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 your 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 to14 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.

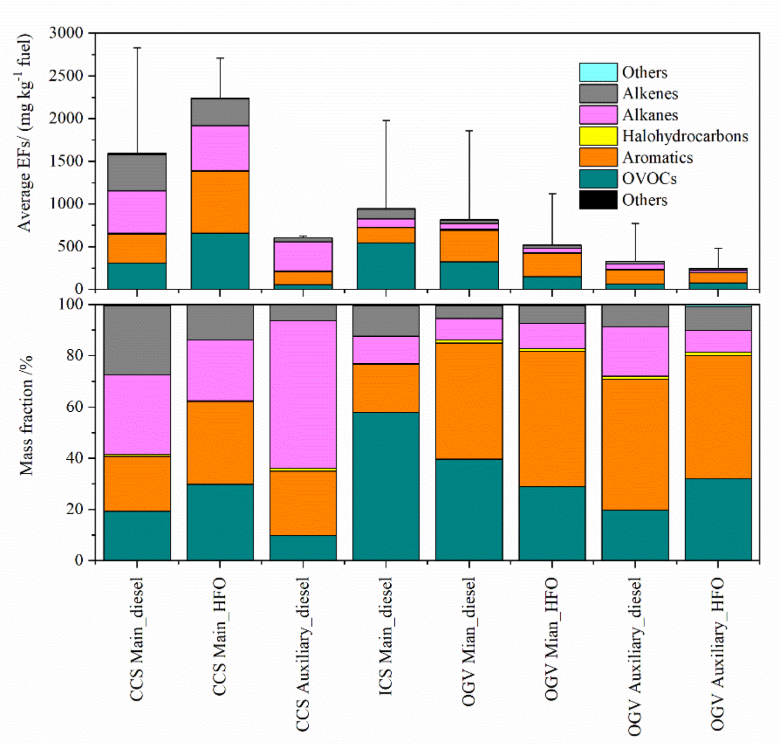

Figure S1 Average EFs of VOCs components and their mass fractions under different ships with different fuels. (These average EF_{VOCs} were calculated through unweighted average of different actual operating modes)

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:

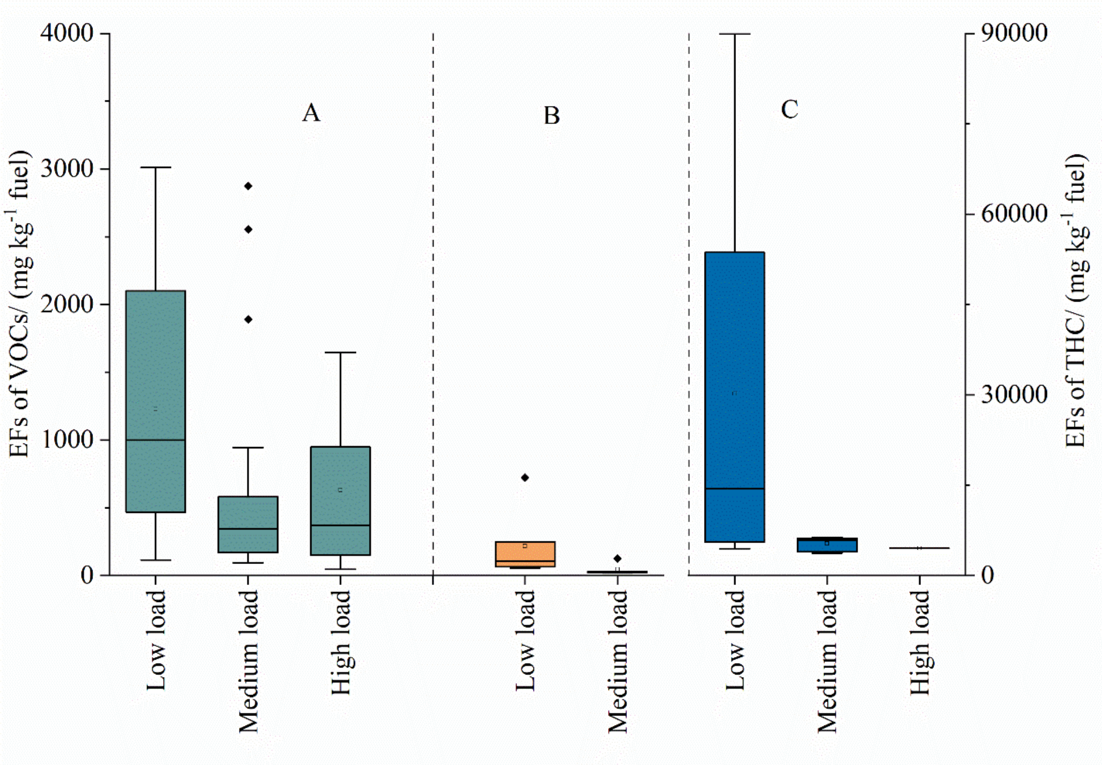

Figure S2 EF_{VOCs} from ship exhausts under different operating modes A, This study; B, (Huang et al. 2018); C, (Radischat et al. 2015), because the THC emission factors were reported in this study with mg/kW h, the EFs presented in this figure were calculated by assuming that the fuel consumption rate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

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

where EF_X is the EF for VOC species X (g/kg fuel), $\triangle X$ and $\triangle CO_2$ 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 (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).

Referee 2#

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.

Major comments

The discussion does not make a connection between the 106 VOCs measured in this study and the ozone and SOA FP. The manuscript needs to answer the questions:

a) Why measure these specific 106 VOCs? Does this subset of VOCs cover the major species observed in previous mass balances?

b) Why assume that these VOCs can explain the ozone and SOA FP? If the SOA FP of 5 unmeasured VOCs was 100x larger than these 106, then the results of the study would not be representative.

c) How do different environmental conditions influence the accuracy of the SOA and ozone formation?

These are questions which have been considered by previous studies, and the authors should be able to address them relatively easily by adding citations and comments in the introduction, methods, and discussion sections.

Reply: Thanks for your comment.

a) There are thousands of VOC species emitted from exhausts. We can't identify and quantitate all the VOCs. However, the measured VOCs in this study can be considered as the main VOC components and basically reflect the emission conditions of ship exhaust. These specific 106 VOCs, including 11 oxygenated volatile organic compounds (OVOCs), 17 aromatics, 29 alkanes, 11 alkenes, 35 halohydrocarbons and 4 other species were tested according to comprehensive consideration of literatures and determination standards of VOCs, which are showing as follows:

Firstly, alkanes, alkenes, aromatics, and OVOCs are mainly concerned from previous studies from ship exhausts, such as 29 alkanes, 19 alkenes, acetylene, 16 single-ring aromatics, and 23 OVOCs from Huang et al. (Huang et al., 2018), 13 OVOCs, 16 aromatics, 11 alkanes and 6 C4-C8 HCs from Agrawal, H., et al. (Agrawal et al., 2010), 29 alkanes, 21 alkenes, 1 alkyne and 17 aromatics from Wu et al. (Wu et al., 2020). These VOCs have been proved and recognized as the main VOC species from ship exhausts. Our measured VOCs contain almost all of these VOC species, which are also easy to be compared and cited for peers. Even though we can't give accurate proportion of the measured VOCs to total VOCs, results from previous studies about motor vehicles show that the identified 57 VOCs (28 alkanes, 15 alkenes, 11 aromatics, and 5 OVOCs) could explain more than 62% of the total nonmethane hydrocarbons (NMHC) (Hung-Lung et al., 2007), and the identified 53 individual VOCs accounted for similar to 80% of the NMHCs from light-duty vehicles, with the most abundant VOCs were ethene (13.8%), acetylene (9.0%), isopentane (7.1%), toluene (5.6%), and n-butane (5.5%)(Araizaga et al., 2013). Therefore, the measured 106 typical VOCs in this study are the major species and have been covered almost all of previous observed VOC species.

Secondly, the detection method used in this study is from USEPA TO-15. This method documents sampling and analytical procedures for the measurement of subsets of 97 VOCs that are included in the 189 hazardous air pollutants (HAPs) listed in Title III of the Clean Air Act Amendments of 1990. These 106 VOCs are measured by Gas Chromatography/Mass Spectrometry (GC/MS) according to USEPA TO-15, which is a very mature and accurate method. Almost half of the 97 VOCs are included in our measurement, especially for halohydrocarbons and aromatics, which can reflect the emission of hazardous VOCs to a certain extent. Besides, VOCs involved in measurement standard for source emission that of most concern in China are also included in this study.

Thirdly, generally speaking, alkanes, alkenes, aromatics and carbonyls with carbon number > 6 in VOCs can form SOA (Grosjean, 1992;Grosjean and Seinfeld, 1989). Previous studies find that aromatics and alkanes contribute most to SOAFP from diesel exhaust, with single-ring aromatics such as toluene, benzene and xylene et al. are the most contributors (Gentner et al., 2012;Che et al., 2023). As for O_3, alkenes, aromatics and OVOCs contribute most to OFP (Che et al., 2023). Wang et al. point out that naphthalene, butene, toluene, benzene, and dodecane etc. are the most contributors to OFP from exhausts of diesel trucks (Wang et al., 2020). Almost all of these VOC species have been identified in our study.

b) To be honest, these measured VOCs in this study couldn’t explain the actual OFP and SOAFP from VOCs. Some potential OFP and SOAFP precursors such as formaldehyde, acetaldehyde and benzaldehyde were missing that could led to underestimate of OFP and SOAFP. However, as mentioned above, the measured VOCs can be considered as the main ozone and SOA precursors and basically reflect the OFP and SOAFP conditions of ship exhaust. These data can also be used for comparison with other studies due to the similar detected VOC species. What's more, based on the typical concerned VOCs, it is intuitive to figure out the impact of the implementation of ship emission control policies on OFP and SOAFP, which is meaningful for further policy formulation.

c) In this study, OFP is estimated using the maximum incremental reactivity (MIR) coefficient method, which represents the maximum contribution of VOC species to the underground O_3 concentration under optimal conditions. (Carter, 1994;Carter, 2010) While SOAFP is calculated using the SOA yields under both high-NO_x and low-NO_x conditions. (Ng et al., 2007) OFP and SOAFP given here are the direct estimated contributions of VOCs from ship exhausts, just like other studies to figure out the effect of fuel switching (Wu et al., 2019;Wu et al., 2020). However, unfortunately, we can't evaluate the accurate contributions of VOCs to SOAFP and OFP in actual atmospheric environment here because they are affected by complicated conditions, such as local temperature, lighting condition, other precursors, atmospheric oxidation, etc., which usually needs to further simulate with air quality models. (Fu et al., 2023)

Relevant contents have been added in lines 219-222 in the revised manuscript.

Minor comments

The abstract summarizes results for CCS and OGVs but not ICS. Please add ICS to the abstract, which will help readers better anticipate the contents of the work.

Reply: Thanks for your comment. ICS has been added in the abstract, showing as follows:

Results showed that emission factor of VOCs (EF_{VOCs}) varied largely from 0.09 to 3.01 g kg^{-1} fuel, with domestic coastal cargo ships (CCSs) had the highest level, followed by inland cargo ships (ICSs) and ocean-going vessels (OGVs).

I find Figure S1 quite valuable and recommend moving the information to the manuscript. However, please find a way to add error bars (or some other measure of variability, like a second Y axis of "standard deviation") to the plot. Please modify the figure caption to explain how the data from different ships were summarized -- it looks like the authors are plotting the mean values? Please comment on the variability between ships? (In contrast, Figure S2 appears to be a summary of Figure S1 and I would leave it in the SI.)

Reply: Thanks for your comment. Figure S1 has been updated with adding the error bars as follows and moved to the manuscript as Figure 2. What needs to be explained to the review is that due to the consideration of costs of the VOCs testing and difficulty of ship exhaust sampling, only 15% of the samples were parallel sampled. Results of the parallel samples showed that the average ratio of Standard Error of Mean (SEM) to the total EF_{VOCs} was 20.8%, which was thought to be acceptable. Then error bars were added in Figure 2 according to both actual standard deviation and this average ratio. These detailed data were also given in Table S5 as sampling errors.

Updated Figure 2 presents the detailed EFs of VOC components for all the test ships under different operating conditions with different fuels. They are not the mean values. While updated Figure S1 are average EFs of VOCs components and their mass fractions for different ships with different fuels. These average EF_{VOCs} were calculated through unweighted average of different actual operating modes.

Brief variations of the total EF_{VOCs} for all the test ships are given in the second paragraph, Section 3.1. Because the EF_{VOCs} is influenced by multiple factors, such as ship type, engine type, operating mode and fuel type, then more detailed discussions about the differences under these factors are presented in Section 3.2.

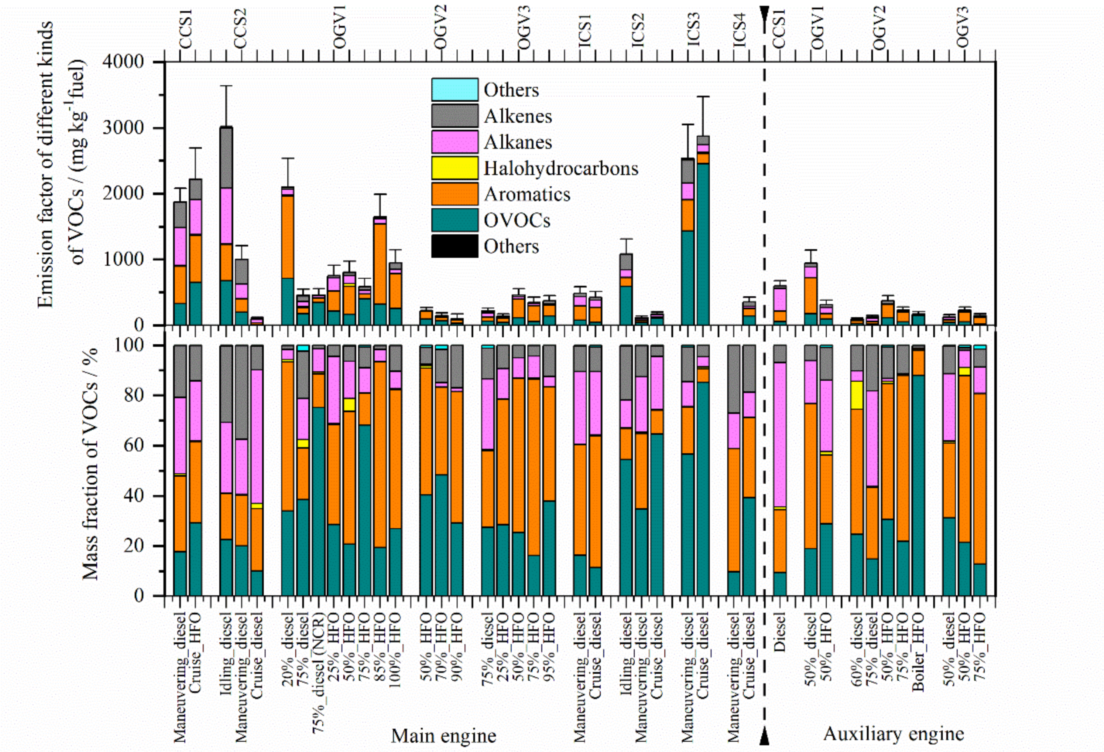

Figure 2 EFs of VOC components and their mass fractions

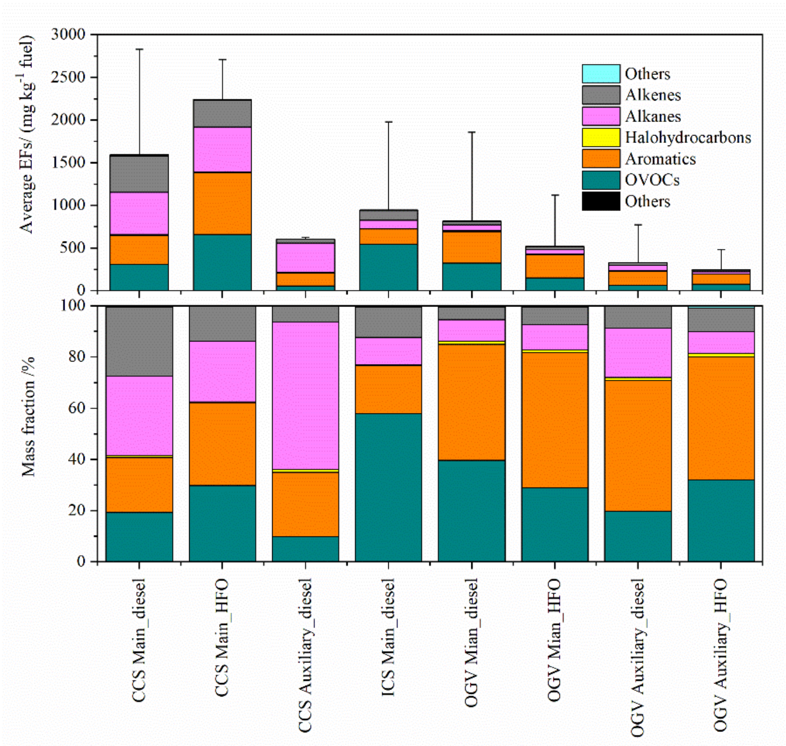

Figure S1 Average EFs of VOCs components and their mass fractions under different ships with different fuels. (These average EF_{VOCs} were calculated through unweighted average of different actual operating modes)

In contrast, Figure 3 is very detailed and not really digestible to the reader. I would move this figure to the SI, and report all data as downloadable data files so that readers requiring this level of detail can use it. I cannot read the x axis of Figure 3. Consider plotting this instead as "mass fraction of Alkanes" with subcategories of the measured alkanes (i.e. subdivide Figure S2) instead.

Reply: Thanks for your comment. Figure 3 has been updated as Figure 4 in the revised manuscript and shown as follows. Because halohydrocarbons, tetrahydrofuran, carbon disulfide, and 1,4-dioxaneand only account for very small mass fractions of the total test VOCs (0.55%-3.06% of total VOCs), they are removed from Figure 4 to enhance the readability for reader. While detailed mass fractions of all the test VOC species in this study also have been added in Table S7 as the reviewer suggested. However, profile of VOCs is a very important characteristic of ship exhausts, which can show the differences of VOC mass fractions intuitively among different ships with different fuels, and therefore we still want to keep this figure in revised manuscript.

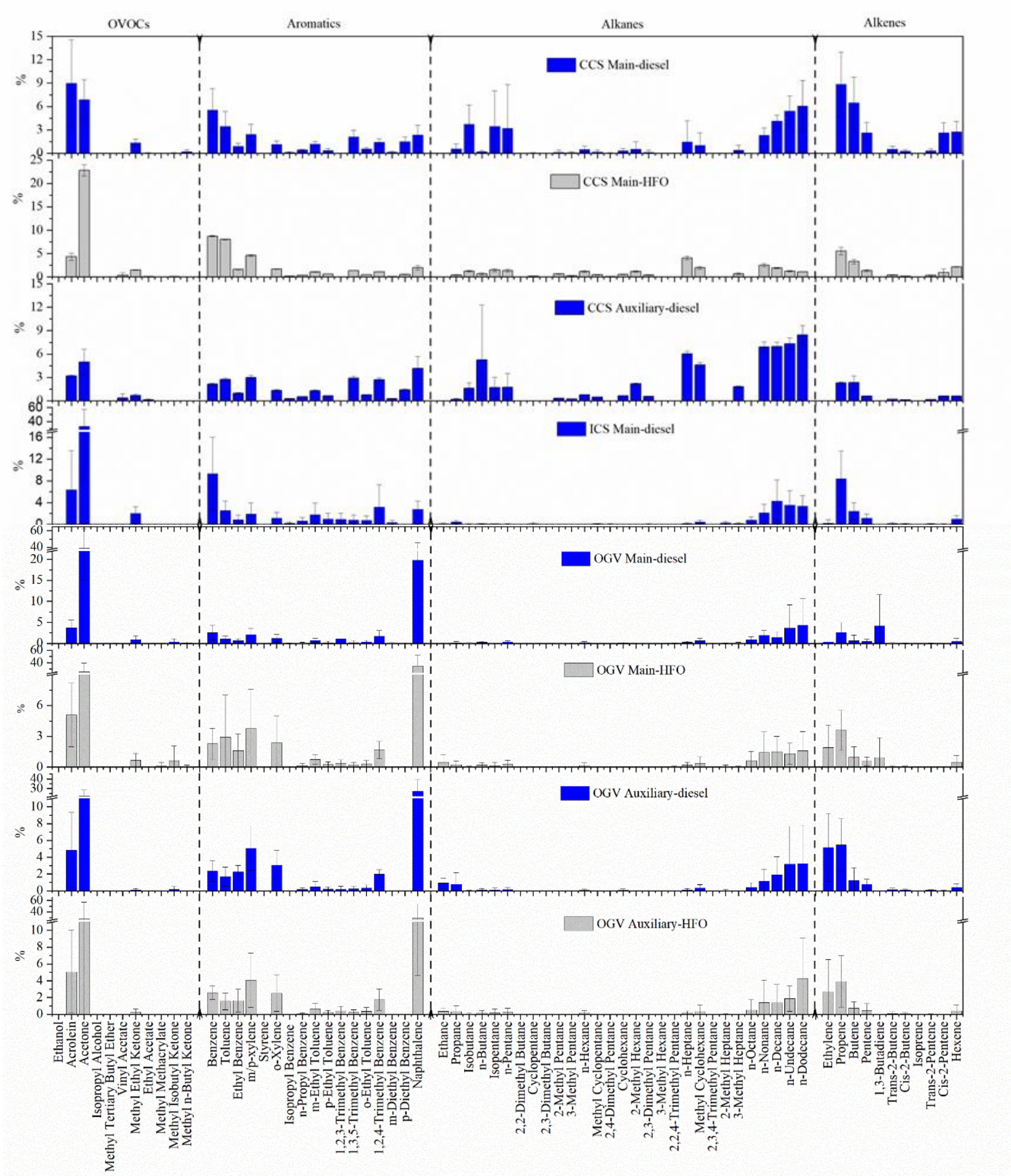

Figure 4 Mass fractions of individual VOCs from test ships under different engine types and fuels (except halohydrocarbons, tetrahydrofuran, carbon disulfide, and 1,4-dioxaneand due to their very small mass fractions)

As noted above, the introduction should mention and cite studies which explain the connection between VOC chemistry and SOA formation potential.

Reply: Thanks for your comment. Relevant contents have been added in lines 70-81 in the revised manuscript as follows:

Generally speaking, alkanes, alkenes, aromatics and carbonyls with carbon number > 6 in VOCs can form SOA (Grosjean, 1992;Grosjean and Seinfeld, 1989).

While O_3 is formed from the photochemical interactions of volatile organic VOCs and oxides of nitrogen (NO_x), with alkenes having the highest Maximum Incremental Reactivity (MIR), followed by aromatics and OVOCs (Carter, 1994). Typical aromatics, alkenes, and alkanes are the most concerned VOCs from diesel exhausts. For example, Previous studies find that aromatics and alkanes contribute most to SOAFP from diesel exhaust, with single-ring aromatics such as toluene, benzene and xylene et al. are the most contributors (Gentner et al., 2012;Che et al., 2023). Wang et al. (2020) point out that naphthalene, butene, toluene, benzene, and dodecane et al. are the most contributors to OFP from exhausts of diesel trucks.

The introduction should introduce the concepts of IVOCs and OVOCs, which appeared in line 169 without definition. Especially since these definitions are different from the common IVOC, SVOC, LVOC categories.

Reply: Thanks for your comment. IVOCs is intermediate volatile organic compounds, while OVOCs is oxygenated volatile organic compounds. Since IVOCs and OVOCs reported by Liu et al. (2022) are not the focus of this study. The ambiguous sentence as following has been deleted in the revised version.

I/OVOCs samples were obtained by automatic sampler to get IVOCs and OVOCs samples that had been reported in other study (Liu et al., 2022).

Line 142, for the reader's benefit, please add the range of years meant by "older", based on Table 1, after making this subjective comment.

Reply: Thanks for your comment. The ranges of years have been added in lines 174-176 in the revised manuscript as following:

It’s worth noting that the ocean-going vessels were newly constructed ships, while the inland cargo ships had older engines (6 to14 years) compared with other types of ships (less than 10 years).

Table 1, which engines were sampled? Note this in the caption, please.

Reply: Thanks for your comment. VOCs from all the engines listed in Table 1 had been collected in this study. The introduction has been added in lines 206-207 in the revised manuscript.

VOCs samples were collected by summa canister from both main engines and auxiliary engines of all the ships listed in Table 1.

Line 323, "emission of EF" change to "the EF" (EF is "emission" already)

Reply: Thanks for pointing it out. The sentence has been revised in the improved manuscript in in lines 377-379.

As mentioned before, fuel type could influence the EF_{VOCs} significantly (Wu et al., 2019;Wu et al., 2020), which also would be one of the most important influence factors in the future under the background of increasingly strict ship oil policy.

Figure 4 caption should point to the section where the source ratios are cited, please.

Figure 4: I do not disagree with the authors that the B:T:E ratios could be used as tracers for ship emissions. But please add a box to highlight the region you are suggesting. Specify the recommended region explicitly in the text for clarity.

Reply: Thanks for your comment. Figure 4 and the caption have been revised and improved as follows.

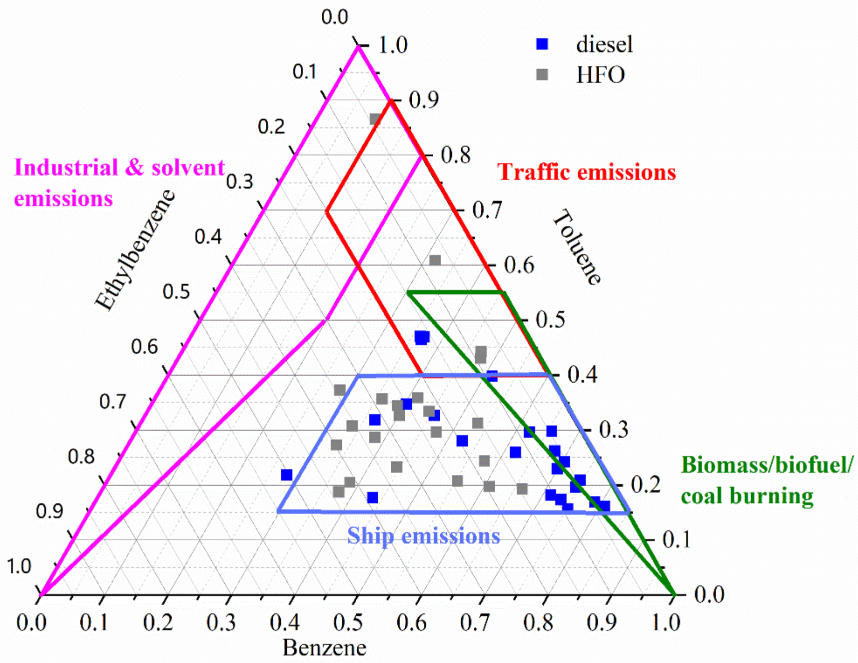

Figure 4 Relative proportions of benzene, toluene and ethylbenzene from the ship exhausts. B:T:E ratios from other sources were cited from Zhang et al. (2016b) that summarized 28 examples from biomass burning, 35 examples from biofuel burning, 17 examples from coal burning, 11 examples from diesel vehicle exhaust, 31 examples from gasoline vehicle exhaust, 24 examples from gasoline evaporation, 25 examples

from roadside or tunnel tests, and 66 examples from industrial processes and solvent applications.

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