Interactive comment on "Investigation of gaseous and particulate emissions from various marine vessel types measured on the banks of the Elbe in Northern Germany" by J.-M. Diesch et al.

Anonymous Referee #1

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As there are many factors that influence ship emissions and still few measurements of these under real conditions available the paper is a valuable contribution to investigating the influence of ship emissions on air quality. However, the presented results do not reflect all relevant influences properly. Thus, the improvements and the remaining lacks should be worked out more clearly. Without correction, proof or removal of the findings that are in my opinion unjustified (see below) I cannot recommend to publish the paper. Despite of this the paper is written clearly, well structured and readable.

Major:

In section 2.5. the authors describe that they determined different emission factors discerning ships by type and size (which is proportional to the engine power as they say). They do not take into account engine and propulsion type which has also an important influence on emissions. Considering this, there might be some restrictions in the applicability of the EFs which is confirmed by the (partly) large error ranges. This should be discussed in the paper.

Author: Using the available AIS information for the probed ships we found a direct proportionality of ship gross tonnage and engine power (R=0.97). This is also in agreement with findings of Hulskotte and van der Gon, 2010.
In order to identify relationships between our measured emission factors and ship characteristics we investigated correlations of all emission factors determined for the various ships with all available quantities that are available from the AIS data. Unfortunately the AIS data do not include information on engine and propulsion type for the vast majority of the vessels. Only for a small – and thus not representative – fraction of the ships the information "diesel engine" is provided. While certainly it would be interesting to include this type of information into the analysis it is unfortunately not possible due to the lack of available information. Within the available ship characteristics the clearest (even though not very clear) relationship with the emission factors was found for the ship size. Therefore we used this variable to investigate emission factor variations for the ships that were probed and to separate the ships into different categories. This issue is now discussed in the paper:
"The gross tonnage is a measure for the ships volume and depends on the ships length by a power function as also mentioned by Hulskotte and van der Gon, 2010. The ships gross tonnage was found to be directly proportional to the engine power (R=0.97)." And: "Within the available ship-related variables from the AIS data ship size (i.e. gross tonnage) and the related engine power were the variables that showed the clearest relationship with our measured emission factors. Indeed, ship size/gross tonnage and engine power are not the only crucial variables that determine the EFs. Engine type, the propulsion system and engine load are expected to play an important role as well. However, since these variables were not (or only for a small fraction) available from the AIS data no relationship between the measured EFs and these variables could be investigated. Likely, the mixture of combinations in engine and

propulsion type and engine load leads to additional scatter in the data, indicated in the figures by the partly large error bars."

292, section 3.2, 3.3, Table 2: It is in my opinion questionable to speak of a correlation if the coefficient is low and not having tested it. I would assume that a correlation coefficient below 0.3 means no significant correlation. Please do not speak of low correlations in such cases without a statistical proof. There's obviously no correlation between EFs and the engine power. I'm quite convinced that there would be a correlation with the load, i.e. the percentage of the power actually used. The load could be estimated by comparing the design speed of the vessel to its actual speed. However, in the optimum case also the engine type and type of propulsion should be considered. Some modern ships with diesel electrical engines, e.g., try to keep the load as constant as possible.

Author: As mentioned above it is unfortunately not possible to investigate potential correlations between the measured emission factors and engine load or engine and propulsion type due to the lack of this information. The clearest relationship between emission factors and ship characteristics was found for the ship size, i.e. the engine power. We are certainly aware that correlations with a correlation coefficient below 0.3 are likely not significant. Our intention to show correlations with a small correlation coefficient was to present potential relationships between emission factors and ship characteristics. However, as suggested, we removed cases with a correlation coefficient below 0.3 in the text together with the corresponding discussions. A number of correlations with rather small correlation coefficients (between 0.3 and 0.5) for correlations between emission factors that can be expected to be related (e.g. BC/CPC, CPC/SO2, CPC/S, CPC/PM1) have not been removed from the text because we think they convey valuable information. Nevertheless, for such correlations the discussion of the relationship was modified to clearly show that there are only potential relationships between the investigated variables. In the following Figure such a correlation for particle number EF with SO2 EF is shown. It has a correlation coefficient of R=0.41 and the linear fit has a coefficient of determination of R^2=0.17. Even though these correlation coefficients are rather low, a general increase of PN EF with increasing SO2 EF can be seen. Therefore we did not remove such correlations from the paper.
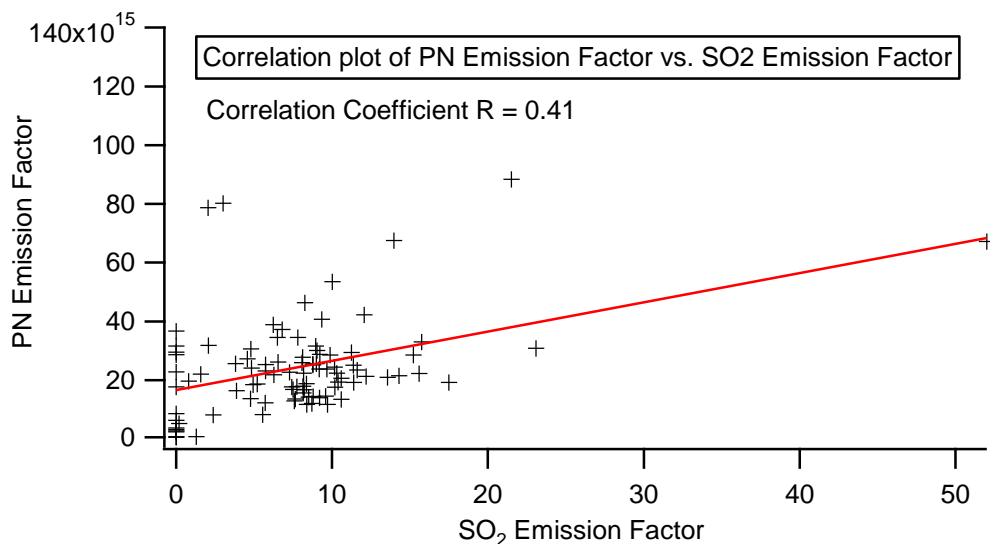
Since we (and the reviewer) want to focus on the strength of correlations instead of the strength of linear fits to the data we now present the correlation coefficients R instead of the coefficients of determination R^2 in the text.

306: "A lower vessel speed implies a lower engine load and less complete combustion process for which reason BC is formed." Generally yes, but this depends on the engine/power/design speed. How do the authors come to their conclusion.

Author: Ship information from AIS did not provide engine load. For this reason, we removed the discussions dealing with the engine load. However, we mention in the manuscript, that we assume BC EFs depend mainly on the operating conditions of the engine like speed, as for high BC emitter the lowest vessel speeds were observed (see Table 2).

456: The point cloud in Fig. 5 doesn't justify this statement in my opinion. The authors should use an appropriate statistical test to underline their statement.

Author: The reviewer's point here is unclear to us. In the lines before line 456 the fact that average particle number emission factors are smaller for the larger ships is mentioned. In line 456 it is stated that a possible explanation for this finding is the larger exhaust system of larger ships in which the aerosol has a longer residence time, supporting coagulation of exhaust particles, finally leading to smaller particle number concentrations. This is not related to Figure 5. In addition the statement is clearly formulated as a possible explanation for the finding of particle number EF dependence. We therefore do not see what should be changed here.

468: "The higher the gross tonnage levels the higher the NOx EFs [...]" This is for me another example for an unproved assertion.

Author: The reviewer is correct that this is an unproven statement. Since the correlation coefficient for this potential relationship is below 0.3 we removed this statement from the text. However, a statement was included that the larger average EFs found for the larger ship types could suggest such a relationship: "In contrast, NO_x (NO, NO_2) is generated by high-temperature oxidation processes with nitrogen in air. For this reason, NO_x EFs are combustion temperature-dependent. The higher NO_x EFs and the NO/NO_2 ratios of the larger vessels are potentially due to higher combustion temperatures within their engines."

483ff: what means "strongly"? There appears to be a difference between Type 1 and the other types, but no difference between Type 2 and 3. This at least is what I get from fig. 7 and table 1.

Author: We reworded this sentence to stress the strength of the differences less: "Comparing the size distributions and EFs for particle components of the different vessel types we found the size of the ship engine exhaust aerosol to show some dependency on the size of vessel that was probed." Indeed, the major difference was identified between "Type 1" and "Type 2 and 3 " vessels. However, although the shapes of the size distributions of the "Type 2 and 3" vessels are similar (figure 7), the number concentrations clearly differ. Additionally, the PM1 mass concentration and the thereby associated variables (sulfate, organics, PAHs) clearly differ dependent on the vessel size.

490: How do the authors derive a third mode above 100 nm for "high BC emitters" from fig. 7? I can only see that the curve flattens out, but this doesn't justify to assume an additional (number) mode.

Author: Compared to the other size distributions, those of the high BC emitters (brown trace on Figure 7) show clearly enhanced concentrations for particles larger than approximately 100 nm. The shape of the trace (the slope changes relatively abruptly around 120 nm) suggests an additional mode here. However,

since not a clear mode can be seen in this size distribution we reformulated the sentence to: "In contrast, "high BC emitters" exhibit clearly enhanced concentrations for particles larger than 100 nm compared to the other vessel types." We also reworded all descriptions of this increase in particle concentrations for larger particles accordingly.

In line 442 the authors write: "Higher black carbon concentrations imply a larger surface area where potential new particle formation precursors condense onto instead of nucleating. Additionally, high black carbon emissions inhibit the growth of freshly formed particles as they are scavenged while coagulating." and in line 486: "[...] and "high BC emitters" emit particulate matter composed of freshly formed particles likely from sulfuric acid nucleating in the expanding plume [...]". Isn't this a contradiction?

Author: Generally we find that increased black carbon concentrations are found together with reduced particle number concentrations, likely because large particle concentrations, associated with large particle surface scavenges vapor that otherwise could form new particles by nucleation. However, this is of course not a situation that is either one way or the other way. Therefore a nucleation mode even for vessels that show enhanced black carbon concentrations is not a contradiction to the previous statement. To make this clearer we changed the first statement to: "Higher black carbon concentrations imply a larger surface area where potential new particle formation precursors preferentially condense onto instead of nucleating."

526: "[...] also the engine type and operation additionally play important roles." This is certainly true, not only for particulates but also for NOx as test bed measurements confirm. There are, however no convincing verifications of this statement in the paper. Maybe the distribution of the vessels into 3 type classes or the criteria by which they are discerned are not suitable.

Author: We agree on the first statement; however AIS did not provide information on engine load for the vessels during operation. Also engine type is only given for about 30 % of the vessels. For this reason, a representative separation of the data according to these features is not possible and the best possible separation of the vessels is according to vessel size.

559: "[...] the gross tonnage is an important parameter [...]" Considering the GT alone is not enough.

Author: Certainly, the gross tonnage is not the only quantity which determines gaseous and particulate emissions. For this reason, we revised the text as follows: "For this reason, in addition to several other quantities the gross tonnage is an important variable distinguishing different kinds of vessels in terms of emission characteristics…".

Section 5: My general impression is that the emission factors proposed by the authors reflect very general emission factors for average ships, useful e.g. for estimating global emissions or bulk emissions for ECAs. They are not suitable for detailed case studies, estimating emission changes when changing the fleet composition or the like. Provided the authors agree to my impression, this should be worked out in the conclusions. Further, the discussion and conclusions might be rethought after removing findings that cannot be verified from the data.

Author: We agree with the reviewer's impression that the measured emission factors are not suitable for detailed case studies as mentioned. However, even without detailed information on load or engine type for the individual ships typical general emission factors for the different vessel classes (based on ship size), measured under typical operation conditions in this ECA have been determined. To make this clear we added in section 5: "As there are many factors that influence ship emissions and still only few measurements of these under real world conditions, the work presented here is valuable for the investigation of the influence of ship emissions on air quality in an emission control area. Additionally, the work presented here is clearly useful for comparing emission factors with those from non-ECA studies as data on ship emissions measured in ECAs are still sparse. Although emission factors were evaluated for a range of vessels and measured quantities, due to the limited AIS information on e.g. engine type, engine load or propulsion system, the presented emission factors generally reflect emission factors for average ships of certain size ranges, useful e.g. for estimating global or bulk emissions for ECAs. These results are not suitable for detailed case studies, estimating e.g. emission changes when changing the fleet composition." As mentioned, we also revised the discussion and conclusion section dependent on data which are statistically relevant.

Minor:

The authors use frequently the word parameter for measured quantities. I'm aware that this has become common practice. It is, however, at least ambiguous. A parameter is by definition a fixed value that cannot be measured but calculated or estimated. The mean of a sample population is for example a parameter that is derived from a number of variable measurements. I'd suggest to use (measured) variable, quantity, concentrations, ...

Author: Thank you for this advice. We changed "parameter" in the manuscript to "variable", "quantity" or "concentration".

Line 139: A table to summarize the the instrumentation and setup would be useful.

Author: According to the request of the reviewer we added a table that summarizes the instrumentation used for the current study (now Table 1). Initially we did not include this table since it is included in the publication where the mobile laboratory is presented (Drewnick, et al., 2012) and we wanted to avoid lengthening the manuscript further.

146: remove "respective". "relative small" relative to what? I wouldn't put the important information of this sentence into brackets.

Author: As we added Table 1 with the corresponding particle losses for each measured quantity, we revised the sentence as follows (not including the word "respective"): "In the size range where the majority of data were measured particle losses are small (see Table 1)." For this reason, the occurring particle losses were neglected as the measurement results were not significantly influenced and the ambient aerosol was measured widely unbiased.

175: Can it be excluded that part of the organics are also detected by the MAAP?

Author: The MAAP measures light absorption and light scattering at the collected aerosol material simultaneously. The purpose of this combined measurement is to avoid the detection of non-absorbing aerosol components as soot (i.e. non-soot material erroneously identified as soot). Even though we

cannot absolutely exclude the possibility that organic (or inorganic) material is mistakenly detected as soot, we assume that the MAAP really measures soot only.

223: [: : :] and PC is assumed to be relatively small. On what is this assumption based?

Author: Complete combustion would lead to carbon dioxide and water. Due to incomplete combustion further gas phase species and particulate carbon compounds are emitted. By comparing the mass concentrations of all measured carbon-containing particulate species (i.e. organics and soot) with those of the gaseous species (mainly CO_2), we found that gaseous constituents exceed by a factor of 1000 those of particulate components. Thus the assumption that CO_2 is a good estimate of total emitted carbon seems reasonable.

246: achieved instead of reached.

Author: Changed.

249: Did the authors check this or can they give a reference?

Author: Yes, both statements in this line have been checked: "The gross tonnage is a measure for the ships volume and depends on the ships length by a power function" was checked in our data set and also found, as mentioned, by Hulskotte and van der Gon, 2010. Additionally, "The ships gross tonnage is typically directly proportional to the engine power" was also found in the measured data set as stated in the text.

264 Does this have a notable influence on the EF?

Author: Dependent on the instrument, the ship emission plumes were registered with different but constant delays and broadenings. These effects result from the transport times through the sampling line that slightly differ for the individual instruments and the internal measurement time constants of the instruments. Both the time delay and the broadening did not change the mean values for the individual vessels for all quantities. For this reason, they did not influence the EFs.

450: How did the authors find out the engine load?

Author: Ship information from AIS did not provide engine load. However, we assumed that a lower vessel speed implies a lower engine load and less complete combustion process for which reason BC is formed. Nevertheless, as this cannot be proven, we delete this context in the manuscript.

Figure 7: What is the point in showing the curve for all vessels? The many curves and (useful) error bars decrease the readability of the figure.

Author: In Figure 7 we want to present the typical characteristics of the size distributions found for the different vessel types which are also discussed in the text. Even though individual size distributions do not differ strongly we do not want to remove arbitrarily some of the curves. In order to improve readability of the figure we removed most of the error bars and show exemplarily only those for the "all plumes" category.

515: marine fuel oil: Is this heavy (or residual) fuel oil? Marine diesel oil has typically a lower S-content

Author: In their publication Corbett and Köhler, 2003 write: "We estimate that world fleet fuel consumption, calculated for all main and auxiliary marine engines in the internationally registered oceangoing fleet (including military vessels), is ~289 million metric tonnes annually. Heavy fuel oil (HFO) represents nearly 80% of the fuel consumed by these engines." For this reason, we changed "marine fuel oil" in "heavy fuel oil".