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Discussions

Interactive comment on "Size-segregated characteristics of OC, EC and organic matters in PM emitted from different types of ships in China" *by* Fan Zhang et al.

Fan Zhang et al.

fzhangtj@tongji.edu.cn

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Response to the comments

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Title: Size-segregated characteristics of OC, EC and organic matters in PM emitted from different types of ships in China

Author(s): Fan Zhang, Hai Guo*, Yingjun Chen*, Volker Matthias, Yan Zhang, Xin Yang, Jianmin Chen Corresponding author: Hai Guo (hai.guo@polyu.edu.hk)

Yingjun Chen (yjchenfd@fudan.edu.cn)

The constructive comments of the reviewers are highly appreciated. We have revised the manuscript accordingly. Our point-by-point responses (in black) to each comment are listed below. And the modifications in the revised manuscript are marked in red. Please see the manuscript for details.

Reply to Referee 1#

I lack a discussion on the relevance of the study in a larger context. It should be elaborated upon from the viewpoint that the engines are relatively small, high speed marine engines, and only representative for smaller marine craft.

Response:

The comment is well noted. Explanation about the studied vessels has been added in Section 2.1 according to the comment. Most of the studied vessels in this study were indeed with high-speed marine engines. However, YK was a large ocean-going low-speed engine ship with heavy fuel oil used, while DFH was a medium-speed engine ship with marine diesel used (See Table S1 for details). Hence, we considered that they were representative of large/medium marine crafts in China to some extent.

Revision was made as follows (Lines 154-156, Section 2.1):

"Notably, most of the vessels had high-speed marine engines in this study, except for YK and DFH, which had low-speed engine and medium-speed engine, respectively."

I find that very little attention is given to the engine and combustion characteristics. It is stated that "Overall, fuel type, fuel quality, engine type might have higher influence on particle mass distributions from ships than the operating mode", but it seems from Figure 1 that also operational mode is important for individual ships. Results on EC and OC at different engine loads are not discussed. It is necessary that the authors also describe whether the presented results are average values from all samples from the tested engines. This would imply mixing of samples representative for different engine

loads. #

Response:

Thanks for pointing out these. More parameters of the tested engines have been added and shown in Table S1. The influence of combustion efficiency on the size distribution, organic formation, and PAHs distribution from different types of ships was briefly discussed in this manuscript in lines 248 to 256, lines 292to 297, lines 385 to 388, etc. Since all the tests were onboard (in-situ) measurements, rather than the bed tests that could control all the conditions in this study, we could not draw absolute conclusions about the engine and combustion characteristics here. Therefore, inferences caused by comprehensive influence factors were given and discussed in this study. Further study of a bed test for a low-speed HFO engine is in progress, from which we hope we can get more detailed and direct influence characteristics.

As for the size distributions of particles in different operation modes, we did not obtain an absolute conclusion before because not all the ships showed consistent size distribution patterns in different operation modes. However, more coarse particles were indeed found in operating modes with higher engine loads. According to the comments of the reviewer, the sentence has been revised in lines 257 to 264 in the revised manuscript as follows: "The size-segregated particle mass distributions in different operating modes were also compared in this study. The results showed that when the ships were operated with higher engine loads, there were more particles in coarse modes for most of the ships, which was similar to a previous study on mass distribution from measurements onboard of three ships (Fridell et al., 2008), but different from another study on a marine engine that reported the particle mass distribution centered at 0.1-0.2 μm with much fewer coarse particles under at-berth condition compared to maneuvering and ocean-going conditions (Chu-Van et al., 2017)." The conclusion has also been improved as "Overall, fuel type, fuel quality, engine type, and operating mode might have comprehensive influence on particle mass distributions from ships." (Lines 269 to 271 in the revised manuscript).

Actually, emission factors and the ratios of OC and EC at different engine loads have been discussed in our previous studies (Zhang et al., 2016; 2018). In this study, we focused on the size-segregated OC and EC distributions. Therefore, no further discussion of OC and EC at different engine loads was given here. Besides, due to the relatively small sampling size of size-segregated samples under different engine loads, not all the operating conditions could be included in the onboard tests. Hence, no solid conclusion could be drawn. However, as we mentioned above, a bed test for a low-speed HFO engine is in progress under different engine loads (0%, 25%, 50%, 75%, and 90%). We hope that we could get detailed and robust results of OC and EC distributions at different engine loads from this bed test.

At last, we would like to clarify that the current results are obtained based on the average values of all samples from the tested engines. Relative contents have been added in lines 275 to 276 and lines 472 to 474 in the revised manuscript.

"Notably, OC and EC levels given in this study were the average values of all samples that were classified into these three types."

"Same as OC and EC, the levels of PAHs and n-alkanes provided in this study were the average values of all samples that were classified into these three types."

The abbreviations of ship types appear inconsistent and subjectively applied. Inconsistent in the sense that two types are named by installed power (high or low) and a third by fuel (HFO). Further, it is difficult to follow the specific ship name abbreviations. I suggest to use a different naming for ships that makes it easy to relate names to the relevant investigated category. An argumentation to support the cutoff in engine power between the chosen categories is necessary. It is not apparent to me why it should matter whether the engine is on a fishing vessel, a research vessel or an engineering vessel etc. Rather, categories should be based on whether it is a 4-stroke cycle or 2-stroke cycle and the engine speed. To use "diesel" as a term for distillate marine fuels can be confusing since diesel is mainly referred to when discussing engine types

rather than fuels (also heavy fuel oil is used in marine diesel engines). “Marine distillate oil” could replace “diesel” to avoid the confusion on whether a fuel or engine type is discussed. #

Response:

Thank you for pointing out these confusions. Firstly, according to the reviewer’s suggestion, we added the engine type (2-stroke or 4-stroke) in Table S1. YK was a 2-stroke low-speed engine ship, and the others were 4-stroke engine ships (1 medium-speed engine and 10 high-speed engines). Among the 4-stroke engine ships, fishing boats were classified as a special type, because they usually had much smaller engine power with older engine, and used non-standard diesel with much larger quantities (6 times the amount of water transport vessels in China) than other water transport vessels. Secondly, the fishing boats with engine power less than 300 kW could account for 75.6% of the total fishing boats in China, and the China registered ships with engine power less than 250 kW could account for 71.6% of the total registered ships in China. Therefore, combined with the studied ships, we considered 300 kW as a threshold between high engine power and low engine power in this study. Thirdly, fuel type is also a significant influence factor for the emitted pollutants. YK used typical heavy fuel oil as fuel, and the fishing boats used non-standard diesel as fuel, while the other ships used typical marine distillate oil as fuel. Therefore, after comprehensive consideration of the engine type, engine power and fuel type, we classified the studied ships into three categories in this study. We renamed the categories and abbreviations accordingly, namely, 4-stroke low-power diesel fishing boat (4-LDF), 4-stroke high-power marine-diesel vessel (4-HMV) and 2-stroke high-power heavy-fuel-oil vessel (2-HHV). All the abbreviations have been changed in the revised manuscript. Finally, due to the fact that our research on shipping emissions was a complete series, including the emission factors and characteristics of gaseous pollutants, PM and its chemical composition, the influence factor, the organic matters, and the total emissions in our previously published papers (Zhang et al., 2016, 2018, 2019), we would like to retain the specific ship

name abbreviations in this study for easy follow-up.

The “diesel” has been changed to “marine distillate oil” or “diesel fuel” in the revised manuscript according to the helpful comments.

The work includes a lot of literature references and reasoning about the particle number size distributions, which could possibly be cut down to make the manuscript more concise. No own results of PN is presented.

Response:

We thank the reviewer for his/her valuable comments. The contents and literature references in lines 217 to 222 in the original manuscript have been deleted. The reasoning about the particle number size distributions was also removed in lines 338 to 339 in the original manuscript. The description was also improved in lines 358 to 365 in the revised manuscript.

“In these processes, several factors can influence the size of the emitted particles: 1) surface growth involves the attachment of gas-phase species to the existing particles and results in increase of PM diameter and mass; 2) aggregation leads to chains and clusters of primary soot particles and an increase in particle size (to 10-100 nm in diameter) (Heywood, 1988); and 3) oxidation occurs during the formation and growth processes of the particles, resulting in the formation of gaseous species and reduction of soot particles and their precursors.”

Influence of measurement methods can be very important. I strongly recommend that the sampling system, the dilution ratios and the fuels used are described at least in the supplementary material. I have not had a chance to review this.

Response:

Thanks for the comments. In this study, the size-segregated particle samples were collected directly from onboard tests through two sampling systems shown in the following Figures R1 and R2. It is noteworthy that these two sampling systems were carefully introduced in our previous studies (Zhang et al., 2016, 2018). Therefore, we did not put them in the supplementary material in this study. Please note, sampling system (I) was used for the high-power diesel vessels of HH, DFH, XYH, and sampling system (II) was used for the other vessels. The difference of the two sampling systems was that the sampling system (I) had no dilution system due to the limitation of the sampling system used in our early sampling period, and consequently all the particles were collected directly from the flue gas. Dilution ratios for all the tested ships ranged from 1.0 to 4.2 according to the actual sampling conditions, which were added in Table S3 in the supporting material. Moreover, the detailed parameters of all the fuels used for each tested vessel are given in Table S2. Some descriptions about the fuels were also added in lines 158-160 in Section 2.1.

"Marine distillate oil was used as the fuel of the high-power-diesel vessels. Non-standard marine distillate oil was used for the 4-stroke low-power diesel fishing boats, which was introduced in our previous study (Zhang et al., 2016), while heavy fuel oil was used for the tested HFO vessel in this study."

Figure R1 Sampling system (I) of the onboard test

Figure R2 Sampling system (II) of the onboard test

I find that in the results and discussion section it can be difficult to understand what is findings from this study, and what was previously known. E.g. page 16 lines 371-390 and page 21 line 501 to 505. I think this can be solved by changing the tense in the presentation.

Response:

Sorry for the confusion. The tense was changed in some sentences, and some descriptions were improved in order to distinguish the findings of this study from previous studies. Revisions were shown in lines 371 to 390 and lines 499 to 504 in the revised manuscript.

"Soot particles are mainly formed through pyrolysis of diesel fuel and lubrication oil, and the organic fraction is formed through incomplete combustion of fuel and lubrication oil (Zetterdahl, 2016). Incomplete combustion of fuel and lubrication oil can significantly enhance the formation of fine particles, which might be the reason for the higher OC to EC ratio in particles with Dp < 0.43 μm in this study. This study also found that most EC contents were in the particles with Dp < 1.1 μm, coincident with the fact that soot is primarily in accumulation mode with particles of 0.1 < Dp <1 μm (Kasper et al., 2007). Although OC was also concentrated in fine particles with Dp<1.1 μm (Section 3.1), its percentage was higher in particles with Dp<0.43 μm, and lower in particles with 0.43<Dp<1.1 μm, compared to the EC distribution. Therefore, OC to EC ratios were the lowest in particles with 0.43<Dp<1.1 μm for all the tested ships. For the coarse particles in this study, though non-carbonous components were the dominant parts, OM increased after the cooling of the exhaust gas while content of EC and ash remained unchanged through adsorption of hydrocarbons and other volatile compounds (Zetterdahl, 2016), which enhanced the formation of coarse particles. Consequently, OC to EC ratios also showed high values in coarse particles with Dp>3.3μm. 2-HHV showed higher OC to EC ratio than the diesel fuel ships, which might be caused by the relatively lower combustion efficiency. Similarly, because 4-HMV ships had higher combustion efficiencies due to the higher fuel quality and better engine maintenance in this study, OC to EC ratios of 4-HMV showed the lowest levels."

And,

"The relatively longer exhaust gas channel and higher temperature in the channel of 2-HHV enhanced the formation of new secondary nucleation-mode and/or accumulation-mode particles from gas-phase organic compounds, leading to higher proportions of organic matters in fine particles, which has been proved in a previous study (Kittelson, 1998)."

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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-363/acp-2019-363-AC1-supplement.zip

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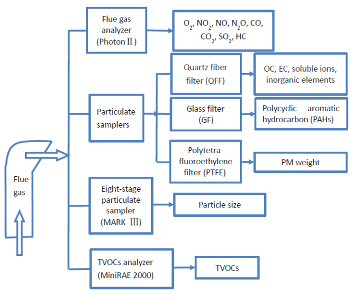

Figure R1 Sampling system (I) of the onboard test

Fig. 1. Figure R1

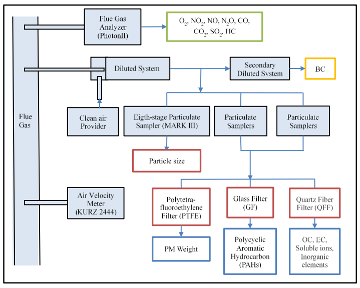

Figure R2 Sampling system (II) of the onboard test

Fig. 2. Figure R2