Response to Reviewer Comments

Ship Speed, Fuel Quality and Exhaust Gas Scrubbing

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We thank the reviewers for their time and comments. We have addressed all concerns and have outlined the changes made in the text below.

<u>Referee #1</u>
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The paper does an excellent job of compiling and comparing previously published data on factors that determine BC emission factors for marine vessels. It should be pub- lished in ACP as it provides a very timely review on what is known about the impacts of ship speed, fuel quality, and exhaust scrubbing on emissions of BC. The paper is well written and the data are clearly presented. I have only the few, very minor comments listed below.
p. 3513, line 12: Should this be “catalyze FORMATION of BC” and not COMBUSTION?
The text as written is correct. Heavy metals can increase combustion of BC as it forms within the flame.

p. 3515, lines 11 – 15: Clarify what is meant by “almost all techniques mentioned vary for ship engine exhaust measurement”. Does this mean the different methods show poor agreement or yield different results for ship exhaust?
We have clarified this:
“Most of these techniques have been applied to the measurement of EC or BC from diesel engine emissions (Burtscher, 2005). However, most of the techniques mentioned here show poor agreement when compared when measuring ship engine exhaust despite the fact that they can show excellent agreement with each other under controlled laboratory conditions with controlled samples”

p. 3516, line 24: A brief description of the “ISO-8178 standard” should be provided.
We have added the following when we discuss the ISO standard:
“Note that the ISO 8178 standard specifies the test load cycles for emissions measurements from non-road vehicles. In addition it specifies a minimum dilution of the emitted exhaust prior to sampling.”

p. 3517, line 6: change to “. . .indicates that the mass absorption coefficient used, 7.5 m2/g, is generally valid. . ...”
Done

p. 3522, line 18: Describe/define “speed proportional to load relationship” upon first use.
This has been done in section 3.1

p. 3524, line 22: Change to "..on BC EMISSIONS is more complex. . .."
Changed

Referee # 2:

1. Section 2.2: Petzold et al. (2010) showed in their paper that for the performed test rig studies BCFilter and BCFSN are highly correlated while the difference between BCFilter and BCTOA is anti-correlated to the ratio of organic carbon (OC) to BCTOA (named EC in this study). They suggested a potential cross-sensitivity of BCTOA determination to the organic fraction of the carbonaceous aerosol. Erroneously high BCTOA values may be caused by pyrolytic conversion of OC to BCTOA during the thermal analysis step particularly for samples with a high OC fraction of total carbon. This interpretation is supported by a method intercomparison study (Schmid et al., 2001). On the other hand, close agreement is found between BCFilter and BCTOA for an aerosol with low organic content, see e.g. Petzold et al. (2011). This finding offers a potential reason for the observed discrepancy. Furthermore, the authors refer to this explanation in Section 4.1 (page 3526, line 14ff) without giving details in Section 2.2.

In section 2.2 we have added the following:

In a study on gas turbine engines, Petzold et al. (2011b) showed that a similar discrepancy between BC_{Filter} and EC_{TOA} was anti correlated to the amount of particulate organic matter (POM) present, and suggested that pyrolysis of POM to EC_{TOA} could occur. This interpretation was supported by the broader inter-comparison study of Schmid et al. (2001).

In section 2.6 (measurement methods summary) we have added the following:

however there appears to be some inconsistency between EC_{TOA} and BC_{Filter} in field measurements of ship exhaust, very likely due to the POM content of the exhaust causing biases in the measurement (Petzold et al., 2011b;Schmid et al., 2001). EC_{TOA} and BC_{FSN} show good agreement in one study, suggesting that biases were not significant in that study. This suggests that the EC_{TOA} method is not nearly as reliable a tool for measurement of BC.

2. Section 3.1: Eq. (1) is introduced as calculating fuel consumption per nautical mile of travelling. In its current version this equation reports fuel consumption per hour of operation. Please correct the equations or adjust the text accordingly.
We have adjusted the equation to reflect the fuel per nautical mile unit.

3. Section 3.3: The expression "speed / engine load3" is misleading. I suggest introducing it as "cruising speed proportional to engine load to the power of 3 (speed/ engine load3)". This avoids confusion.
In section 3.1, where we first define the cubic relationship between engine load and vessel speed, we have done what the reviewer suggests.

Page 3517, line 15: translated into BC
Fixed
Page 3520, line 3: some studies show a direct relationship
Fixed
Page 3521, line 23-24: delete "..."
Deleted
Page 3523, line 13: combining safe speed/IDN data and the rated speed data
Fixed
Page 3528, line 9: BC from ship exhaust
Fixed
Page 3532, line 16: each ship's load distribution
Fixed

Referee # 3:

1. Please show the R^2 values in figure 1.

Done

2. Pg 3517 line 15 "into" BC

Added

3. Figure 6. Are these the study numbers in table 3? The legend is too small to read. What is the difference in the dotted and solid lines?

We have increased the size of this legend to make it clearer. The difference in the dotted and solid lines is the study type, which can be found in the table. This is made clearer with a larger legend.

4. Pg 3528 line 9 remove "within"

Deleted.

Reviewer: J. Ristimäki

General comments: In the topic of the paper three aspects for ship emitted black car- bon is presented. In addition to these three aspects (ship speed, fuel quality and ex- haust gas scrubbing) definitions and measurement of black carbon is widely discussed in the paper. Compared to the total length of text in the paper (24 text pages) 5 pages of discussion about methods feels quite a lot. Although maybe relevant for the issue authors should consider referring to other papers focusing purely on the definitions of black carbon and/or measurement methods (e.g. Slowick et al. AST 41,2007 DOI: 10.1080/02786820701197078) to shorten this section.
Within this review paper we strived to minimize the length of the manuscript. We feel that because this is a review paper that we need to include this section at its current length. The small amount of data that is available on this topic is spread across multiple operational definitions of 'black carbon'. There is a substantial body of research that discusses comparisons of the measurement methods for these definitions. To effectively use all of this data we need to spend some time showing that they can be confidently compared in the next sections of

the manuscript.

In the references used to evaluate the effect of fuel quality (table 3) on BC emissions, one reference (Petzold et al 2011) represent over 50% of the total number of points. As Petzold et al used only one single cylinder test engine with different fuels a significant bias to real world case can occur. This possibility to bias should be noted in the text.
We have added the following:

"These studies include EF_{BC} measurements from in-use marine engines, full, and reduced size test bed engines and provide converging evidence that improved fuel quality is linked to reductions in EF_{BC} for marine diesel engines."

Although the Petzold test bed engine is a single cylinder and not representative of a full-scale engine it was operated at appropriate conditions for a marine diesel engine. Because we are reporting ratios, to we do not agree with the reviewer that any bias will exist because any issues due to the smaller engine will be replicated at both high and low fuel sulfur levels.

Authors have previously published data which show opposite trend in BC emissions as a function of fuel sulphur (e.g. Lack D., Corbett J.J. et al 2009 "Particulate emissions from commercial shipping: Chemical, physical and optical properties") and now discard these results in chapter 4.2 by referring to data that is not yet publicly available for evaluation (Buffaloe et al). Author should consider if it is a scientific approach to discard published data based on unpublished data.
We have not discarded our previous data. Because this review is comparing the BC emissions from low and high quality fuel, we need to have a robust baseline. The Lack 2009 data does not have a baseline that can be established with confidence because BC emissions from single ships were not measured at low and high quality fuels. Also, the high variability in vessel loads and operating conditions complicate the emissions profile for that region. Under these conditions, the average (and other statistics) from the main dataset is a more reliable number than considering individual points. Comparing to averages from California allows us to set a high quality fuel "regional baseline" in addition to investigate the mean values of EF_{BC} in a region using high sulfur fuel and higher engine loads compared to a region using low sulfur fuel and operating at lower engine loads. Producing these averages is in fact the best scientific method to utilize the Lack etal 2009 data. In addition, we have been very careful to state that this data is only "suggestive" of a link, not conclusive.

Further it should be noted that nearly 50% (8/19) of the compar- isons in the table 3, are to fuels which are not produced enough to be real alternative to HFO. I suggest that results are re-evaluated while discarding the biogenic fuels from the evaluation as they are not a true alternative to HFO.

We respectfully decline to remove this data. Although the biogenic fuel may not

be a viable alternative fuel on the large scale, these fuels do represent a high quality fuel in comparison to low quality fuel within the same engine. It is therefore an important dataset to provide an indication on what the effect of a shift in fuel quality will do to BC emissions.

Detailed comments: Introduction: As the share of global CO2 emissions of ship- ping are mentioned (3.3%), it would be very informative for the reader to also ob- tain the contribution of shipping to global BC and SO2 emissions.
We have added the estimated global SO2 and BC contributions in the text (see below):

Further, it would be informative to the reader to get an estimate (e.g. in %) for the most signif- icant BC sources of the BC deposited on the arctic – including shipping.
We have added the following text:
Shipping contributes about 2% to global BC emissions (Lack et al., 2008b), and currently very little in the Arctic (Corbett et al., 2010a). The vast majority of current Arctic BC is sourced from outside of the Arctic region (Quinn et al., 2011), and future ship traffic in the Arctic will be one of the few direct emissions of BC into the Arctic (Quinn et al., 2011). In addition, these added direct emissions will likely add to the climate impacts in that region (Corbett et al., 2010a;IMO, 2010a).

Page 3513 line 7 and 11. Comparing the number of references in the two sentences "Each of these impurities is known..." (1 reference) and "There are some evi- dence.." (2 references) the phrasing should be vice versa: "There is some evi- dence that these impurities...(American bureau of shipping)" and "It is known that heavy metals catalyze...(Maricq 2007...)".
We have changed the text so that both read "it is known".

Authors should note that there are much more scientific articles reporting enhanced oxidation of soot due to metal additives (e.g.Jung et al Combustion and Flame 142 (2005) 276–288) and several commer- cial applications of metals catalyzing oxidation of soot exists. E.g. fuel borne cat- alysts in PSA group automobiles and products manufactured by . Innospec, Tech- nol, SFA international, Conseal international. http://www.meca.org/galleries/default-file/MECA%20Diesel%20White%20Paper%2012-07-07%20final.pdf page 21 on fuel C866borne catalysts states "Fuel-borne catalysts are a colloidal dispersion of base metal oxides or organic compounds containing precious or base metal ions such as plat- inum, cerium or iron and are added to the diesel fuel prior to the combustion process." and "The direct contact between catalyst particles and soot particles reduces the tem- perature required for ignition of trapped particulate matter that is collected together on the filter media."
Noted.

Chapter 3.1 line 15. “conditions away from stoichiometric combustion”. References supporting this claim should be provided as typically diesel engines operate with excess air conditions (lambda >1) e.g. away from stoichiometric conditions.
We have changed “stoichiometric” to “ideal”.

Chapter 6.2 line 4 on page 3531: “There is limited evidence that heavy metals catalyze the com- bustion...” Based on comments above on 3513 this sentence should also be revised.

We have adjusted this sentence to read:

“From the data presented, we are unable to conclude if heavy metals catalyze the combustion of BC.”