Reply to RC C2755, Anonymous Referee #2

We thank the referee for carefully reading our manuscript and the valuable comments and suggestions. We have included responses to the reviewer's comments below and indicated where changes were made to the text (blue).

Pirjola et al. report in their paper emission factors (EF) for 11 individual ships equipped with different aftertreatment system, travelling at ports of Helsinki and Turku. The authors applied the plume catchment method performed from the shore by a mobile laboratory equipped by the necessary instruments, moving the van to the best measurement location determined by wind conditions. The number of papers on the related field increases, indicating the growing importance of ship emission on human health, air quality and climate worldwide. However, this work pursues the series of previous studies, the number of relevant citations is limited and the comparative analysis of the data is also incomplete.

We have added some recent citations in Introduction on page 4: Alföldy et al. (2013); Berg et al. (2012); Williams et al. (2009); Chen et al. (2005); Juwono et al. (2013); Moldanova et al. (2013); Petzold et al. (2010); Schlatter et al. (2010). The following sentence was added page 4: "The reported emission factors are in the range of 2.9-44 and 22-109 g (kg fuel)$^{-1}$ for SO_2 and NO_x, respectively, in the range of (0.2-6.2)x10^{16} (kg fuel)$^{-1}$ for particle number, and in the range of 0.5-5.3 g (kg fuel)$^{-1}$ for $PM_{2.5}$."

On the other hand, the paper concerns numerous important topics such as particle size distribution, volatility of the emitted aerosols, effects of aftertreatment systems, comparison between seasons, etc. For the above reasons I suggest to accept the manuscript for publication after mayor revisions based on the following comments.

1. The reported data concern for SECA during an important period, namely after the reduction of fuel sulphur content (FSC) from 1.5% to 1%, and prior to further reduction from 1% to 0.1%. Even though, the presented work focuses mainly on the aerosol emission, the paper would document the effect of the FSC reduction, since particle emission and FSC are tightly related as previous works demonstrated. For this reason, I miss a detailed comparison with previous works done by SECA prior to the recent reduction of FSC. For example, whereas Alfoldy et al. (2013) published particle emission factor up to 5.7 10ˆ16 (kg fuel)ˆ-1, this paper reports particle EF below 2.26 10ˆ16 (kg fuel)ˆ-1, as an evident sign of the benefits of FSC reduction.

This was a good point, and has now been discussed in Section 3.4 (pages 14-15).
"In this work the emission factors are measured in the SECA areas after the reduction of the allowed FSC from 1.5% to 1%. The obtained EF_{Ntot} are in the range of (0.32-2.26)x10^{16} (kg fuel)$^{-1}$. As well measured in the SECA areas in 2010, Moldanova et al. (2013) report a low value of 0.5x10^{16} (kg fuel)$^{-1}$ at 30% engine load, and Jonsson et al. (2011) the values in the range of (1.37-3.6)x10^{16} (kg fuel)$^{-1}$. On the other hand, Alföldy et al. (2013) who performed measurements in 2009 in the SECA area (FSC<1.5%), report higher values of (0.8-5.7)x10^{16} (kg fuel)$^{-1}$. For higher FSC, from 2-5%, number emission factors are typically higher, in the range of (1.2-6.2)x10^{16} (kg fuel)$^{-1}$ (Chen et al., 2005; Sinha et al.,2003; Petzold et al., 2010). Unfortunate, literature provides only a limited number of papers concerning EF_{Ntot} for ships. Fig. 8 and previous works (Fig. 13 by Alföldy et al., 2013) demonstrate that particle number emissions and FSC or EF_{SO2} are positively correlated. In spite of many variables affecting EF_{Ntot} this paper is an evident sign of the benefits of FSC reduction."

2. Authors found that the calculated FSCs are significantly lower than the actual limit

at SECA (0,37% vs. 1%). They explain this difference in the first paragraph on page 7164 considering the contribution of the auxiliary engine emission that generally use low sulfur fuel. Authors should mention that same thing was found by previous studies (see e.g. Alfoldy et al., 2013). They should also enhance here that the reported EFs (for O2 and particles) are biased due to the relatively significant contribution of auxiliary engines in ports comparing to the steady state engine operating conditions that is generally current at open sea.

We added in Table 5 the information obtained from the ship owners concerning the sulphur contents of the fuels used in our measurements, and modified the text in Section 3.5 (page 15): "If both main and auxiliary engines use fuel with identical fuel sulphur content as was the case for most of the ships in this study (Table 5), the measured value can be thought as direct indication of the fuel sulphur content used onboard the vessel. In the cases where main and auxiliary engines use fuels with different sulphur content, the sulphur levels will be a combination of these contributions roughly equivalent to the ratio of main engine and auxiliary engine power used at the time of the measurement. Then the measured emission factors for SO_2 and particles might be underestimated. Alföldy et al. (2013) estimated that the reduction of total SO_2 emission factor caused by auxiliary engines' contribution can be 6% at sea and 30% in ports if FSC is 0.5% for MDO and 1.5% for HFO. As seen from Table 5, the measured FSCs do not exceed the values given by the ship owners, and for all ships they are less than 1% which was set as a limit in Baltic Sea on 1 July 2010."

Table 5. Estimated fuel sulphur contents along with standard deviations for all ships studied in this work. Also given is the information from the ship owners.

Ship	FSC (%)	Ship owners
A	0.68±0.26	n.a.
B	0.84±0.10	<1.0%
C	0.62±0.18	<1.0%
D	0.41±0.16	n.a.
E	0.33±0.10	<0.5%
F	0.34±0.18	<0.5%
G	0.31±0.13	<0.5%
H	0.38±0.06	<0.5%
I	0.38±0.15	n.a.
J	0.37±0.13	n.a.
K	0.46±0.16	<0.5%)

n.a. - not available

Section 2.5, page 9: The sentence "Based on the measured carbon mass percent of the fuels (Cooper et al., 2005), a similar equation was derived in a slightly different way by Alföldy et al. (2013)." was deleted, and the sentences "Should be noted that a minor part of the fuel sulphur might be emitted as SO_3 or converted to H_2SO_4 by homogeneous and heterogeneous partways in

the atmosphere (Alföldy et al., 2013; Williams et al., 2009). Thus eq. (4) considers lower limits for FSC." were added.

3. Since one ship was measured twice or more times, statistical analysis would be beneficial for the evaluation of the repeatability of the measurements. Even though standard deviations of the results are given in the paper, no information can be found in the text how were they calculated. A new section regarding the uncertainty analysis of the measurements should be added to the text.

As pointed out by the referee, the main focus of this paper is to study particle number size distribution, volatility of the emitted aerosols, effects of after-treatment systems, comparison between seasons, etc. Due to a limited number of repetitions (5-12) for each ship type, any robust statistical analysis cannot be presented. The results concerning particle number concentration and FSC are the mean peak values along with standard deviations as was mentioned in the text. However, for emission ratios as well as for emission factors the least square slope determination was used. More discussion concerning the uncertainty sources were added:

Section 2.2, page 6: "The uncertainty of the measurement results of the ELPI is difficult to estimate since no clear information of the performance characteristics of the instrument was able to reach from the manufacturer. However, based on the study by EURAMET it is some tens of percent (Schlatter, 2010)."

Section 2.2, page 7: "The expanded uncertainty of the gas analyzers including the contribution of major components of the performance characteristics of the analyzers and the traceable calibration lays approximately ± 10 % but increases considerably at the low concentration levels (ambient background concentration). The uncertainty calculations and the calibration service of the analyzers were provided by the Calibration Laboratory of the Finnish Meteorological Institute, which is designated as a Standard Laboratory for the Air Quality in Finland by the Centre for Metrology and Accreditation."

Section 2.2, page 7: "All instruments were synchronized with the GPS time, and zero checked before and after the measurements."

Section 3.4., page 14: "Linear regression analysis was used to determine the slope of the line."

Section 3.4, page 14: "The uncertainty covers errors from the instrumental uncertainties (Sect. 2.2), errors in calculating ΔCO_2, since it was very sensitive to the background concentration, errors in the least squares slope determination, and errors in the assumptions in eq. (4)."

Concerning the discussion of uncertainty of EF(NOx) and EF (SO_2), the following text was added:
Section 3.4, page 14: "The conversion of NO_2 to HNO_3 and PAN is much slower (Chen et al., 2005; Williams et al., 2009)."

4. Fig. 6 is still not understandable. Authors briefly described in their reply how they generated the figure from the raw ELPI data, but they should do the same in the text, detailing how and which Matlab functions were applied. A relevant reference for the applied software is also required. Otherwise the original ELPI data should be presented here. It is a quite important point, since they haven't presented what they measured, but a modified figure was inserted instead.

We have used a very common way to present the time evolution of particle number size distributions if a commercial program Matlab is available (see eg. Kulmala et al. 2005; Pirjola et al., 2012). The measurements should be performed by an instrument which can classify particles

into some number of stages such as SMPS, EEPS or ELPI. The Matlab command needed is pcolor(X,Y,C), where X and Y are vectors or matrices, which produces a pseudocolor plot on the grid defined by X and Y (Matlab R2013a). In our case X refers to time (h), Y to particle diameter Dp (m) and C to dN/dlogDp (cm^{-3}), all these come from the raw ELPI data (10-s data). With a command shading('interp'), each cell has color resulting from bilinear interpolation of the color at its four vertices.

Because the commands used are Matlab's own commands, it is not usual to present the script in the paper. We have added a sentence "The figure was produced by a commercial program MatlabR2013a." on page 11, Section 3.2.

However, the Matlab script is below and a part of the data is attached (data_fig6.dat). In the data file, the cells 2-13 in first line refer to diameter Dp (m), 1st column refers to time (h), 2-13 colums to dN/dlogDp (cm^{-3}).

```
load data_fig6.dat
v=data_fig6;
[m n]=size(v);
Zdata=log10(abs(v(2:m,2:n))+1e-6);
pcolor(v(2:m,1),v(1,2:n),Zdata');
shading interp
colormap(jet(250));
caxis([1 6]);
set(gca,'yscale','log')
axis([9.25 10 1e-8 1e-5]);
```

References:

Alfoldy et al. (2013) Measurements of air pollution emission factors for marine transportation in SECA, Atmos. Meas. Tech. 6, 1777-1791.

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