Reply to Referee 2#

Thanks very much for your comments. Before giving our reply, we would like to present several statements to help you understand further about the current shipping emission situations in China, which will be added and explained in the revised manuscript.

Firstly: The measurement data of shipping emissions are in urgent need. Laws and regulations for shipping emissions have already managed to make, which require the basic measurement data very much. Besides, estimating contribution of ships to air and calculating emission inventories of ships based on local emission factors are essential in China because of the differences of ships with other countries. So this study is focusing on adding the measurement database of shipping emissions in China. And to our knowledge, only very limited study has carried on on-board measurement of ocean vessels in China. Even though our work is not comprehensive, it is a start to have a look at the emission conditions of vessels in China.

Secondly: High speed and medium speed engines are the predominant engines used in vessels of offshore and inland rivers in China, which always take light diesel as fuel. Though only three offshore vessels' data were reported in this study, they were typical offshore diesel vessels that could, to some extent, represent the emission conditions from a low level to a relative high level in China.

Furthermore: On-board test is really hard due to the unpredictable of field work, the expensive rent for vessels, unwillingness of vessel owners, and also the lower online operating parameter devices on the ships, etc. We have finished five vessels till now, and is carrying on measurement of heavy fuel vessels now. More accurate data will be provided to scientific research and policy-making in our follow-up work.

Our replies are given as following according to your comments:

p.23509 14-18...low engine power vessel, ... higher engine power vessel this is a bit confusing description of the vessels, especially as also medium-speed and high speed engines and different engine loads are used to describe the experiments. It looks like there are two smaller and one larger vessels, maybe this, or using the vessel abbreviations would make the text easier. # Thanks for your comment, and the reply is given as following: The vessels' abbreviations have been added in the revised manuscript (Line14-17, Page 2).

Observed concentrations and emissions factors for carbon monoxide, nitrogen oxides, total volatile organic compounds, and particulate matter were higher for the low engine power vessel (HH) than for the two higher engine power vessels (XYH and DFH).

p.23512, l. 15 IMO legislation in ECAs is not decided by EU environmental ministers, the same rules apply for the North Sea & English Channel and for the Baltic sea through all years, not only 2004-2010.

Thanks for your comment, and the reply is given as following: The legislations set by IMO that applied in ECAs have been checked and rewritten in the revised manuscript (Line 25-28, Page 5, Line 4-6, Page 6), which are shown as following:

Ships operating in the emission control areas (ECAs) (the Baltic Sea, the North Sea, the North America and the Caribbean of US) should use fuels with sulfur less than 0.1% m/m since January 2015.

Emission standard of Tier II for NO_x set by MARPOL VI has been executed since January 2011 in ECAs, and more stringent rules of Tier III will be executed from January 2016.

Other legislations in EU and USA also have been checked and rewritten in the revised manuscript (Line 28-30, Page 5 and Line 1-4, Page 6), which are shown as following:

Even more stringent limits have been laid down in some national or regional regulations. For example, in some EU ports, seagoing ships at berth are required to switch into using fuels of under 0.1 % m/m sulfur since 2010 (The Council of the European Union: Council Directive 1999/32/EC of 26 April 1999, Official Journal of the European Communities, 13-18); both marine gas oil and marine diesel oil used in water area within 24 nautical miles of coastline in California should have sulfur content less than 0.1 % m/m since 2014 (California Code of Regulation Titles 13 and 17).

Besides, the first draft aimed to limit the emissions from marine engines in China

is on soliciting opinions now, and the details are shown in revised manuscript (Line 8-22, Page 6), as shown below:

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.

p.23514, Operating modes – the normal is to express operating modes as % of max engine load and not ship speed which is affected by external conditions as currents, wind-speed e.c.t. The comparison with other data is difficult when using vessel speed when most of the other published factors are based on engine load.

Thanks for your comment, and the reply is given as following: Vessel speed was used in the study to give different operating modes. According to ISO 8178-E3, speed and torque of the test engine need to be measured during the sampling to calculate the effective power, which could give the load rate. But unfortunately, we were not allowed to install any detector for the engine of the vessels, only real-time engine speed could be read through the tachometer. We were more focused on the actual navigation conditions, and more than three samples for each mode were collected to give an average value to reduce the influence form external conditions.

Besides, vessel speed used as a variable to check out the variations of pollutants from ship was applied before, and good results have also been obtained (Cappa et al.,

2014). We considered it was also feasible using vessel speed to give different operating modes.

p.23516, Formula 2: The flue gas emission rate R(FG) is essential for the calculation, how was it obtained? Has the correction for CO2 in the engine inlet air been implemented in the carbon balance equation? In formula 3 – What is meant with 'background subtracted?

Thanks for your comment, and the reply is given as following: 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, CO2, and TVOC) and carbon-containing particulate matter. So Formula 1 was given as shown below:

$$C_{\rm F} = R_{\rm FG} \times (c(C_{\rm CO}) + c(C_{\rm CO_2}) + c(C_{\rm PM}) + c(C_{\rm TVOC}) \tag{1}$$

 $R_{\rm FG}$ 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 CO_2 concentration (the CO_2 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.

p. 23517 Part 3 – what is reason of presentation and comparison of concentrations in exhaust? These vary largely among the different engines and operation conditions and do not allow any general comparison.#

Thanks for your comment, and the reply is given as following: As we mentioned in the Introduction, concentrations in exhaust of inland ships on the Grand Canal of China, the only test vessels reported in China, were also presented and also other studied (Sinha et al., 2003;Corbett et al., 1999;Williams et al., 2009;Berg et al., 2012). Even though there were big differences among different engines and operation conditions, comparison could be done to a certain extent, such as the differences for different power engines. Besides, this study is focused on presenting detailed basic data, initial concentration data was also given for other studies to compare or recheck.

p. 23519 l. 1-3 CO2 emissions - ship HH had actually rather bad and not 'high'

combustion efficiency with 2-7.5% C emitted as other but CO2. p. 23521 – OC depends very much on dilution of the exhaust analyzed and OC analyzed on PM sampled without dilution cannot be directly compared with OC analyzed on samples from diluted exhaust.#

Thanks for your comment, and the reply is given as following: The sentence was rewritten in the revised manuscript (Line 6-10, Page 13), shown as following: Under actual conditions, CO_2 emissions were 2940–3106, 3121–3160, and 3102–3162 g kg⁻¹ fuel for HH, DFH and XYH, respectively, which means they had combustion efficiencies with 92.5–97.8%, 98.5–99.7% and 97.8–99.7% in terms of CO_2 for these three vessels.

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. We just give the actual OC to EC ratio under undiluted situation. During our later sampling, both samples with and without dilution were collected to give the differences between them. The details are shown in revised manuscript (Line 30, Page 15 and Line 1-8, Page 16), as shown below:

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 cruse and transient modes even though higher in cold-start/idle and creep modes (Shah et al., 2004).

p. 23522 – Section 3.3 – How were the emission factors for different operation modes averaged? There are standardized methods for averaging, were these applied?#

Thanks for your comment, and the reply is given as following: Average EFs for each vessel were calculated based on actual operating conditions, as shown in Formula (4) (Line 3, Page 10):

 $\underline{EF_{XA}} = \sum_{Xi} \underline{EF_i} \times \underline{P_i} \tag{4}$

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.

There is no standardized method for averaging emission factors for different operating modes in China. Only a draft aimed to limit the emissions from marine engines set by Ministry of Environmental Protection (Limits and measurement methods for exhaust pollutants from marine compression ignition engines (CHINA I, II)) is on soliciting opinions. Even though other standard such as ISO 8178-4 (Reciprocating internal combustion engines -- Exhaust emission measurement -- Part 4: Steady-state test cycles for different engine applications) has standardized method for the calculation of weighted emission factors, it is calculated as:

$$E_{WM} = \frac{\sum_{i=1}^{i=n} (m_i \times WF_i)}{\sum_{i=1}^{i=n} (p_i \times WF_i)}$$

where E_{WM} is the overall weighted emission factor (g/kW-hr), m_i the emission factor for i mode (g/hr), WF_i the weighted factor for i mode, and p_i the engine load for i mode (Khan et al., 2013). In previous study, weighted emission factor was given for vessels under different engine loads in order to have comparison of measured emission factors with literature data (Agrawal et al., 2008); average emission factor were also given for load conditions of 85-110% from one serial 4-stroke medium-speed marine diesel engine (Petzold et al., 2010). Unfortunately, we have no measurement engine load, so a weighted average emission factor was calculated based on the actual operating modes (added in supporting information as Table S6) in this study.

p. 23523 l. 3-4 – How were the Tier-1 limit emissions calculated in g/kg-fuel? The specific fuel consumption needed for the calculation need to be shown. This is also the case for the power-based emission factors Table 3. Fuel-based emission factors – The table is mostly missing information about fuels used by the vessels and

their sulphur content which is essential for EFs both for SO2 and for PM.#

Thanks for your comment, and the reply is given as following: Another formula that 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: $EF_{X,P} = EF_X \cdot FCR \qquad (5)$ 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 (kg fuel (kW h)⁻¹).

According to Formula 5 and the fuel consumption rates that obtained from Engine Performance Curve of each vessel combined with real-time engine speed in each operating mode, power-based emission factors could be calculated, which have been presented in Table 4.

The IMO Tier I emissions limit for NO_x is $45.0 \times n^{-0.2}$ g kWh⁻¹ (n, rated speed, 130 < n < 2000 rpm). The rated speeds *n* for the vessels were shown in Table 1, so we could get the power-based emissions limit. Fuel-based emissions limit could be calculated combined with fuel consumption rate of each vessel that was also given in Table 1. Thus, the emissions limits for HH, DFH, and XYH would be 54.5, 57.5, and 56.5 g kg⁻¹ fuel, respectively.

All the fuel used of the test vessels were diesel. And the fuel analysis results were shown in Table 2, where sulfur content for each kind of diesel could be found. All of these fuels had relatively low sulfur contents ($\leq 0.13\%$ m) and low metals concentrations (V, Al, Si, Pb, Zn, Mn, etc.).

| | Units | HH | DFH | ХҮН |
|-----------------------|---------------------|--------|---------|---------|
| Total calorific value | MJ kg ⁻¹ | 45.44 | 45.40 | 45.50 |
| Net calorific value | MJ kg ⁻¹ | 42.51 | 42.48 | 42.55 |
| Ash content | %m | 0.001 | < 0.001 | < 0.001 |
| Sulfur (S) | %m | 0.0798 | 0.0458 | 0.130 |
| Carbon (C) | %m | 86.66 | 86.40 | 86.49 |
| Hydrogen (H) | %m | 13.32 | 13.22 | 13.44 |
| Nitrogen (N) | %m | < 0.2 | < 0.2 | < 0.2 |
| Oxygen (O) | %m | <0.4 | <0.4 | <0.4 |

Table 2 Results from the fuel analysis (diesels)

p. 23524 – section 3.5 – Since the Tier is based on power-based EF it would be good to look at these as well, these EFs are usually stable. The variability of the fuel-based EFs is related to the power-based ones through inverse specific fuel consumption which can have similar shape as seen on fig. 4#

Thanks for your comment, and the reply is given as following: Both fuel-based EFs and power-based EFs were given in this study, as different kind of data for different purpose, such as detailed fuel-based EFs are more useful for inventory estimating and power-based EFs are more easier for looking at the emission situation and comparing among different vessels.

The fuel consumption rates have little change in different operating modes for the test vessels, which are queried from the Engine Performance Curve. Power-based emission factor for NO_x would have the similar variability trends of fuel-based emission factor as shown in Fig.4. Furthermore, fuel-based emission factor is calculated directly from the measurement data, which would be closer to the actual condition than power-based emission factor.

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Best regards,

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