

Reply to Editor

Thanks very much for your comments. We would like to ask you if we can change the authors and the relevant affiliates because of the change of supporting project.

Our replies are given as following according to your comments:

Both reviewers supported the measurement methods and data quality. However, one reviewer mentioned the need to consider the dilution when evaluating organic carbon, and to present more standard emission measures. The other reviewer requested a better treatment of uncertainties.

Thanks for your comment, and the reply is given as following:

The residence time of soot particle in the pipe could affect the formation of PM, which led to the different composition of PM. The dilution sampling was not used in this study that might be one reason of the lower OC to EC ratio. Besides, TOR was used to measure OC and EC in PM, which always had a lower OC content compared with other methods because of the different definitions of OC and EC. They are shown in the revised manuscript (Line 30, Page 15 and Line 1-8, Page 16) as following.

The non-dilution sampling was the main reason of the lower OC to EC ratio in this study. Besides, TOR was used to measure OC and EC in PM, which always had a lower OC content compared with other methods (such as TOT) because of the different definitions of OC and EC (Khan et al., 2012). Compared with other diesel engines., the ratios of OC to EC in this study were higher than that of automobile diesel soot, in which EC comprises 75–80 wt% of the total PM (Clague et al., 1999), and also higher than heavy heavy-duty diesel trucks (HHDDT) with OC to EC ratios below unit for cruise and transient modes even though higher in cold-start/idle and creep modes (Shah et al., 2004).

The real-world measurement system for vessels including on-board test picture and schematic diagram of the portable measurement system has been added in Figure S1. Detailed instruction of the sampling system also has been added in Supporting information, shown as following:

Real-world Measurement System for vessels

Detailed compositions and procedure of the on-board measurement system were given as follows: The whole measurement system was placed on deck next to the exhaust pipe of the vessel. A slender tube was placed into the vessel exhaust pipe to lead out the flue gas. Then it was divided into five subsamples through a manifold for different analyses and evacuation of the excess gas. The on-board test picture (Figure S1, a)) and schematic diagram of the portable measurement system (Figure S1, b)) are shown in Figure S1.

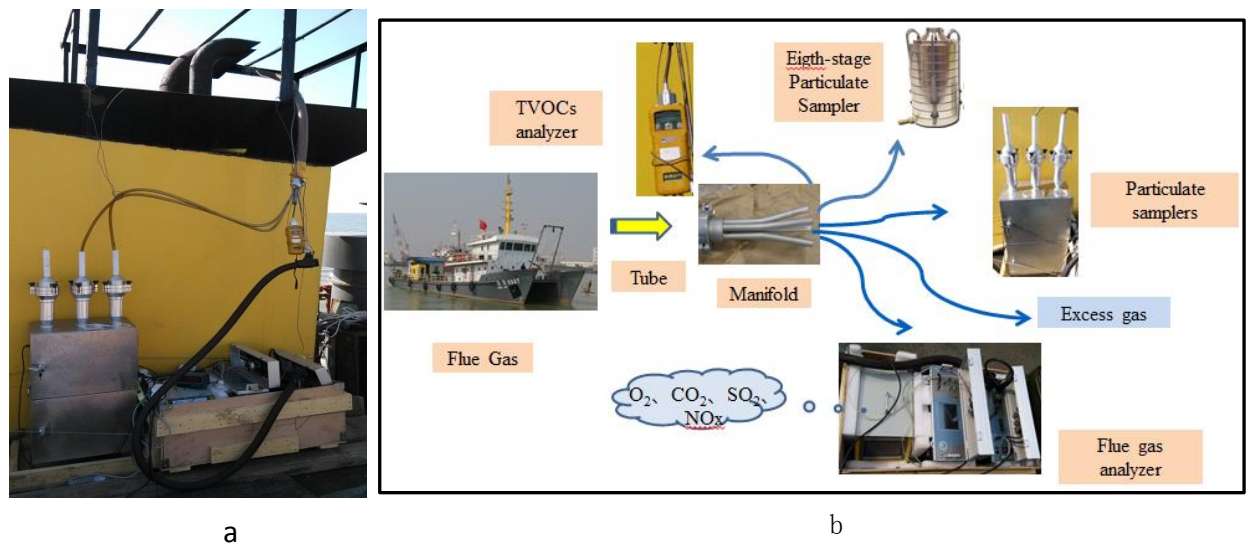


Figure S1 Real-world measurement system for vessels: a) on-board test picture, b) schematic diagram of the portable measurement system

The detection parameters for the gaseous matters have been added as an accessory in Table S3, including the detection method, range, resolution and accuracy etc. We can see that all the detection uncertainties are within the relative error of 5%. During our sampling, 3 to 5 replicate samples for each operating mode were collected, which could give the total error shown in Table S4 and Table S5, including the detection error and the artificial error.

Table S3 Detection parameters for the gaseous matters

<u>Component</u>	<u>Method</u>	<u>Range</u>	<u>Resolution</u>	<u>Accuracy</u>	<u>Time (T₉₀)</u>	<u>Conformity</u>
<u>O₂</u>	<u>Electrochemical sensor</u>	<u>20.95%</u>	<u>0.01%</u>	<u>±5% rel.</u>	<u>45 s</u>	<u>ISO 12039, CTM-030</u>

<u>CO₂</u>	<u>NDIR</u>	<u>5%</u>	<u>0.01%</u>	<u>±3% rel.</u>	<u>45 s</u>	<u>ISO 12039, OTM-13</u>
<u>CH₄</u>	<u>NDIR</u>	<u>5%</u>	<u>0.01%</u>	<u>±3% rel.</u>	<u>45 s</u>	
<u>NO</u>	<u>NDIR</u>	<u>1000ppm</u>	<u>1ppm</u>	<u>±3% rel.</u>	<u>45 s</u>	<u>ISO 10849, Method 7E</u>
<u>NO₂</u>	<u>NDIR</u>	<u>1000ppm</u>	<u>1ppm</u>	<u>±3% rel.</u>	<u>45 s</u>	<u>ISO 10849, Method 7E</u>
<u>SO₂</u>	<u>NDIR</u>	<u>1000ppm</u>	<u>1ppm</u>	<u>±3% rel.</u>	<u>45 s</u>	<u>ISO 7935, Method 6C</u>
<u>N₂O</u>	<u>NDIR</u>	<u>2000ppm</u>	<u>1ppm</u>	<u>±3% rel.</u>	<u>45 s</u>	<u>ISO 21258</u>
<u>VOCs</u>	<u>PID</u>	<u>10000ppm</u>	<u>0.1ppm</u>	<u>±5% rel.</u>	<u>-</u>	

NDIR, Non-dispersive Infra-red

PID, Photo Ionization Detectors

Both reviewers indicate a need for context in this manuscript. That context will need to improve to make the manuscript publishable. Simply stating that one needs Chinese emission factors won't be sufficient. While China is a large and important country, especially with regard to shipping, this manuscript does not contain an understanding of WHY Chinese ships might be different. China's fleet probably contains a wide range of ships and fuels. In that case, the emission factors presented here may not be representative of the Chinese fleet. Other emission factors, not measured in China, also may or may not be representative. Without an understanding of what causes a difference in emissions, one can't discuss whether measurements are representative.#

Thanks for your comment, and the reply is given as following:

The estimated contribution of shipping emissions on port cities, the type composition of offshore vessels in China, the fuel consumption and the latest policy made by Chinese Ministry of Environmental Protection aiming to limit the emissions from marine engines have been added in the revised manuscript (Line 5-7, Page 4, Line 14-17, Page 8, Line 8-22, Page 6).

It was estimated that 8.4% of SO₂ and 11.3% of NO_x were emitted from ships in China in 2013 with port cities were the worst effect areas (http://news.xinhuanet.com/politics/2015-06/08/c_127890195.htm). Conditions in

China differ substantially from those in other countries, such as in vessel types (more small motor vessels and the type composition of offshore vessels is shown in Table S1), different fuel standards compared with other countries (fuel meeting the GB/T 17411-2012 standard with sulfur contents of less than 3.5% m/m; however, the ISO 8217-2010 international standard has the maximum sulfur content according to the relevant statutory requirements that always have lower values, such as less than 0.1% in emission control areas), age of vessels (Chinese commercial vessels have an average age of 19.2 yr compared with 8.0 yr and 8.9 yr for Japan and Germany, respectively).

According to statistical data, the total oil consumption of vessels in China was 20.99 million tons in 2011, including 10.99 million tons bonded oil and 5.93 million tons domestic trade oil, with light fuel oil account for 40% of the domestic trade oil and 25% of the total consumption (shown in Table S2).

But because of the serious air pollution these years in China, emission limits for the main sources such as vehicle exhaust, coal combustion, biomass combustion and raise dust have becoming more and more stringent. A draft aimed to limit the emissions from marine engines set by Ministry of Environmental Protection, which is named Limits and measurement methods for exhaust pollutants from marine compression ignition engines (CHINA I , II), is on soliciting opinions. It has set the limits of CO, HC, NO_x and PM for different kinds of vessels, which mainly based on the Directive 97/68/EC set by EU and 40 CFR part 1042 set by EPA. Besides, an implementation plan has released by Ministry of Transport of the People's Republic of China in December 2015 aiming to set shipping emission control areas to reduce SO₂ emissions in China (Ministry of Transport of the People's Republic of China, 2015). All the regulations were set mostly based on other directive and regulations. And therefor, detailed measurement data in China are in urgent need for the further policy making that more fit current situations of vessels.

Initially, it was hoped that the choice of measurement ships would reflect the shipping fleet in general, i.e. in terms of engine type (engine speed and power output), fuel used, engine age and mode of operation, with more than 10 vessels planned to

test. However, consideration was given to the practicalities involved with the measurements, i.e. installation of sampling systems, external conditions, etc. Besides, time and economic constraints weighed heavily and only several shipowners willing to participate in the project. Thus, the chosen vessels of different engine powers with diesel used represent a compromise.

Three offshore vessels with different total tonnage and engine power were tested in our study. We inferred that the engine type was the most important influence factor on shipping emissions. All the three vessels are high-speed and medium-speed engines. Statistics have reported that high-speed and medium-speed engines could account for more than 95% of the latest produced vessel engine since 2008 in China, with most of which used in inland vessels and offshore vessels. So, the test vessels have a certain degree of representativeness from this point.

We may not fully understand what causes a difference in emissions. But the simple fact that these ships are measured in China is not the cause of a difference in emissions. These results may be applicable beyond China, and other emission measurements may be valid for Chinese ships. Both reviewers ask for a discussion that relies on physical factors, such as fuel or ship type. Even a simple breakdown for Chinese ships or usage in Chinese waters would be helpful. When comparing with previously published results, comparison with regard to the type of ship would be more instructive. I suggest that authors provide discussion to help readers understand how these measurements, as well as other measurements, would best be used to develop an emission inventory. This could be accomplished, in part, by comparing with a wider range of measurements, as the first reviewer requests. The second reviewer also suggests that authors update their understanding of how measurements are typically presented. Both of these improvements would provide a better context for the potentially useful measurements presented here.#

Thanks for your comment, and the reply is given as following:

Influence factors such as engine type and fuel type have been discussed in the revised manuscript (Line 13-27, Page 18). We inferred that engine type could have more impact on the emission factors.

In order to compare the differences of emission factors from vessels in this study and other non-road diesel engine vehicles, fuel-based emission factors for CO, NO_x and PM were given in Table S7, including military non-road heavy duty diesel vehicles, excavator and wheel loader and other diesel trucks. It could be deduced that engine types have significant impact on emission factors such as non-road heavy duty diesel vehicles always have much higher NO_x emission factors compared with common diesel trucks. Besides, one interested thing should be mentioned that Chinese diesel engines always have higher NO_x and PM emission factors which may be caused by the less strict emission standard applied for diesel vehicles in China. Similarly, the engine type might be an important cause of the different emissions, such as HH had much higher pollutants emissions with an engine produced in China and yet DFH's engine produced in Germany. Besides, emission test for a high-speed marine diesel engine with different kind of diesels showed that, diesel type had limited influence on emissions such as NO_x, CO and CH, but a significant impact on PM emission (28.9-41.5%) because of the different sulfur content in fuel (Xu, 2008).

Because there is no public data of the distribution of engine types in China, we could not give official statistics. But through personal relationships of Marine Affairs Bureau, we got the distribution of vessels through gross tonnage in 2014 in offshore area of Yangtze River Delta that is one of the three largest shipping areas, which is shown in the following table. Unfortunately, we are not allowed to public the data in the manuscript. The gross tonnage of our test vessels are 307, 3235 and 602, respectively, accounting for 34.7% of the total vessels, which could has certain degree of representation.

Distribution of vessels through gross tonnage in 2014 in offshore area of Yangtze River Delta

Gross tonnage (t)	Percentage (%)
10000-49999	3.7
3000-9999	12.7
1000-2999	50.3

500-999	13.8
100-499	8.2
<99	7.0

More power-based emission factor data are added in the revised manuscript in Table 4, including data from US EPA, Khan et al., Agrawal et al.(2008), Moldanova et al.(2009), Celo et al.(2015) and so on.

Besides, detailed calculation method about average EFs for each vessels, converting power-based emission factor to fuel-based emission factor and Carbon balance method have been added and explained in the revised manuscript.

Average EFs for each vessel were calculated based on actual operating conditions, as shown in Formula (4) (Line 3-6, Page 11):

$$EF_{X,A} = \sum_{X,i} EF_i \times P_i \quad (4)$$

where $EF_{X,A}$ is the average EF for species X, EF_i is the EF for operating mode i for species X, and P_i is the percentage of time spent in operating mode i during the shipping cycle.

The converting power-based emission factor to fuel-based emission factor was added as Formula 5 in the revised manuscript (Line 7-11,Page 11), shown as following:

$$EF_{X,P} = EF_X \cdot FCR \quad (5)$$

where $EF_{X,P}$ is the power-based emission factor for species X (g kW h^{-1}), FCR is fuel consumption rate for each vessel ($\text{kg fuel (kW h)}^{-1}$).

Carbon balance method was used in this study to give the emission factors, which assumes that all carbon in the fuel was emitted as carbon-containing gases (CO , CO_2 , and TVOC) and carbon-containing particulate matter. So Formula 1 was given as shown below:

$$C_F = R_{FG} \times (c(C_{CO}) + c(C_{CO_2}) + c(C_{PM}) + c(C_{TVOC})) \quad (1)$$

R_{FG} could be calculated according to this formula since all the other parameters could be measured during or after the sampling.

The correction for CO_2 has been implemented in the carbon balance equation.

Background CO₂ concentration (the CO₂ concentration of ambient air) was subtracted to ensure all the carbon was transformed from the carbon in the fuel. In the same way, when other emission factors were given, background concentrations had also been subtracted, such as CO, NO_x, etc.

Other minor revisions made will be shown in the revised manuscript.

Thanks again.

Best regards,

Fan Zhang, Representative of all the authors