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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 2#

1. # It's authors' duty to explain what's new in this paper compared to several other papers published by the same group, especially the one on Atmospheric Environment 2019. Both titles indicate similar contents. #

Response:

Thanks for your comment. The paper published on Atmospheric Environment 2019 was focusing on the emission factors, profiles and characteristics of organic matters from the total particles emitted from ships. This information could provide some basic data for inventory estimation, source apportionment, and implication for source identification and health influence of ship emissions. However, this manuscript is focusing on characteristics of OC, EC and organic matters from size-segregated particles. As shown in the manuscript, the proportions and characteristics of these compositions varied significantly in different particle sizes, which were worthy being analyzed carefully. This information could provide further knowledge of the composition of particles in different sizes, implication for particle formation mechanism of ship exhaust, and also potential health impact, source apportionment of particles in different sizes. For example: the different profiles of chemical components in size-segregated particles implied that size-segregated chemical profiles should be considered when source apportionment was conducted. Furthermore, this study found more toxic organics such as PAHs in small particles emitted from fishing boats, suggesting the necessity of more stringent control on this type of boats in China.

2. # The results were not organized in a clear way which made them very hard to follow. 1) For different particle size bins, what are the chemical component profiles? No figure gives a comprehensive component profile. Only the OC/EC percentages compared among bins were provided. Authors failed to touch the whole picture of the "size-segregated characteristics", which should be the most important part of this study. For example, EC and OC were found very low for coarse PM. Then, which components are the major part for coarse PM? In addition, without a total mass analysis, it's impossible to judge the reliability of the sampling and analysis. 2) If the whole picture of size based chemical profile could be provided, then it's OK to discuss the distribution for each component in different size bins. However, this information is not very important compared with part 1). Currently, authors spent too much pages on discussing this, including Fig. 2, 6 and etc. 3) All figures were displayed in percentage or ratio. The mass of OC, EC, PAH or others should be provided directly. Is it still necessary to provide so many figures if the mass could be given? 4) Figure 1 is confusing. 'YK, GB1...' should be replaced with ship types, e.g. 'HDPV'. #

Response:

The valuable comments are highly appreciated.

1) Unfortunately, we could not get the detailed chemical profiles of the size-segregated particles in this manuscript. We only focused on OC, EC, organic matters of 16 priority PAHs, and n-alkanes in the particles. Other inorganic matters such as water-soluble ions and metal elements were not analyzed because the sample volume was too low of the particles in each size bin, especially in coarse particles. We inferred that inorganic matters of ash and hydrated sulfates could be the dominant component in coarse particles, which has been confirmed by a previous study (Moldanová et al., 2009). The detailed chemical profiles of particles in different size bins still need further investigation, which is also a target we are working on. Besides, we compared the total PM concentrations between the two sampling methods, namely the TSP sampling method and the Andersen sampling method. The result showed that they had a Pearson cor-

relation coefficient of 0.917, and the correction was significant at the 0.05 level. As described in Sections 2.2, 2.3 and 2.4, all the sampling processes and chemical analysis were carried out according to standard methods, which showed reliable quality assurance and quality control. Therefore, we believed the sampling and analysis were reliable.

2) As explained above, we could not quantify the size-segregated inorganic matters in this study. Instead, carbonaceous matters of OC, EC, PAHs and n-alkanes in the PM were the focus in this study. Even though the detailed chemical profiles of size-segregated PM were not able to give, the results were still meaningful for the implications of climate change, source apportionment, health influence, and formation mechanism of organic matters, as presented and discussed in this manuscript. We fully agree that the whole picture of size based chemical profile is very important and needs to be figured out correctly in the future.

3) Firstly, the emission factors of the total PM and size-resolved particle mass distributions are given in Figure 1 in the manuscript. It could be seen that the PM emission factors varied significantly among different ships, from 0.08 to 19.01 g (kg fuel)-1. When the mass was distributed to different size bins, there were still large variations among the different types of ships (such as the OC and EC emission factors in different particle size bins shown in the following Figure R1). Due to the significant variations of the absolute mass/emission factor, it would be difficult to obtain the common pattern of the distributions and characteristics of OC, EC, and organic matters. Therefore, figures in percentage or ratio were displayed in this manuscript. Besides, since the distribution patterns of total OC, EC, PAHs and n-alkanes in different particle size bins are different, we consider that figures such as Figure 2 and Figure 6 are necessary in the manuscript.

Figure R1 OC and EC emission factors in different particle size bins

4) Figure 1 has been improved in the revised manuscript, and 2-HHV, 4-HMV, and

4-LDF have been added in the figure (shown as follows).

Figure 1 Emission factor for total PM and size-resolved particle mass distributions with different modes for the 12 tested ships

3. # Presentation quality in text also needs to be improved. 1) In abstract, line 30, 'in fine particles, OC and EC were the dominant components'. line 34, 'OC and EC have the lowest values for 0.43 to 1.1 um'. Are they still dominant? 2)Line 34, What are the OC1, OC2 and OC3? 3) Line 214, how can 5% be called the large proportion? 4) line 282, HFOV vessels should be HFOV. and 'HPDV ships' should be 'HPDV'. And this sentence is confusing. What's the meaning by 'HPDV accounted for 23%....'? Compared with what kind of ships? #

Response:

Thank you for pointing out these. OC and EC were indeed the dominant components in fine particles. In line 34, we meant that the OC to EC ratios had the lowest values for particles between 0.43 and 1.1 μm. They are not contradictory.

1) As explained in lines 392 to 397: OC and EC were tested according to the IMPROVE-A protocol in the thermal-optical carbon analysis. OC1, OC2, OC3, OC4, EC1, EC2, EC3, and pyrolysis carbon fragments were tested under different temperatures and conditions, which could be read directly from the result file (shown in the following Figure R2 as an example). Then OC and EC could be calculated according to the protocol. We analyzed the OC1 to EC3 fragments in this manuscript to help understand the different formation processes of particles.

Figure R2 Example of test results for OC and EC fragments

2) Sorry for the confusion. "large proportions" has been changed to "non-ignorable proportions" in the revised manuscript in line 221.

3) "HFOV vessels" has been revised to "2-HHV", and "HPDV ships" has been changed to "4-HMV" in the revised manuscript in lines 285 to 290. The abbreviations of ship

types have been changed to 4-stroke low-power diesel fishing boat (4-LDF), high-power-diesel vessel (4-HMV) and 2-stroke high-power heavy fuel oil vessel (2-HHV) (see Table S1 for details).This sentence has been improved as "For example, 4-HMV only accounted for 23% OC and 27% EC in particles with Dp < 0.43 μm compared to 2-HHV which had 75% OC and 66% EC in particles with Dp < 0.43 μm. This is in accordance with the characteristics of total PM mass distributions; that is, diesel fuel vessels had relatively smaller proportions of fine particles with Dp<0.43 μm and larger proportions of coarse mode particles than HFO ships."

Reference:

Moldanová, J., Fridell, E., Popovicheva, O., Demirdjian, B., Tishkova, V., Faccinetto, A., and Focsa, C.: Characterisation of particulate matter and gaseous emissions from a large ship diesel engine, Atmos. Environ., 43, 2632-2641, 10.1016/j.atmosenv.2009.02.008, 2009.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-363/acp-2019-363-AC2-supplement.zip

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-363, 2019.

ACPD

Interactive comment

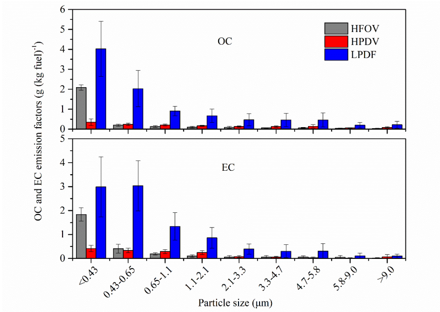

Figure R1 OC and EC emission factors in different particle size bins

Fig. 1. Figure R1

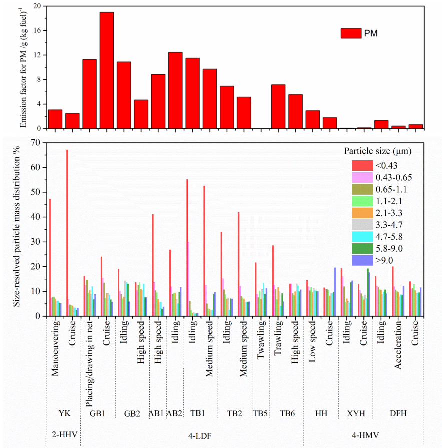

Figure 1 Emission factor for total PM and size-resolved particle mass distributions with different modes for the 12 tested ships

Fig. 2. Figure 1

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		Peak Area		Carbon			
OC1	OC	94	mv-secs	0.17	ug C/cm2	.17	ug C/filter
OC2	OC	764	mv-secs	1.37	ug C/cm2	1.37	ug C/filter
OC3	OC	621	mv-secs	1.11	ug C/cm2	1.11	ug C/filter
OC4	OC	539	mv-secs	0.97	ug C/cm2	.97	ug C/filter
EC1	EC	388	mv-secs	0.70	ug C/cm2	.70	ug C/filter
EC2	EC	16	mv-secs	0.03	ug C/cm2	.03	ug C/filter
EC3	EC	0	mv-secs	0.00	ug C/cm2	.00	ug C/filter
LRPyMin	Py	0	mv-secs	.00	ug C/cm2	.00	ug C/filter
LRPyMid	Py	0	mv-secs	.00	ug C/cm2	.00	ug C/filter
LRPyMax	Py	0	mv-secs	.00	ug C/cm2	.00	ug C/filter
LTPyMin	Py	0	mv-secs	.00	ug C/cm2	.00	ug C/filter
LTPyMid	Py	0	mv-secs	.00	ug C/cm2	.00	ug C/filter
LTPyMax	Py	0	mv-secs	.00	ug C/cm2	.00	ug C/filter

Figure R2 Example of test results for OC and EC fragments

Fig. 3. Figure R2

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