instrument, than as a scientific 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 fluorescent 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 fluores-

cence and why is that specific 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? 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 specifications, but as relevant for Rotterdam conditions.

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 firmly prove that their results have universal validity, the title should reflect the specifics.

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

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

P8929, L1: "statistically representative studies". The authors should define what in their eyes is a 'statistically representative' or 'significant' fleet.

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

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

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?

P8939, L13: "are obtained with filter sampling". The authors should explain what they mean here.

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, fleet sample) conditions were pretty different.

P8941, L5: fourth instead of 'forth'.

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.

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?

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?

REFERENCE

Chen, G., et al., An investigation of the chemistry of ship emissions plumes during ITCT 2002, J. Geophys. Res., 110, D10S90, doi:10.1029/2004JD005236, 2005.