Author response to referee 1

We thank the referee for the very much appreciated and valuable comments and address them accordingly in the sections below. Additionally, we added some further corrections which are referred to in the last section of this response.

Reply to general comments

Comment by referee:
The model appears to work well for international ship traffic for what concerns SO2 and NOx emissions but it strongly overestimates the SO2 emissions from vessels used in domestic shipping. It is, of course, of interest to understand why several ships that technically could use fuels with 1% sulphur content are using fuels with a lower sulphur content. It is explained in the paper that "this reflects the restrictions of fuel used in inland waterway traffic". Here a few lines about the rules concerning SO2 emissions from ships sailing on inland waterways in Russia (if there are any) would be very helpful for understanding if the use of fuels with low sulphur content is related to legal restrictions or to other reasons. It is anyway an interesting observation that compliance with the limit for fuel sulphur content seem to be good.
Reply: To our knowledge, ships that are going on the river within city borders must use diesel oil instead of heavy fuel oil which is due to safety reasons, but not to sulfur emissions. This is information is now added: "*However, there are also many inland vessels for which there is a large positive bias in the model, indicating that the assigned model FSC of 1 % is much too high since the domestically running cargo and tanker ships actually had a measured FSC of around 0.4 % and less. This reflects the restrictions of fuel used in inland waterway traffic, which to our knowledge prohibit the use of heavy fuel oil within the city borders due to ship-operation safety reasons.*"

Comment by referee:
The model simulations overestimate particle mass emissions by a factor 2-3 and this is attributed by the authors mainly to overestimation of the fuel sulphur content. However, I do not think that this explanation is sufficient: According to Figure 5, the passenger ships predominantly used fuels with a high sulphur content, i.e. close to what is assumed in the model; however the disagreement between model and measurement for what concerns the particle mass is typically about a factor of two (Fig. 10) also for these ships. More generally speaking, I find that the fact that for many ships information about fuel sulphur content as well as speed and the characteristics of the engine was available, should make it possible to evaluate how well the model works when all the requested model input information is available and reliable.
Reply: We agree with the referee and conducted a further comparison between modeled particle emission factors based the measured FSCs and the measured particle measured ones. The result has been added to an updated Figure 10 and is discussed in more detail in section 4.4 and the according passage in the text on **page 25949, line 3 – 17**, which has been changed to: "*The modelling of particulate matter emissions is complex due to the uncertainties in assumptions about used FSC, engine load and the mass and composition of the emitted particles. Large differences between modeled and measured emissions can be seen for particles, Figure . The deviations found might be partly due to the assumed unity density (see for example Virtanen et al. (2002), Petzold et al. (2008), Murphy et al. (2009), Barone et al. (2011), Kuwata et al. (2012) and Pennington et al. (2013) for the typical range of densities), the limited size range for the measured data and assumptions made in STEAM. With respect to the latter, there are several uncertainty sources regarding the modelling of the particulate matter emissions.*
First, the instantaneous main and auxiliary engine power level predictions will have an impact on modeled engine loads. Load levels of engines, in turn, will have an impact on calculated emission factors

and instantaneous values of specific fuel consumption thus changing the mass based emission factors in the model. For the model, engine load and load balancing is more straight-forward for a single 2-stroke main engine than for a setup of several 4-stroke main engines, in the case of which the number of active main engines may vary depending on variable power needs of the vessel. A comparison of the model and measured results for EF(PM) of ships using 2-stroke engines did not show a significant improvement in this study. Unfortunately, the lack of observations from the ships' engine rooms regarding the operational state of the engines hinders a more detailed analysis. However, it is unlikely that these uncertainties are the single cause for the deviations between measured and modelled results of more than 100 %.

Second, the FSC will have a major impact on the modeled PM emission factor. In STEAM, the cheapest possible fuel (with higher sulfur content) is assigned to vessels, defined by geographical limitations (SECA/non-SECA, local legislation for port areas) and technical feasibility of using residual fuel. A re-calculation with the model using FSC from the measurement of the individual ships as shown in the bottom plot in Figure did not lead to a major improvement.

Third, some differences between the modeled and measured results can be explained by unknown parameters for certain domestic ships, for which standard parameters for small vessels were used. Small vessels in domestic operations do not need to undergo the IMO registry procedure and the level of technical details of these vessels in STEAM database is low. For this reason, small vessels are assigned the generic tugboat type, which is bound to lead to inaccuracies in vessel performance and emissions calculations.

Fourth, the conditions of experimental measurements, on which STEAM emission factors for PM are based on (see Jalkanen et al, 2012 for details), do not necessarily correspond to the measurement conditions used in the plume chasing approach used in the current work. The emission factors of fresh exhaust are quite different from those of the aged plume and sample analysis in laboratory conditions may have an impact on PM mass determination when compared to in-situ measurements. The aerosol chemistry after the emission of the plume is not trivial and considering that the plumes are already aged by several minutes there are significant changes in number and mass of the emitted particles Direct comparison of PM results from plume chasing studies with emission modeling would necessitate the use of PM emission factors which represent fresh exhaust and consecutive modeling should be done using a plume model with a detailed description of aerosol processes. This was not done during the current work, however, and it requires further study. It may be necessary to develop separate ship emission modeling schemes for short range studies (both space and in time) of aerosols and for regional scale modeling, unless regional transport models can include relevant aerosol physics modules describing gas/particle partitioning in a short time scale (Robinson et al., 2007;Tian et al., 2014).

It was clearly observed in the modeling results that the misallocation of FSC for ships, especially for domestic traffic using low sulfur fuel, will easily lead to SO_x and PM emission factors of which the latter are over twice higher than what was measured in this work. However, the model does not allow higher fuel sulfur content to be used for vessels than what is allowed by current legislation unless the user assigns the fuel sulfur content manually. Currently, there is no centralized registry for the properties of fuel used in each vessel which makes emission modeling challenging for SO_x and PM. In this regard, the work reported improves the knowledge of the fuel sulfur content of the Baltic Sea shipping."

This has been also taken into now in the summary and conclusions section on **page 25951 in lines 10 to 12** as: "*Overall, the NO_x emissions compared well with the modeled results while there is a significant difference concerning the particle emissions which is only partially due to uncertainties in fuel sulfur content assumptions made in STEAM and requires further evaluation.*"

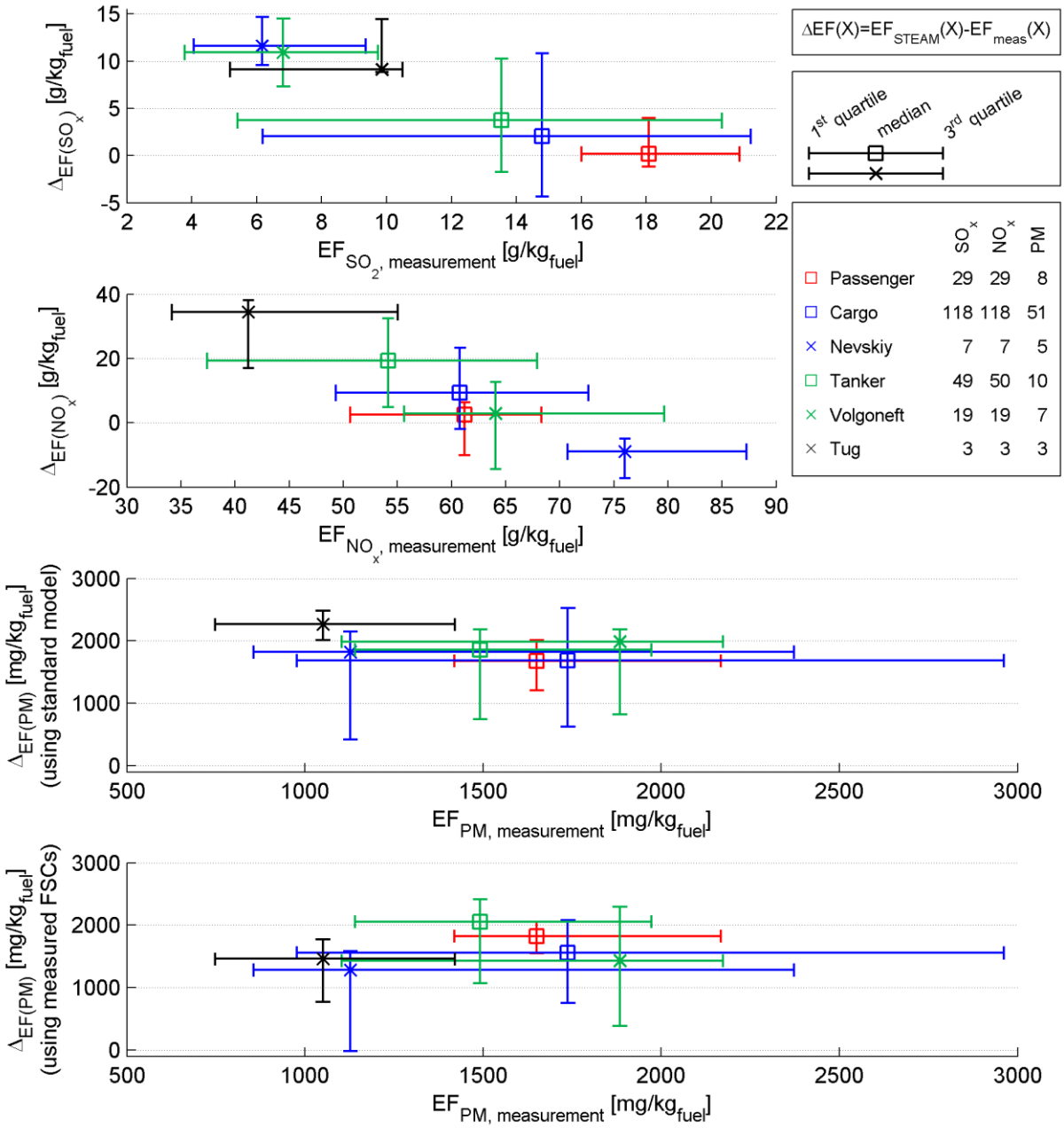

Figure 10: Comparison between modeled data by STEAM and measured data. The differences of the emission factors are shown group wise for individual ships at same speed. Thus it is shown in the graphs by how much the STEAM model statistically exceeds the measured data. It should be considered that the modeled SO_x was compared to the measured SO_2. Furthermore, the sum of the modeled emissions of OC, EC, ash, SO_4 are compared to the measured size distributed data between 5.6 nm and 10 µm under the

assumption of a particle density of 1 g/cm^3, which were calculated using the FSCs originally assumed by the model and the FSCs obtained from the measurements. The number of the compared plumes for each type and species is presented by the number values in the legend.

Reply to specific comments

P. 25934, line 23: "...the measurements in 2011 were carried out at the transition period..": Please mention the date when the 1% sulphur limit came into force in Russia.
Reply: This information has been added. The sentence now reads: "*The measurements in 2011 were carried out at the transition period when Russia ratified the Annex VI protocol in April 2011 requiring maximum 1 % sulfur content in the fuel which came into effect on July 8, 2011 (AMSA, 2014).*"

P. 25934, line 27: Please state clearly if, to your knowledge, there have been other studies of ship emissions in the eastern Baltic.
Reply: This information has been added to the sentence: "*Since very few real world emission measurements of ships have been conducted worldwide, especially in the eastern Baltic where to our knowledge no such studies have been carried out before, there is a need for this type of data to be able to model ship emissions more accurately and subsequently carry out air quality modeling.*"

P. 25935, line 24: "…but in most cases ships emit very little VOC…": Please give reference.
Reply: The reference (Williams et al., 2009) has been added now.

P. 25936, line 15-16: Is it possible that the sampling frequency can be 10 Hz (i.e. 0.1 seconds for each sample) when the response time (t_{90}) is 0.5 seconds?
Reply: The sampling rate is the frequency by which the data is recorded. It only regards the frequency of signal detection, while the response time denotes the time it takes for the instrument to reach a certain percentage of the final value at a step change at the input, which is 90 % in the case of the applied response time t_{90}. For example, if the flow of sample air through an instrument is rather slow, the response time would not necessarily decrease with an increase of the sampling rate. So it is reasonable that the sampling period, i.e. the inverse to the sampling rate, is shorter than the response time.

P. 25937, "2.2 Calculation of emission factors": Although it is clear how the calculations were done, the description is formally incorrect and need to be changed. The authors want to explain, as far as I understand, that the concentration measurements are integrated over a certain time period, but in this case they should not use the summation symbol _ which has a different meaning.
Reply: The equations have been changed to:

$$\mathrm{EF(X)_{g/kg_{fuel}}} = \frac{\mathrm{M(X)}_{g/mol} \cdot \int [\mathrm{X}]_{ppb} - [\mathrm{X_{bgd}}]_{ppb}\, dt}{\mathrm{M(C)}_{g/mol} / _{0.87} \cdot \int [\mathrm{CO_2}]_{ppm} - [\mathrm{CO_{2,bgd}}]_{ppm}\, dt} \quad (1)$$

$$\mathrm{EF(PN)_{\#/kg_{fuel}}} = \frac{\int [\mathrm{PN}]_{\#/\mathrm{cm^3}} - [\mathrm{PN}_{bgd}]_{\#/\mathrm{cm^3}}\, dt}{\int [\mathrm{CO_2}]_{\mathrm{kg/cm^3}} - [\mathrm{CO_{2,bgd}}]_{\mathrm{kg/cm^3}}\, dt} \cdot \mathrm{EF(CO_2)_{kg/kg_{fuel}}} \quad (2)$$

$$[\mathrm{CO_2}]_{\mathrm{kg/cm^3}} = [\mathrm{CO_2}]_{ppm} \cdot \mathrm{M(CO_2)}_{g/mol} \cdot \frac{p_{Pa}}{R_{J/(mol \cdot K)} \cdot T_K} \cdot 10^{-15} \quad (3)$$

P. 25939, line 18: "…usually for several calibration steps 300 ppb for both gases": The meaning of the sentence is not clear.

Reply: This sentence has been rewritten:
"*In 2012 a high flow dynamic gas calibrator (Thermo 146i) in conjunction with a zero-air supply (Thermo 1160) was used to dilute SO_2 at 63.7 ppm±3 % and NO at 64 ppm±5 % in several calibration steps to volume mixing ratios between 0 and 300 ppb for both gases.*"

P. 25939, "2.3 Calibrations" and p. 25940 "2.4 Uncertainties": Generally the uncertainties here are given as a percentage (e.g. on p. 2540, line 9.). However, I would presume that the relative uncertainties on the low concentrations (and consequently normally also on the low emission factors) generally are higher than the uncertainties on the high concentrations and thus on the high emission factors. I would suggest to the authors to address this issue, which is particularly relevant in relation to compliance monitoring in a SECA area, because reliable measurements also of relatively low SO2 emission factors will be needed to check compliance with the limit of 0.1% maximum sulphur content in fuels, which will be in force from 2015.
Reply: The following passage has been added now to page 25941, line 8: "*It should be noted that the uncertainty for emission factor of SO_2 of 20 % was found for FSCs of around 1 %. In a study by Mellqvist et al. (2015) the emission of a ship which was known to run voluntarily on marine gas oil and hence with FSC of 0.1 % or below was repeatedly measured, yet at different occasions, using the same system at a fixed site. The measurements indicate an average FSC of 0.06 % with a standard deviation of about 0.03 %. Under the assumption that the FSC used by the ship would be the same at all measurements, this would indicate an uncertainty of about 50 % for the EF(SO_2) for ships running on low FSC around 0.1 %.*"

P. 25944, "4 Results and discussion": The study provides information about both fuel sulphur content and particle emission factors (number and estimated mass) for the same ships. It might be interesting to use this possibility to look at the dependence of particle emission factors on fuel sulphur content.
Reply: We currently examine this matter is in greater detail as part of a separate study in combination with results from other campaigns to have a bigger picture on this.

Technical corrections according to referee

P. 25933, line 8: "...with about 4% growth.":...per year?
Reply: The growth is related to 2012. Further, the publication date of the reference has been corrected:
"*In 2012 about 9 billion tons of goods were transported by ships, corresponding to almost 80 % of the worldwide merchandise trade by volume, with about 4 % growth as compared to 2011 (UNCTAD, 2013).*"

P. 25935, line 1: "i.e" should probably be "e.g.".
Reply: This has been corrected according to suggestion.

P. 25935, line 15: "..for about 0.3 s.":...each?
Reply: This has been reformulated to: "*…for around 0.3 s per species.*"

P. 25941, line 9: "Following" should probably be "according to".
Reply: This has been corrected accordingly.

P. 25949, line 21: "…the model does not allow higher fuel sulfur content to be used…": probably "higher" should have been "lower" here.
Reply: The model assumes that ships use fuel with FSC according to current legislation. But it takes into account that it is common to use higher quality fuel with lower FSC in the auxiliary engines than for the main engines. It is not assumed that ships use fuel with higher FSCs than permitted.

Further corrections

P. 25933, line 7: The reference (UNCTAD, 2013) was not shown in reference list. This has been corrected.

P. 25935, line 12-14: I added an explanation for the term t_{90}. It now reads:
"*CO_2 was measured with cavity ring-down spectroscopy (CRDS) (O'Keefe and Deacon, 1988) using a modified flight Picarro G2301-m greenhouse gas monitor with a response time, t_{90}, of less than 1 s, which time that is needed at a step change to reach 90 % of the final value.*"

P. 25938, line 4: The value for the assumed pressure was corrected to 101325 Pa (before1013.25 Pa).

P. 25938, line 8: Reference to equation is corrected to "… analogue to Eq. (1) …".

P. 25938, line 17 & line 21: The format and numbering of Eq. (3) has been adapted according to the other equations.

$$\mathrm{GMD}_{nm} = \exp\left(\frac{\sum[\mathrm{n_i}\cdot\ln(\mathrm{D_{pi,nm}})]}{\mathrm{N}}\right) \quad (4)$$

P. 25941, lines 21 to 29: This section has been corrected to: "*The SMPS System which was used in this comparison was validated with standardized polystyrene latex spheres (PLS) of known sizes between 70 and 500 nm. From the deviations it was seen that the particle diameters were underestimated by the SMPS by less than 1 % at an offset of less than 7 nm. A comparison with the CPC indicated an underestimation of the total particle number of about 30 % by the EEPS. A similar discrepancy has been observed in another study (Jonsson et al., 2011), when comparing the same type of instruments.*"

References

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