

Referee 1#

Thanks very much for your constructive comments. So far, we have revised the manuscript accordingly. Our point-by-point responses (in black) to each comment are listed below. And the manuscript also has been improved. Please see the manuscript for details.

specific comments

I worry that since this paper is so dense, there are a lot of important details about the sources you tested that haven't been mentioned in the SI. I would like to see a small discussion in your paper about how representative these engines would be relative to the engines that are typically deployed on inland, coastal, and ocean-going vessels in this region. It would also be nice to see more discussion on vessel information and what type of activity each vessel was involved in. Possibly a travel route with a speed or engine load overlay for the in-use testing. One of the bigger components about mobile source testing that is becoming more and more prevalent is activity, which is defined as the type of activity the source is involved in, what the typical engine loads encountered for this activity are, and how that may affect the emissions signal.

Reply: Thanks for your comment. More discussions about the engines equipped in the test ships and their representativeness have been added in Section 2.1 in the revised manuscript as shown below. The typical activities, travel route and operating modes also have been given. We believe the measured ships in this study could represent the typical cargo ships in China to a certain extent.

VOCs samples from 9 different ships were collected in this study, including 2 coastal cargo ships, 3 ocean-going vessels, and 4 inland cargo ships in Yangtze River. The detailed technical parameters of the sampling ships are shown in Table 1. Different types of cargo ships had different technical parameters in China. For example, the engine powers of coastal cargo ships varied largely, with about 57% are equipped with engines of more than 500 kW. Of the other left coastal cargo ships, 17% of which are ranging from 150 kW to 250 kW. Therefore, one large coastal cargo ship with main engine power of 1470 kW and another small coastal cargo ship with main engine power of 178 kW were selected here. Coastal cargo ships typically transport cargos among different coastal ports, with one to several days per voyage. The main operating modes

are cruise (~75% engine load), maneuvering (low and variable engine loads), and idling.

Ocean-going vessels usually have large tonnages with large power main engines. Statistical AIS data show that engines with power of 4 kW to 10 kW account for the largest proportion (~25%) of the total OGVs in China, followed by 2 kW to 4 kW (~23%) and 10 kW to 20 kW (~20%). Besides, newly built OGVs have a tendency to have larger and larger engine powers. Hence, three ocean-going vessels with different engine powers ranging from 13.5 kW to 15.7 kW were tested in this study. They are designed for transporting goods across borders, usually with several months per voyage. The main operating mode is cruise in the open ocean. While during the processes of in and out of the port, the engines of OGVs typically active in maneuvering mode with relative lower and variable engine loads, which could have great influence on the nearshore environment due to higher emission levels of pollutants.

Most inland cargo vessels are generally equipped with high-speed small main engines of power within 1000 kW (~70%). Among them, the vast majority are below 500 kw. Therefore, four typical inland cargo ships of engine power between 138 kW and 300 kW were chosen in this study. The inland cargo vessels typically active among different inland ports or coastal ports near inland rivers, with several hours to several days per voyage. Affected by the complicated water conditions of inland rivers, cruise and maneuvering are the most important operating modes for inland cargo ships.

In brief, the measured ships in this study could represent the typical cargo ships in China to a certain extent. It's worth noting that the ocean-going vessels were newly constructed ships, while the inland cargo ships had older engines (6 to 14 years) compared with other types of ships (less than 10 years).

Besides, most large cargo ships are equipped with both main engine and auxiliary engine. The main engine provides navigation power, and the engine loads vary greatly with the different operating modes. While the auxiliary engine mainly provides domestic electricity or heating on board, and the engine load is relatively stable with about 75% load. Small cargo ships are equipped only with main engines, such as the tested inland cargo ships and small coastal cargo ships in this study.

On that same note, it was never discussed in the SI how the average emissions factors were arrived at. Were the D-2 and E-3 certification test cycles used or was the average performed unweighted?

Reply: Thanks for your comment. The VOCs samples were collected based on actual operating modes (including idling, maneuvering and cruise) except for OGVs that more samples from more operating modes could be obtained thanks to the testing of the newly constructed ships in this study, which are different from D-2 and E-3 certification test cycles. Detailed sampling information is shown in Table S2. Therefore, these average EF_{VOCs} were calculated through unweighted average of different actual operating modes, which has been added in the caption of updated Figure S1.

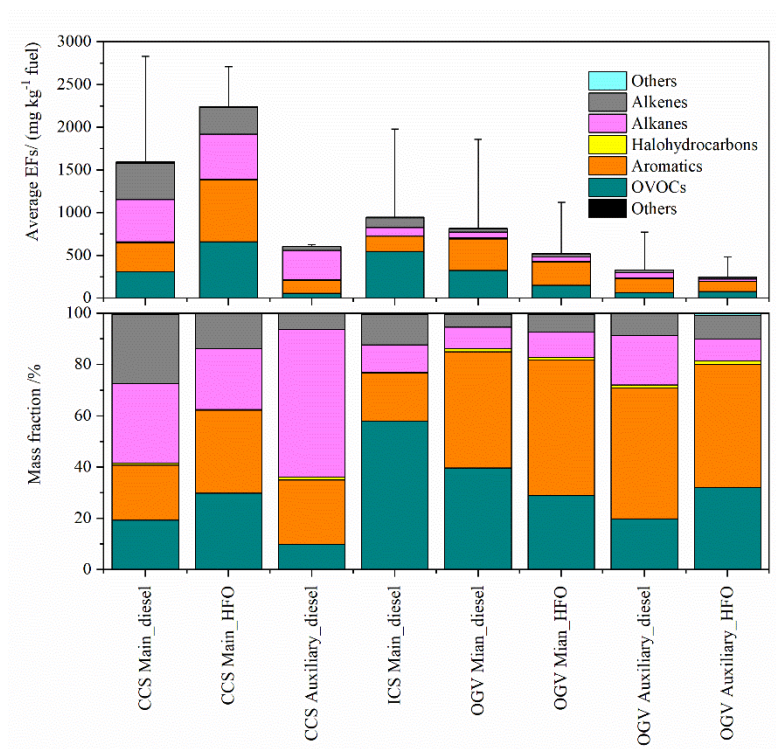


Figure S1 Average EFs of VOCs components and their mass fractions under different ships with different fuels. (These average EF_{VOCs} were calculated through unweighted average of different actual operating modes)

Try to revisit your “low medium and high load” graph in figure 2, do the same low medium and high, except separate OGV, CCS, and ICS. You might see a much tighter resolution on data by load if you incorporate the data points from outside studies that would lead to a better reader understanding of what the engines are doing.

Reply: Thanks for your comment. A new figure has been added in SI as Figure S2 by adding more data from previous studies about ship exhausts. Only few studies have

reported the EF_{VOCs} or EF_{THC} from ship exhausts with different operating modes. Nevertheless, obvious variation trends of VOCs have been shown that EF_{VOCs} had the lowest level when the engines were operating in medium loads, and the highest in low loads, which indicates that engine load could affect the VOCs emission significantly. The revised sentence is also shown in lines 345-347 in the revised manuscript as follows:

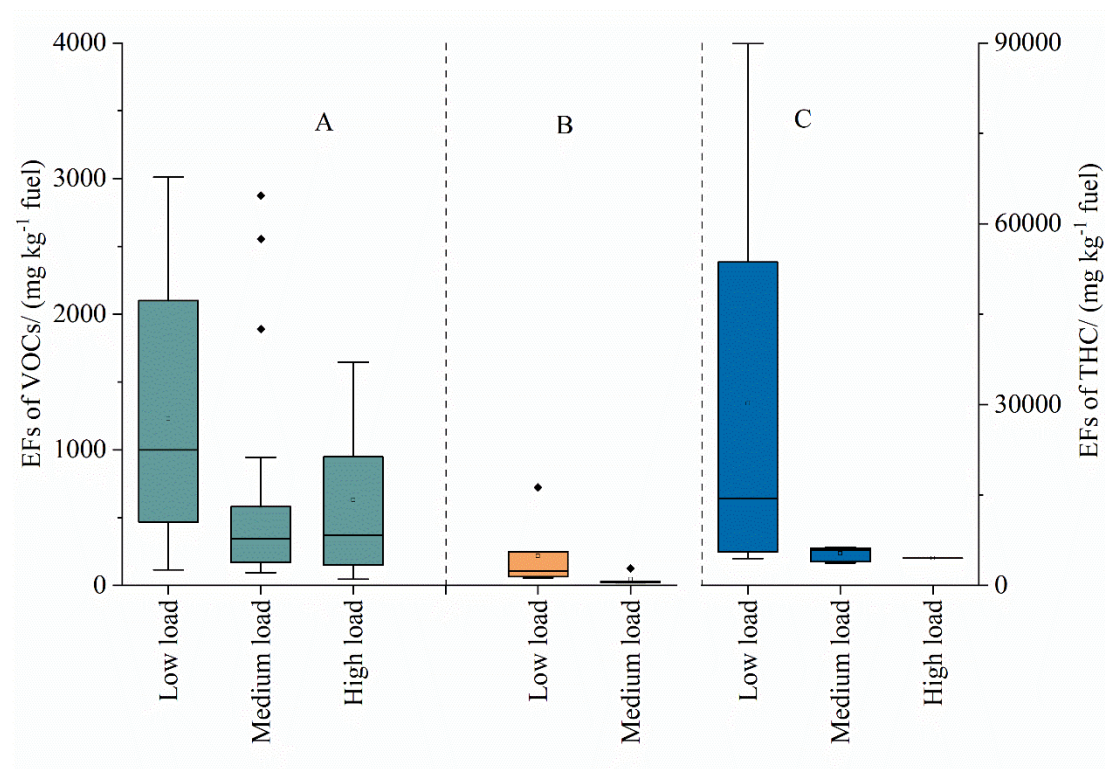


Figure S2 EF_{VOCs} from ship exhausts under different operating modes A, This study; B, (Huang et al. 2018); C, (Radischat et al. 2015), because the THC emission factors were reported in this study with $mg/kW\ h$, the EFs presented in this figure were calculated by assuming that the fuel consumption rate for the test ships was $200\ g\ fuel\ kWh^{-1}$

This was consistent with the results of VOCs emission reported by previous studies such as Huang et al. (2018), Wu et al. (2019) and Radischat et al. (2015), which were also shown in Fig. S2.

There needs to be a more explicit discussion on how you arrived at your modal emissions factors. Incorporate a subsection into your methods for this.

Reply: Thank you very much for your comment. Detailed carbon balance method to calculate the emission factor of VOCs has been added in the revised manuscript as formulas (1) and (2) in Section 2.3.

$$EF_X = \frac{\Delta X}{\Delta CO_2} \cdot \frac{M_X}{M_{CO_2}} \cdot EF_{CO_2} \quad (1)$$

where EF_X is the EF for VOC species X (g/kg fuel), ΔX and ΔCO_2 represent the concentrations of X and CO_2 with the background concentrations subtracted (mol m^{-3}), M_X represents the molecular weight of species X (g mol^{-1}), M_{CO_2} is the molecular weight of CO_2 (44 g mol^{-1}), and EF_{CO_2} is the EF for CO_2 (g (kg fuel)^{-1}).

$$EF_{CO_2} = \frac{C_F}{c(C_{CO}) + c(C_{CO_2}) + c(C_{PM}) + c(C_{HC})} \cdot c^*(CO_2) \cdot M_{CO_2} \quad (2)$$

where C_F represents the mass of carbon in 1 kg diesel fuel ($\text{g C (kg fuel)}^{-1}$), $c(C_{CO})$, $c(C_{CO_2})$, $c(C_{PM})$, and $c(C_{HC})$ represent the mass concentrations of carbon as CO, CO_2 , PM, and HC (g C m^{-3}), respectively, in the flue gas, and $c^*(CO_2)$ is the molar concentration of CO_2 (mol m^{-3}).

Technical corrections

Thank you very much for pointing out these incorrect or inappropriate presentations. All of them have been improved in the revised manuscript.

88 references missing

Reply: The reference has been added in line 98 in the improved manuscript as follows:

The Chinese government also has set the coastal ECAs that require the sulfur content of 0.5% (m/m) since 2019, and 0.1% (m/m) in inland ECAs since 2020 (Ministry of Transport of the People's Republic of China, 2018).

114 healthy

Reply: The word *healthy* has been corrected as *health* in line 125 in the revised manuscript.

115 researches

Reply: *Researches reveal* have been revised as *Research reveals* in line 127 in the revised manuscript.

126 valuated or evaluated

Reply: *Valuated* has been revised as *evaluated* in line 138 in the revised manuscript.

208 controls

Reply: *Control* has been revised as *controls* in line 258 in the revised manuscript.

317-318 confusing wording

Reply: This sentence has been improved in lines 371-373 as follows:

Firstly, as shown in Fig. 2 (b), high-speed and medium-speed engines were equipped for the CCSs, they could lead to higher EF_{VOCs} compared with low-speed engines that equipped for OGVs.

392 needs a semicolon

Reply: A semicolon has been added in in line 450 in the revised manuscript.

397-399 A recent study reported that the addition of additives including naphthalene to low-sulfur fuel during the blended fuel manufacturing process to improve stability could lead to an increase in PAHs, especially naphthalene (Yeh et al., 2023).

You are stating that by adding naphthalene, you increased naphthalene. This is a bit confusing. Are you saying this addition yields increased naphthalene in exhaust? If so, just clarify.

Reply: Thanks for your comment. This sentence has been improved in lines 455-458 as follows:

A recent study reported that the addition of additives of naphthalene-based lubricants to low-sulfur fuel during the blended fuel manufacturing process to improve stability could lead to an increase in PAHs emission in exhaust, with naphthalene being the main pollutant (Yeh et al., 2023).