

We greatly appreciate all the reviews' constructive feedback on our manuscript. In response to the insightful comments, we have thoroughly revised the manuscript. Below, we provide a detailed point-by-point reply to each of the reviews' comments. We believe these revisions have significantly improved the clarity and quality of our work, and we have updated the manuscript accordingly. For further details, please refer to the revised version.

Referee 1#

Specific Comments:

The manuscript presents novel findings concerning ship emissions, with a focus on intermediate and semi-volatile organic compounds. The results reveal the enhancement of organic compounds following the transition from high-sulfur to low-sulfur fuels. The manuscript is well written and provides meaningful information to our knowledge. However, there are several questions and comments that require attention: (1) Lines 63-66: This sentence is ambiguous in terms of the contribution proportion of SO₂. It is suggested to be revised.

Reply: The sentence has been verified against the original reference and revised in lines 64-67 in the improved manuscript as follows:

Specifically, the consumption of heavy fuel oil constitutes 70% of total ship fuel usage (International Maritime Organization, 2016), which is primarily responsible for the majority of SO₂ emissions associated with global shipping. This has considerable effects on coastal regions and the marine environment.

(2) Line 87: Does PM refer to mass or number? It needs to be made clear.

Reply: Thank you for your comment. We have explicitly stated that 'PM' refers to mass in line 89 in the revised version of the manuscript, as shown below:

Previous studies have indicated that the low-sulfur fuel regulation is an effective measure for reducing SO₂ and PM mass emission in many countries.

(3) Line 98: The aging of VOCs to form intermediates is equally important for SOA. Examination of long-time aging process on volatile organic compounds emitted from solid fuel combustion in a rural area of China. Chemosphere 333 (2023) 138957.

Reply: Thanks for your reminding. The aging process of VOCs is not only closely

related to SOA formation, but also plays a significant role in ozone formation and its impact on climate and air quality. Additionally, the intermediates formed during VOCs aging are key precursors for SOA formation. This content has been added to the lines 101 -104 of the manuscript:

VOCs have been getting lots of interests due to their crucial role as common precursors of secondary organic aerosols (SOAs) and ozone (O₃) (Shen et al., 2023;Hui et al., 2019). Additionally, the aging process of VOCs and the resulting intermediates also play pivotal role in SOA formation (He et al., 2023; Srivastava et al., 2022).

(4) Line 108: Provide the full name of “WRF-Chem”

Reply: Thank you for the suggestion. We have now provided the full name of "WRF-Chem" (Weather Research and Forecasting model coupled with Chemistry) in lines 112-113 in the revised manuscript.

(5) Line 118 and 102: The emission of IVOCs from various sources are important for the SOA production. It is recommended to enrich the expression, and the following study can be referred to: Emission of Intermediate Volatile Organic Compounds from Animal Dung and Coal Combustion and Its Contribution to Secondary Organic Aerosol Formation in Qinghai-Tibet Plateau, China. Environmental Science & Technology 2024 58 (25), 11118-11127.

Reply: Thank you for your suggestion. We have enhanced the expression to provide a more comprehensive explanation in lines 117-120 in the revised manuscript.

Clarifying the contribution of I/SVOCs from different sources to SOA formation is crucial for a comprehensive understanding of their role in atmospheric processes and their impact on air quality (He et al., 2024; Srivastava et al., 2022).

(6) Section 2: In this study, some data statistical analyses were involved. It is necessary to add the methods used in the statistical analyses mentioned in the text, such as significance tests, etc.

Reply: We appreciate the reviewer’s insightful comment. The data statistical analyses involved in this study, such as significance test, have been explained in the relevant caption of Figure 2 in lines 430-434 in the revised manuscript.

Figure 1 Box-whisker plots of total EF_{I/SVOCs} for the tested ships under (a) different fuel types, (b) different operating modes, and (c) different engine types. N represents the number of samples. Significant differences between samples were determined using an independent samples T-test. The error bars represent the standard deviation of the measured values, while *** indicates a significance level of $p < 0.001$.

(7) Lines 151-152: The parameters of fuels are the most important information in this study. It is suggested that the Table S2 be placed in the main text.

Reply: Thank you for the suggestion. We agree that the fuel parameters are crucial to this study and have moved Table S2 into the manuscript as Table 1 for better accessibility. See details in line 164 in the revised manuscript.

(8) Line 167: A total of 64 sets of gas-phase and particle-phase I/SVOCs samples were collected in this study, involving various engine types, fuels, and operating modes. Please add a table detailing exactly in what fuel, what type of engine, and under what operating modes, etc. these samples were collected.

Reply: Thank you for your valuable advice. The details of specific fuels, engine types, operating modes, and other relevant conditions under which these samples were collected have been added as Table S3 of the supplementary materials.

Table S3 Engine type, operating mode, and fuel type of each ship for each

measurement							
Ship	Engine	Operating	Sampling	Ship	Engine	Operating mode	Sampling
ID	type	mode	duration	ID	type		duration
OGV1	Main	20%_MGO ^a	20 min	OGV3	Auxiliary	50%_MGO	20 min
	engine	75%_MGO	20 min		engine	50%_HFO	20 min
	Auxiliary	75%_MGO	27 min			75%_HFO	20 min
	engine	(NCR)					
OGV2	Main	25%_HFO	20 min	ICS1	Main	Maneuvering_0#diesel	20 min
	engine	50%_HFO	20 min		engine	Cruise_0#diesel	20 min
		75%_HFO	20 min	ICS2	Main	Maneuvering_0#diesel	20 min
	Auxiliary	85%_HFO	20 min		engine	Cruise_0#diesel	20 min

	engine	100%_HFO	20 min	ICS3	Main	Maneuvering_0#diesel	20 min
		50%_MGO	25 min		engine	Cruise_0#diesel	20 min
		50%_HFO	70 min	ICS4	Main	Maneuvering_0#diesel	20 min
	OGV3	75%_MGO	40 min		engine	Cruise_0#diesel	20 min
		25%_HFO	20 min				
		50%_HFO	10 min				
		75%_HFO	40 min				
		95%_HFO	40 min				

^a, means percentage of engine load under what type of fuel

(9) Line 202: In formula (1), when calculating EF, has the influence of organic carbon in gaseous phase been considered? How large is the error?

Reply: Thank you for your valuable comment. Unfortunately, the concentration of hydrocarbons (HC) was not quantified in this study and thus was excluded from the calculation of the emission factor. We recognize that incorporating organic carbon in the gaseous phase may introduce a degree of uncertainty into our analysis. It is important to note that in our previous research (Zhang et al., 2018), HC was included in the emission factor calculations. However, based on earlier data, the contribution of gaseous carbon (C) was relatively insignificant. Additionally, we have quantified the contribution of carbon from VOCs to the total emitted carbon in this study (Zhang et al., 2024a). The average emission factor of carbon from VOCs is $0.025\% \pm 0.020\%$ of that of CO_2 , indicating a relatively minor contribution. Considering the limited availability of HC data in this study and its minimal impact, it is reasonable to infer that the uncertainty associated with carbon in gaseous organic matter has a negligible effect on the overall results.

(10) Lines 312-319: Here reveals a very interesting research result that high-quality fuel leads to higher organic matter emissions. What do you think are the reasons for the increase in SVOCs/IVOCs emission factors after the switch from high-sulfur oil to low-sulfur fuel?

Reply: Thank you for your question. The increase in I/SVOCs emission factors after switching from high-sulfur to low-sulfur fuels is a multifaceted phenomenon primarily attributable to alterations in combustion efficiency and changes in the chemical composition of the fuel. A detailed analysis of these factors is provided in the latter part of the second paragraph of Section 3.2 (lines 369-376). Specifically, low-sulfur fuels tend to exhibit reduced combustion efficiency, which can lead to increased formation of I/SVOCs. Moreover, the production process of lower sulfur content fuels can also alter the relative proportions of other fuel components (Liu et al., 2022; Zhang et al., 2024), potentially leading to a higher propensity for the formation of I/SVOCs under certain combustion conditions, which may account for the observed increase in emission factors.

(11) Section 3.2: How do the influencing factors such as fuel type, engine type and operating conditions interact to affect the final combustion process and I/SVOCs emissions? Give more explanations.

Reply: Thank you for your insightful question. The interaction between fuel type, engine type, and operating conditions plays significant roles in influencing the combustion process and the emissions of I/SVOCs. These factors affect both the combustion efficiency and the chemical composition of the emissions. Here, we provide a more detailed explanation of how each factor interacts to influence the final emissions:

The chemical composition of the fuel directly affects the types and amounts of I/SVOCs generated during combustion. Fuels with higher aromatic content tend to produce more semi-volatile and particulate-bound organic compounds (Zhao et al., 2015). In contrast, fuels with higher aliphatic content generally produce fewer semi-volatile species (Huang et al., 2018b). The presence of oxygenates in some fuels can alter the combustion temperature and the oxidation pathways, leading to different emission profiles for I/SVOCs. Moreover, the fuel type also influences its vaporization and mixing behavior with air during combustion, which subsequently affects the combustion efficiency (Liu et al., 2022; Zhang et al., 2024). The design and operational characteristics of the engine play a critical role in the combustion process. For example, in internal combustion engines, the air-fuel mixture, combustion chamber design, and fuel injection timing can significantly influence the temperature and pressure during combustion (Sayin and Canakci, 2009). These factors affect the thermal decomposition of hydrocarbons and the subsequent formation of I/SVOCs. Moreover, engine speed and load can influence combustion efficiency, with poor combustion typically leading to higher emissions of I/SVOCs. The operating conditions of the engine, including load, speed, and temperature, also influence the emission characteristics (Rajendran et al., 2023). Under low-load or idle conditions, incomplete combustion often results in higher emissions of I/SVOCs, as fuel is not fully vaporized or oxidized (Liu et al., 2022). High temperatures generally promote more complete combustion, but may also lead to the formation of more highly reactive intermediate species, which can contribute to I/SVOC formation in certain scenarios. The air-to-fuel ratio is another important

factor—leaner mixtures (excess air) can result in lower I/SVOC emissions, while richer mixtures (more fuel) may increase the formation of unburned hydrocarbons and intermediate compounds. In summary, fuel type determines the primary chemical composition of the emissions, engine type influences the combustion efficiency and reaction pathways, and operating conditions dictate the degree of combustion completeness and the formation of intermediate compounds. These factors interact in such a way that they lead to varying degrees of incomplete combustion, resulting in the formation of a complex mixture of I/SVOCs.

(12) Lines 353-357: Incomplete combustion is regarded as an important reason for the increase of I/SVOCs with low-sulfur fuel. Can more convincing evidence, such as combustion efficiencies of the engines with different types of fuels, be provided in this study to support the conclusion given here?

Reply: Thank you for highlighting this important aspect. As you suggested, we have calculated the modified combustion efficiencies (MCE) with different fuel types (as shown below). It can be observed that the MCE values decrease significantly as the sulfur content of the fuel decreases. By comparing MCE values across different fuels, we can more robustly establish the relationship between incomplete combustion and the observed increase in I/SVOC emissions. These calculations offer compelling evidence to further support and reinforce the conclusions drawn in this study. This supplementary analysis has been integrated into the manuscript, with detailed information provided in lines 371-376 of the revised version:

To further elucidate the impact of reduced sulfur content on combustion efficiency, we calculated the modified combustion efficiencies (MCE) for various fuel types (Figure S2). The results indicated a notable decline in MCE values as the sulfur content decreased, suggesting that incomplete combustion was a key factor contributing to the elevated levels of I/SVOCs in low-sulfur fuels.

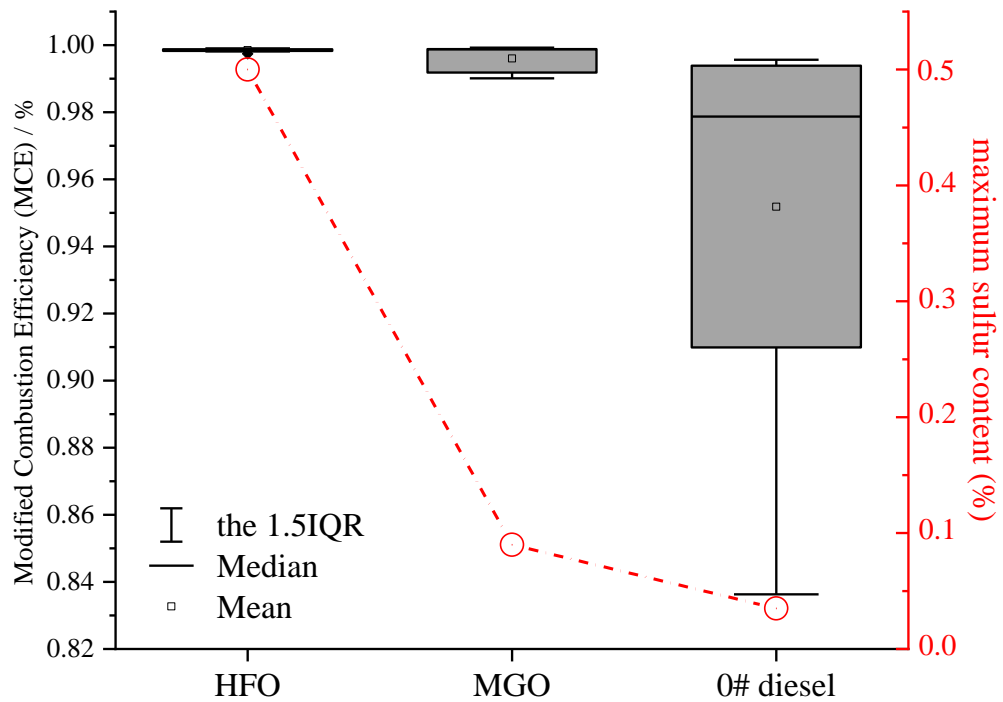


Figure S2 Box-whisker plot of the MCE with different fuel types

(13) Figure 2: Explain the meanings of the different symbols in this box plot, such as which one represents the average value, the median, etc. Besides, what method was used to calculate the P value?

Reply: Suggestion taken. The meanings of the different symbols in the box plot have been added in Figure 2. Significant differences between samples were determined using an independent samples T-test, which is shown in the caption of Figure 2.

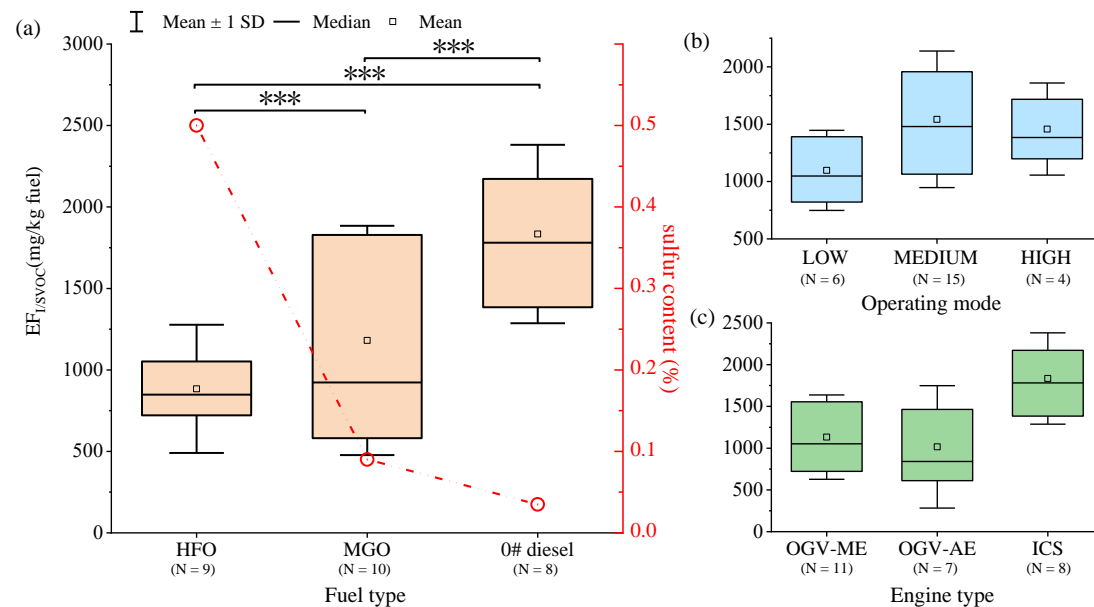


Figure 2 Box-whisker plots of total $EF_{I/SVOCs}$ for the tested ships under (a) different fuel types, (b) different operating modes, and (c) different engine types. N represents the number of samples. Significant differences between samples were determined using an independent samples T-test. The error bars represent the standard deviation of the measured values, while *** indicates a significance level of $p < 0.001$.

(14) Lines 410-412: Given that UCM accounts for a very high proportion of total I/SVOCs, what do you think the future research on UCM should be carried out, and is it necessary to further identify more compounds and what analytical methods should be used?

Reply: Thank you for your insightful question. Given that UCM (Unresolved Complex Mixture) accounts for a significant portion of total I/SVOCs, future research should prioritize refining analytical techniques to better characterize and identify the compounds within UCM. Since UCM is composed of a complex array of compounds that can often overlap in conventional single-dimensional Gas Chromatography (GC), employing Two-Dimensional Gas Chromatography (GC×GC) offers a powerful approach for separating and identifying more components (Tang et al., 2023). GC×GC enhances separation by utilizing two orthogonal columns with different stationary phases, thus allowing for improved resolution of complex mixtures like UCM (Marriott, 2004). Moreover, Orbitrap mass spectrometry provides ultra-high resolution and high mass accuracy, which can significantly improve the identification of individual compounds in the UCM (Kösling et al., 2022). Its capability to differentiate isomeric compounds and identify low-abundance species with high sensitivity is essential for the characterization of complex mixtures. Although GC × GC and Orbitrap MS are individually powerful analytical techniques, their integration with complementary methods such as high-resolution liquid chromatography (HRLC) or high-resolution mass spectrometry (HRMS) would significantly enhance the characterization of UCM. These combined approaches could enhance the detection of a wider range of compounds, including semi-volatile and non-volatile species, which may be more challenging to analyze with GC×GC alone. In conclusion, the identification of more

compounds in the UCM and the application of advanced analytical techniques, such as GC×GC and Orbitrap MS, are crucial for advancing our understanding of UCM's role in I/SVOC emissions. These approaches offer superior separation, sensitivity, and resolution, making them invaluable tools for future research on UCM characterization.

Detailed information also has been provided in lines 707-710 in the revised manuscript:

Future research can obtain more comprehensive and accurate organic component emission data by using advanced methods such as comprehensive two-dimensional gas chromatography-mass spectrometry (GC × GC-MS) and Orbitrap mass spectrometry.

(15) Section 3.4: It is reasonable to divide the volatile range with normal alkanes as indicators. However, when discussing the differences in volatile distribution under different fuel types, only the phenomenon was described. How did the differences in fuel composition eventually lead to changes in volatile distribution? Can a summary of this part be made based on the component differences of different fuels?

Reply: Thank you for your insightful comment. The variations in fuel composition indeed play a crucial role in shaping the volatile emissions during combustion. Fuels with different chemical compositions, such as varying carbon chain lengths, aromatic content, and sulfur levels, lead to different combustion behaviors. These differences influence the volatility of the organic compounds released during combustion, which subsequently alters the distribution of I/SVOCs (Liu et al., 2022). Additionally, the sulfur content can also affect combustion efficiency, which may further alter the volatile distribution by influencing incomplete combustion processes.

In summary, the changes in volatile distribution can be attributed to the complex interactions between fuel composition, combustion conditions, and the inherent chemical properties of different fuel types. We have now added a more detailed explanation of these interactions and their impact on volatile emissions in lines 620-625 in the revised manuscript:

The compositions and physicochemical properties of different fuel types vary, leading to differences in the volatile organic compounds they contain. Consequently, the type of fuel played a significant role in determining the distribution of volatile

fractions for each individual I/SVOC component. The composition and combustion efficiency of fuel are important factors affecting the emission and distribution of I/SVOCs.

(16) Figure 4: In the figure involving Hopanes, some error bars are covered.

Reply: Thank you for highlighting this issue. We have adjusted the figure to ensure that the error bars are fully visible and not obscured in the revised version of the manuscript. See details below:

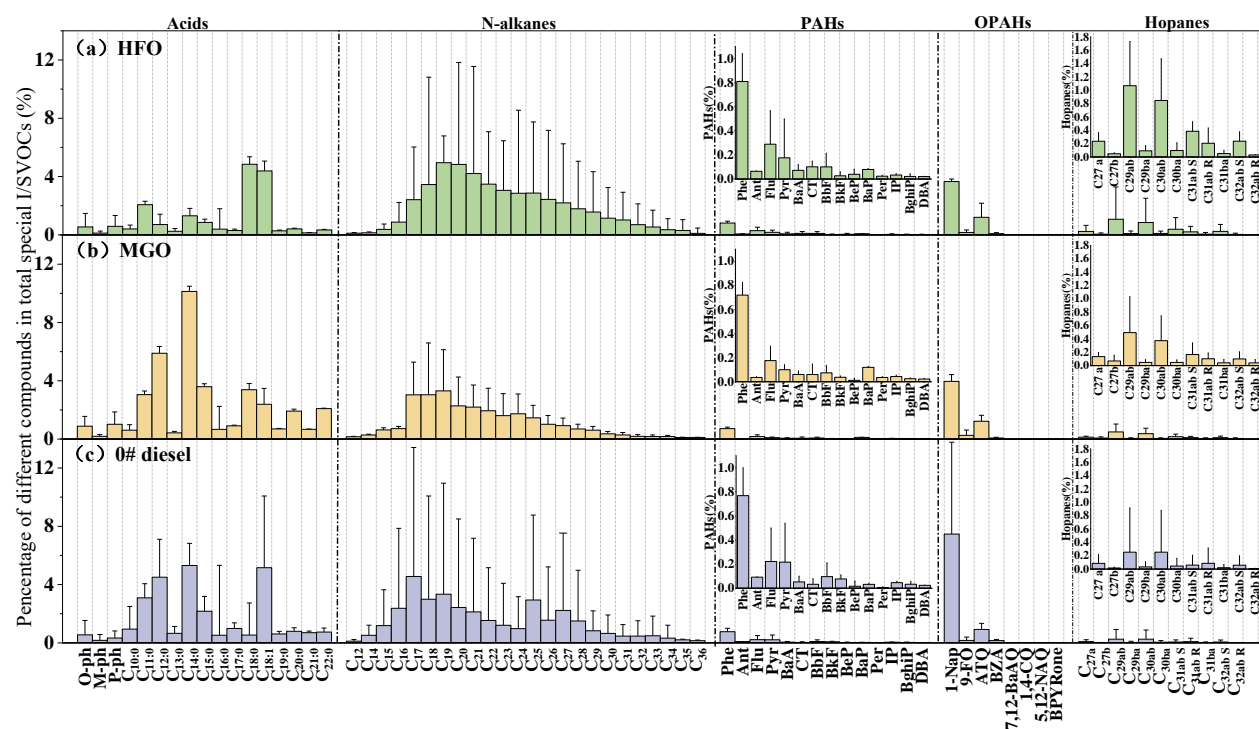


Figure 4 Profiles of I/SVOCs in ship exhausts under different fuels

(17) Figure 5 and Figure 6: The meanings of the Y-axis titles have not been clearly explained.

Reply: Suggestion taken. We have revised the explanations for the Y-axis titles in Figures 5 and Figure 6 to ensure greater clarity. These clarifications are now included in the revised manuscript. See details below:

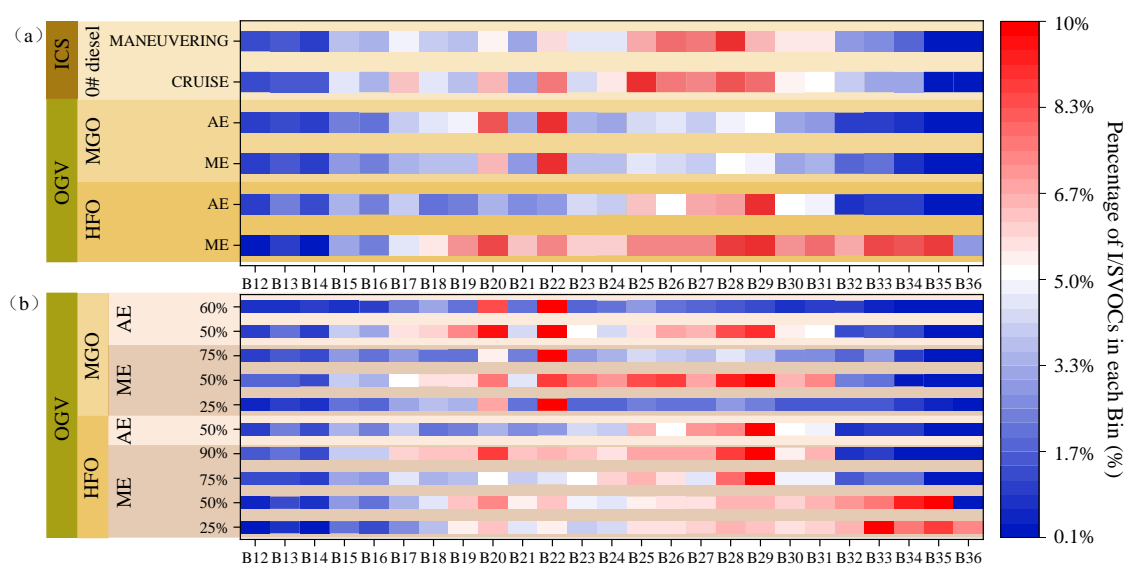


Figure 5 Split-bar heat plot of the I/SVOCs proportions in each volatile bin under (a) different fuel types for different engines, and (b) different operating conditions (25% - 90% operating modes) for OGVs

(18) Figure 6: In the legend, the word "Hopanes" is misspelled.

Reply: Thank you for pointing that out. The misspelling of "Hopanes" in the legend has been corrected in the revised version of Figure 6. Moreover, Figure 6 also has been revised as the volatility distributions of I/SVOCs based on the volatility basis set (VBS) framework from different fuels for improved clarity.

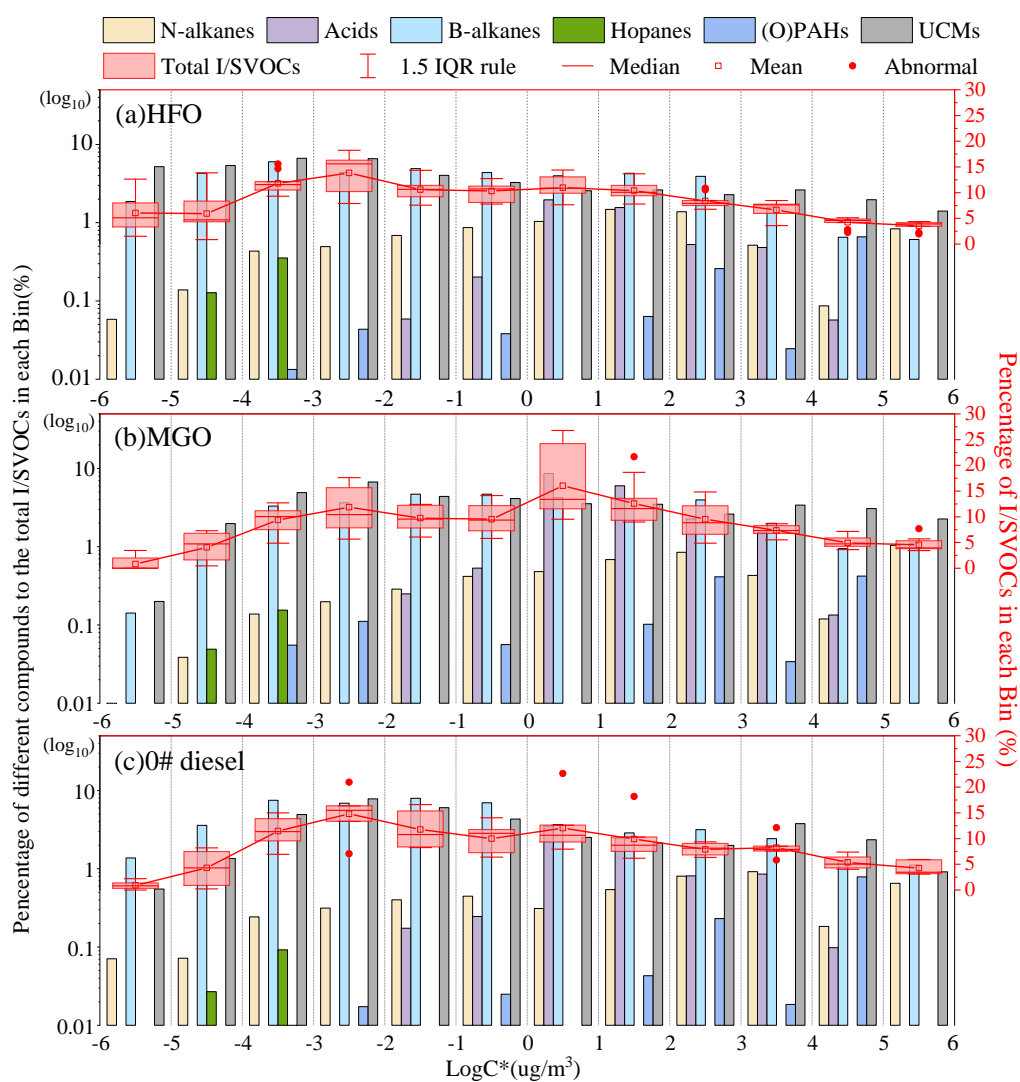


Figure 3 The volatility distributions of I/SVOCs based on the volatility basis set (VBS) framework from different fuels

(19) Figure 7: Could the total error bar of SOAFP be given?

Reply: Suggestion taken. The error bars for SOAFP have been added in the revised Figure 7.

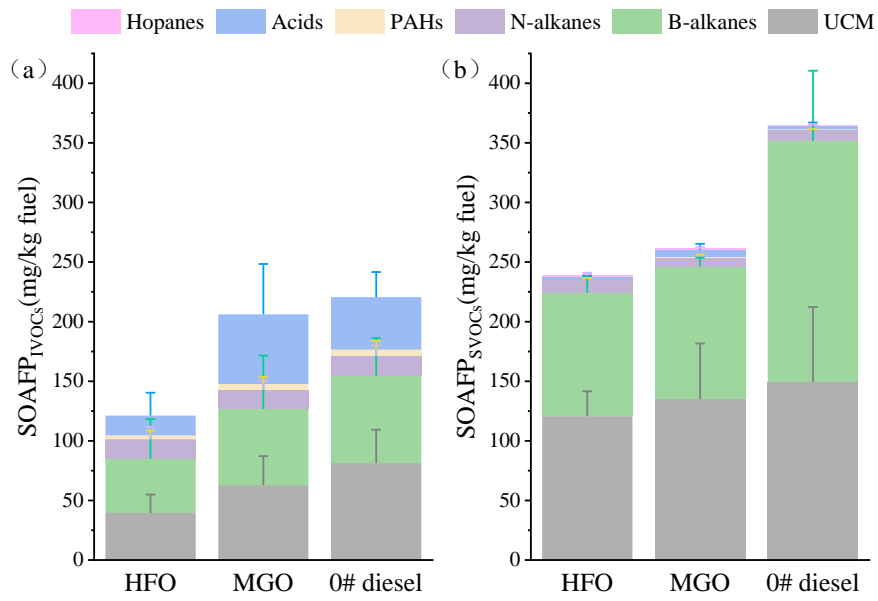


Figure 7. SOAFP from (a) IVOCs and (b) SVOCs

Referee 2#

In this study, the authors estimated the emission factors of speciated and unspecified I/SVOCs from ocean-going vessels and inland cargo ships with different fuel types, engine types and operating conditions. They found that the emission factors of I/SVOCs increased as the sulfur content in fuels decreased. Although the methods, analysis, and results are not particularly novel which was built on previous studies of IVOCs emissions from vehicles (Zhao et al., 2015, 2016), the measurement data presented in this study are very helpful for estimating SOA formation from ship exhausts. I recommend the publication of this work after the authors could address my comments below.

Major comments:

(1) My major concern lies in the representation of volatility distributions. Though I totally understand that the authors classified volatility based on carbon numbers (Lines 188-194), following the methods in Zhao et al. (2015, 2016), I strongly recommend the authors can plot volatility distributions using the volatility basis set (VBS) framework (Donahue et al., 2006), similar to Figure 2 in Zhao et al. (2015) and Figure 4 in Zhao et al. (2016). The VBS framework is widely used in air quality models for simulating SOA formation. Representing volatility distribution within this framework would provide essential information for inputting the measured data into chemical transport models in future studies.

Reply: We sincerely appreciate your constructive comments regarding the representation of volatility distributions in our manuscript. Based on the recommendations, we utilized the effective saturation concentration (C^*) of n-alkanes in each bin as a proxy for the volatility of intermediate and semi-volatile organic compounds (I/SVOCs) to derive their volatility distribution. The improved Figure 6, generated using the VBS framework, is presented below. The associated text has also been updated in lines 601-625 in the revised manuscript. The pertinent revised content is outlined as follows:

In order to figure out the volatility distributions of detailed chemical compositions of I/SVOCs from different fuels. The average volatility distributions of UCM, b-alkanes,

n-alkanes, PAHs, acids and hopanes based on the volatility basis set (VBS) framework are given in Figure 6. Results revealed that the volatility distributions of UCM exhibited distinct bimodal characteristics, with peak values occurring at $\log C^* = -3$ to -2 and $\log C^* = 3$ to 4 . Notably, the concentration was more pronounced in the low-volatility region. Moreover, the bimodal characteristic of UCM in 0# diesel was more pronounced compared to HFO and MGO. Additionally, HFO exhibited a higher proportion of low-volatility UCM relative to 0# diesel and MGO. The volatile distributions of n-alkanes exhibited a bimodal structure, with peaks occurring at $\log C^* = -4$ to -2 and $\log C^* = 5$ to 6 , respectively. Furthermore, from HFO to MGO and finally to 0# diesel, the VBS peak of n-alkanes progressively shifted towards the higher volatility range. This shift could be attributed to the distinct characteristics of these fuels, where MGO and 0# diesel had lower boiling points and contained fewer carbon atoms in their hydrocarbon chains compared to HFO (Liu et al., 2022). The volatility distributions of other specific I/SVOCs were consistent with their molecular sizes. (O)PAHs emitted from ships were predominantly small molecules with high volatility, primarily enriched in the $\log C^*$ range of 4 to 5 . Acids were mainly concentrated in the higher volatility bins ($\log C^* = 0$ to 5), whereas hopanes exhibited a primary concentration in the lower volatility intervals. The compositions and physicochemical properties of different fuel types vary, leading to differences in the volatile organic compounds they contain. Consequently, the type of fuel played a significant role in determining the distribution of volatile fractions for each individual I/SVOC component. The composition and combustion efficiency of fuel are important factors affecting the emission and distribution of I/SVOCs.

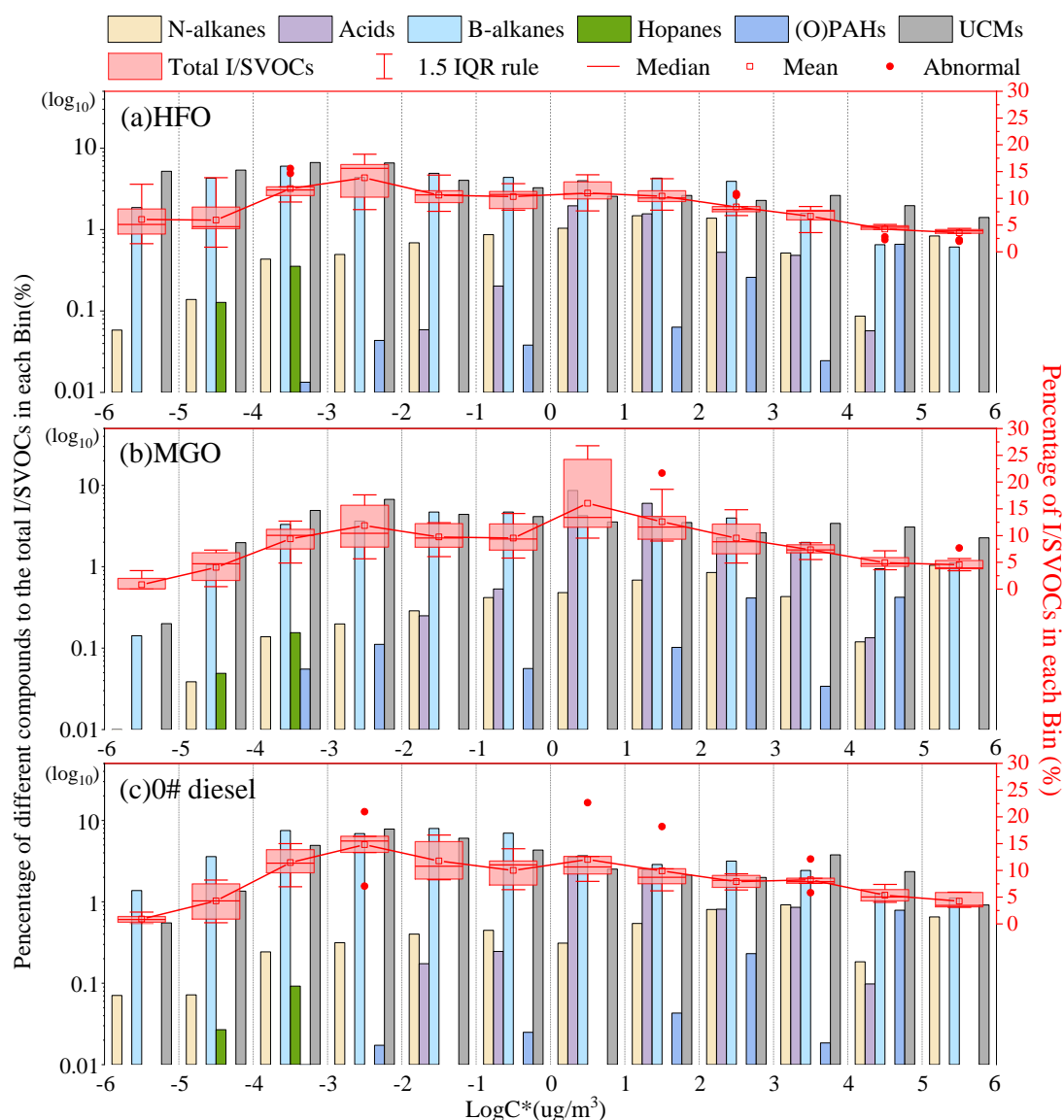


Figure 4 The volatility distributions of I/SVOCs based on the volatility basis set (VBS) framework from different fuels

(2) My second major concern is related to the method used for calculating SOA formation in Section 2.5 and the SOA formation potential (SOAFP) showed in Fig.7. As shown in Eq.3 in Line 218, the formed SOA is a function of reacted precursor concentration [HC], oxidation time, and assumed OH concentrations. I am very confused how the authors derived the SOAFP in Figure 7 without providing details on the amount of precursor reacted, oxidation time, or OH concentrations. Please refer to Figure 5 in Zhao et al. (2016) and include more detailed information on how SOA formation was calculated in this study. Furthermore, it would be helpful to compare the SOA production from ship exhausts with previous studies (Morino et al., 2022; Zhao et

al., 2015, 2016). Is the SOA production from ship exhaust higher or lower than SOA formed from vehicle exhaust?

Reply: Thank you for your insightful comments. We appreciate your attention to the details of the SOA formation calculation in Section 2.5 and the SOA Formation Potential (SOAFP) presented in Figure 7. In response to your concern, we acknowledge that the calculation of SOA in Figure 7 may lack clarity, particularly due to the omission of explicit information on the amount of precursor reacted, oxidation time, and OH concentrations.

The equation utilized for the estimation of SOA production via IVOCs in this study is as follows:

$$\Delta SOA_{IVOCs} = \sum_j [HC_j] \left(1 - e^{-k_{OH,j} [OH] \Delta t}\right) \times Y_j$$

where $[HC_j]$ represents the concentration of IVOCs species involved in the reaction, Y_j is the yield coefficient of IVOCs species, and k_{OH} is the OH reaction rate constant of precursor j at 25°C ($\text{cm}^3 \cdot \text{molecules}^{-1} \cdot \text{s}^{-1}$); $[OH]$ is the OH concentration ($\text{molecules} \cdot \text{cm}^{-3}$), which is assumed in this study to be $1.5 \times 10^6 \text{ molecules} \cdot \text{cm}^{-3}$; Δt is the photochemical age (h); and Y_i is the SOA mass yield of precursor j . And the OH reaction rate constants ($\text{cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$) and SOA yields used in this study were the same as Zhao et al. (2016), which reacted (Δt) after 48 h photo-oxidation at the OA concentration of $9 \mu\text{g}/\text{m}^3$, as explained in detail in Table S5 and Table S6.

The refined description of the SOAFP estimation method is presented in lines 229-238 of the revised manuscript.

where $[HC_j]$ represents the concentration of IVOCs species involved in the reaction, Y_j is the yield coefficient of IVOCs species, and k_{OH} is the OH reaction rate constant of precursor j at 25°C ($\text{cm}^3 \cdot \text{molecules}^{-1} \cdot \text{s}^{-1}$); $[OH]$ is the OH concentration ($\text{molecules} \cdot \text{cm}^{-3}$), which is assumed in this study to be $1.5 \times 10^6 \text{ molecules} \cdot \text{cm}^{-3}$; Δt is the photochemical age (h); and Y_j is the SOA mass yield of precursor j . And the OH reaction rate constants ($\text{cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$) and SOA yields used in this study were the

same as Zhao et al. (2016), which reacted (Δt) after 48 h photo-oxidation at the OA concentration of $9 \mu\text{g}/\text{m}^3$, the specific values of Y_j and k_{OH} under different environmental conditions were obtained from the simulation study of smoke chamber (Table S5-S6).

Furthermore, to enhance the comparative analysis, we have incorporated a detailed discussion that contrasts the SOA production from ship exhausts with findings from previous studies (Lines 647-655).

Previously, there has been limited research on SVOCs and their SOAFP, resulting in a scarcity of relevant comparative studies. Compared to diesel vehicles ($430 \pm 574 \text{ mg (kg fuel)}^{-1}$), gasoline vehicles ($39 \pm 79 \text{ mg (kg fuel)}^{-1}$) and nonroad machinery ($424 \pm 138 \text{ mg (kg fuel)}^{-1}$) reported in previous IVOC studies (Zhao et al., 2015; Zhao et al., 2016; Qi et al., 2019), SOA emissions of total I/SVOCs from ships using 0# diesel exhibit significantly higher formation potential ($634 \text{ mg (kg fuel)}^{-1}$). This discrepancy highlights critical knowledge gaps in current assessments, where the scarcity of research on SVOCs and their SOAFP has led to incomplete comparisons.

Specific comments:

The writing quality needs some improvement. Several sentences would benefit from commas instead of periods. For instance, a comma should be used after "detection methods" in Line 449. I also recommend that the authors pay closer attention to the use of conjunctions throughout the manuscript to enhance readability and clarity.

Reply: We appreciate your valuable feedback. We have conducted a thorough review of the manuscript and significantly enhanced its writing quality by refining sentence structures and improving readability. Specifically, we have replaced periods with commas where appropriate, such as after "detection methods" in line 478, to maintain the continuity of sentence. Furthermore, we have revised the use of conjunctions throughout the manuscript to ensure a smoother flow and greater clarity. All these modifications have been integrated into the revised manuscript.

Referee 3#

This study systematically evaluated the impact of improving marine fuel quality on I/SVOC emissions. An innovative finding revealed that transitioning from low-sulfur to ultra-low-sulfur oil led to a significant increase in I/SVOC emissions, which in turn elevated the secondary organic aerosol formation potential (SOAFP). Additionally, it also found that I/SVOC emissions from inland ships are substantial and should not be overlooked. The findings provide valuable insights for the development of future ultra-low-sulfur oil policies. However, several questions still require further elaboration and explanation.

(1) I fully understand the challenges the authors face in conducting ship emission tests. However, from the perspective of improving the quality of the paper, since one of the key innovations is updating the emission factors, there should be further discussion on how the new emission factors impact the pollutant emissions in the inventory or the total amount of SOA formation.

Reply: Thank you for your thoughtful feedback. We agree that further discussion on how these updated emission factors impact pollutant emissions in the inventory and the total amount of SOA formation would significantly enhance the quality of the paper. However, Given the complexity associated with developing a precise ship emission inventory and estimating SOA, we conducted a simplified case study to assess the impact of low-sulfur fuel usage on ship emissions and the overall SOA levels.

In accordance with your recommendation, we have conducted a preliminary evaluation utilizing our recently updated I/SVOC emission factors. Based on existing emission inventories derived from prior studies, the total IVOC emissions from non-road mobile sources are estimated to be 238 Gg, of which ships account for 6.16% (14.7 Gg) (Zhao et al., 2022). Assuming these ships use low-sulfur fuel (with a sulfur content of below 0.5% (m/m)) and applying the emission factors derived from this study for re-estimation, the previously unaccounted SVOCs would result in an underestimation of pollutant emissions by 11.9 Gg. If the sulfur content of the fuel is further reduced to

0.1% ultra-low sulfur fuel oil (with a sulfur content of below 0.1% (m/m)), the revised IVOCs emissions from ships would be 17.7 Gg, and SVOCs emissions would be 23.6 Gg, representing a 64% increase in total emissions compared to using low-sulfur fuel. The results demonstrated that the implementation of updated emission factors resulted in significant alterations in pollutant emissions. Notably, the transition to lower sulfur fuels increased the contribution of IVOCs and SVOCs, consequently enhancing the overall potential for SOA formation potential. These findings highlight the indispensable role of current emission factors in precisely quantifying the contributions of shipping activities to SOA generation and enhancing the reliability of emission inventories, especially within the context of evolving regulatory frameworks and fuel standards. We have expanded this discussion in the manuscript and will continue to refine our analysis in response to additional feedback, as detailed from manuscript lines 686-698.

Due to data limitations, previous emission inventories for ships often underestimated the emissions of organic matter, particularly from I/SVOCs. This underestimation has led to an incomplete assessment of the contribution of ship emissions to air quality, particularly in terms of SOA formation. Specifically, according to existing emission inventories based on previous studies, the total IVOCs emissions from non-road mobile sources amount to 238 Gg, with ships contributing 6.16% (14.7 Gg) of this total (Zhao et al., 2022). Assuming these ships use low-sulfur fuel (with a sulfur content of below 0.5% (m/m)) and applying the emission factors from this study for re-estimation, the unconsidered SVOCs would lead to an underestimation of 11.9 Gg pollutant emissions. If the sulfur content of the fuel is further reduced to 0.1% ultra-low sulfur fuel oil (with a sulfur content of below 0.1% (m/m)), the revised IVOCs emissions from ships would be 17.7 Gg, and SVOCs emissions would be 23.6 Gg, representing a 64% increase in total emissions compared to using low-sulfur fuel.

I would like to extend our sincere gratitude for your insightful suggestions, which have significantly enhanced the academic depth and clarity of our work.

(2) It is suggested that Table S2 be placed in the main text.

Reply: Thank you for your suggestion. We agree that placing Table S2 in the main text would improve its accessibility to the readers. Consequently, we have relocated Table S2 to the main body of the manuscript, where it is now presented as Table 1.

(3) Line 243-244: In the study of emissions from different types of ships (OGVs and ICSs), are there any significant differences in the emission characteristics of I/SVOCs between new ships in this study and old ships from previous studies of the same type? If so, what could be the reasons for these differences?

Reply: Thank you for your thoughtful question. The emission characteristics of I/SVOCs from different types of ships (OGVs and ICSs) in this study indeed show some differences compared to those observed in previous studies on older vessels of the same type. Compared to ship measurement results using ultra-high sulfur fuel oil prior to 2016, the emission factor derived from this study was lower than previously reported values (Perrone et al., 2014). Compared to similar low-sulfur airborne measurement results from OGVs, the average EF_{IVOC} level in this study was consistent (Huang et al., 2018b). Furthermore, there are notable differences in the emission characteristics of I/SVOCs between newer and older ships. Taking polycyclic aromatic hydrocarbons (PAHs) as an example, older ships typically use high-sulfur residual fuel oil (with sulfur content of more than 3.5%) and outdated engine technology, resulting in higher PAH emission intensities. In certain types of vessels, such as oil tankers and fishing ships, the use of traditional high-sulfur heavy fuel oil (HFO) resulted in higher levels of polycyclic aromatic hydrocarbons (PAHs) emissions (Perrone et al., 2014; Zhang et al., 2014). In contrast, ships that use low-sulfur light fuels, such as marine gas oil (MGO) or other cleaner fuels, exhibited lower emissions of PAHs (Liu et al., 2022). Under the guidance of the International Maritime Organization (IMO) policies, there has been a further reduction in sulfur content in fuels to mitigate environmental pollution. However, despite the perceived environmental benefits of low-sulfur oil, some studies have shown relatively higher levels of PAH emissions compared to high-sulfur oil. This phenomenon can be attributed to the intricate chemical reactions and

transformations that occur during the desulfurization process of the fuel, resulting in an increased generation and release of PAHs. Additionally, the operational conditions and types of vessels also influence the emission characteristics of PAHs. The emission of I/SVOCs from ships is influenced by multiple factors, including fuel type, engine type, and engine conditions. Among these factors, fuel type plays a predominant role in shaping the emission characteristics. The differences in fuel types utilized in the ships examined in this study, compared to those used in older vessels from previous studies, likely account for the observed variations in I/SVOC emissions. Additionally, advancements in engine technologies and modifications in operational conditions can substantially impact emission factors and profiles.

(4) Line 423-425: Considering the importance of fatty acids in ship exhausts, what are the possible sources of fatty acids in the atmosphere besides fuel combustion? And how can their contributions from different sources be differentiated and quantified?

Reply: Thank you for your insightful question. Fatty acids in ship exhausts constitute a significant portion of the organic matter in atmospheric aerosols, particularly in marine aerosols. However, they can also originate from various other sources such as biomass burning (especially cooking and heating), industrial activities (e.g., chemical production), and animal farming (manure and waste). (Hu et al., 2023; Kawamura et al., 2010; Li et al., 2020). Moreover, oceanic microalgae and phytoplankton also produce fatty acids, which can be transferred to the atmosphere as part of marine aerosol particles (Kawamura et al., 2017). In addition, fatty acids can also be generated in the atmosphere through photochemical reactions or by the oxidation of precursor compounds such as unsaturated hydrocarbons or aldehydes (Li et al., 2020).

To differentiate and quantify the contributions of fatty acids from these various sources, several approaches can be used. Techniques like positive matrix factorization (PMF) can be applied to identify and quantify source contributions based on the chemical fingerprint of fatty acids and other co-emitted species in the atmosphere (Molnár et al., 2014; Wang et al., 2015). Isotopic signatures of fatty acids can also be

used to distinguish between biogenic and anthropogenic sources. For instance, a study by Swales and Gibbs (2020) investigated the isotopic signatures of fatty acid soil biomarkers under varying land use scenarios, offering valuable insights into how land use changes impact these signatures. Additionally, a review by Twining et al (2020) discussed the application of stable isotopes of fatty acids in ecological studies, emphasizing their utility in tracing energy flows and identifying sources. (Twining et al., 2020). Moreover, specific fatty acids or fatty acid ratios can serve as tracers for distinct sources. For instance, particular fatty acids may be indicative of specific marine organisms or combustion byproducts, thereby facilitating source identification through chemical profiling. Incorporating these methodologies into atmospheric studies can provide a more accurate understanding of the origins of fatty acids and their relative contributions to atmospheric fatty acid concentrations.

(5) Section 3.5: In the investigation of SOA formation potential, the contribution of different I/SVOC components is discussed. How sensitive is the SOA formation potential to variations in the relative proportions of these components? Could a minor alteration in the ratio of specific I/SVOCs significantly influence overall SOA formation?

Reply: When studying SOA formation potential, the SOA formation potential (SOAFP) shows a notable sensitivity to changes in the relative proportions of I/SVOC components. Studies have shown that the relative proportions of these components play a crucial role in determining overall SOA formation. This is particularly evident when analyzing emission factors from different fuel types, where SOAFP is significantly influenced by the ratio of VOCs to SVOCs. Taking the OGV in this study as an example, switching from HFO with a sulfur content of 0.5% to MGO with a sulfur content of 0.1% led to marked changes in emissions and SOA production. SVOC emissions increased by 4%, resulting in a 3% increase in SOA yield. Conversely, intermediate volatility organic compound (IVOC) emissions rose by 74%, corresponding to a substantial 55% increase in SOA production. The predominant contribution to the total SOAFPs was primarily attributed to b-alkanes and UCM, comprising 81.1% to 87.8%. However, it is crucial to emphasize that the role of acids, especially in IVOCs, should

not be overlooked. For example, the emission factor contributions of acids for 0# diesel, MGO and HFO were 5%, 10% and 3%, respectively, while their corresponding SOAFP contributions reached 7%, 12% and 5%, respectively. Consequently, even minor variations in the I/SVOC ratio can significantly influence overall SOA formation. Therefore, future research should thoroughly investigate the specific impacts of these proportion changes on SOA generation and systematically analyze the variations in their sensitivity under diverse environmental and policy conditions. This will provide a robust scientific foundation for the development of effective emission reduction control measures.

(6) In the conclusion section, in addition to summarizing the main findings, it would be valuable to propose potential future research directions based on the limitations identified in this study. For instance, with respect to the UCM analysis, it is crucial to elaborate on the specific enhancements required for the experimental methodology. Are there alternative extraction techniques or advanced analytical instruments that could potentially enhance the identification and quantification of UCM components, such as GC \times GC-MS? A more comprehensive investigation into these aspects would substantially enhance the value of the conclusion and more effectively guide future research endeavors.

Reply: Thank you for your insightful suggestion. In the conclusion section, we not only summarize the main findings but also propose potential future research directions based on the limitations identified in this study. Regarding the UCM analysis, we agree that further refinement of the experimental methodology is essential. Since UCM is composed of a complex array of compounds that can often overlap in conventional single-dimensional Gas Chromatography (GC), employing Two-Dimensional Gas Chromatography (GC \times GC) offers a powerful approach for separating and identifying more components (Tang et al., 2023). GC \times GC enhances separation by utilizing two orthogonal columns with different stationary phases, thus allowing for improved resolution of complex mixtures like UCM (Marriott, 2004). Moreover, Orbitrap mass

spectrometry provides ultra-high resolution and high mass accuracy, which can significantly improve the identification of individual compounds in the UCM (Kösling et al., 2022). Its ability to distinguish isomeric compounds and detect low-abundance species with high sensitivity is crucial for characterizing complex mixtures. While GC×GC and Orbitrap MS are individually powerful techniques, their combination with complementary methods such as high-resolution liquid chromatography (HRLC) or high-resolution mass spectrometry (HRMS) would significantly enhance the characterization of UCM. Integrating these techniques could markedly improve the sensitivity and specificity of UCM analysis. By addressing these aspects, future research could provide a more detailed understanding of UCM characteristics, thereby enhancing the precision of source apportionment studies and the overall assessment of environmental pollutants. We have incorporated these recommendations into the revised conclusion in lines 707-710 to ensure the study provides clear guidance for future research efforts:

Future research can obtain more comprehensive and accurate organic component emission data by using advanced methods such as comprehensive two-dimensional gas chromatography-mass spectrometry (GC × GC-MS) and Orbitrap mass spectrometry.

(7) It is recommended to review and standardize the unit notations for better readability:

For example, in the process of converting original measurement data into emission factors, the presentation of units for each variable in the text is not coherent enough. For the concentration representation of some chemical substances, such as the concentration of certain substances in the gas or particle phase in the description of sampling and analysis processes, there is no explanation in different paragraphs or charts whether different representation methods or units are used. The author should review and standardize the unit notations for better understanding.

Reply: We appreciate your clarification. In response, we have thoroughly reviewed the manuscript to ensure consistent representation and explanation of emission factor units (mg (kg fuel)^{-1}) throughout the text, as all units in the manuscript refer to emission factors.

(8) Figure 4: It is necessary to improve the readability;

Reply: Thank you for your suggestion. The quality of Figure 4 has been improved.

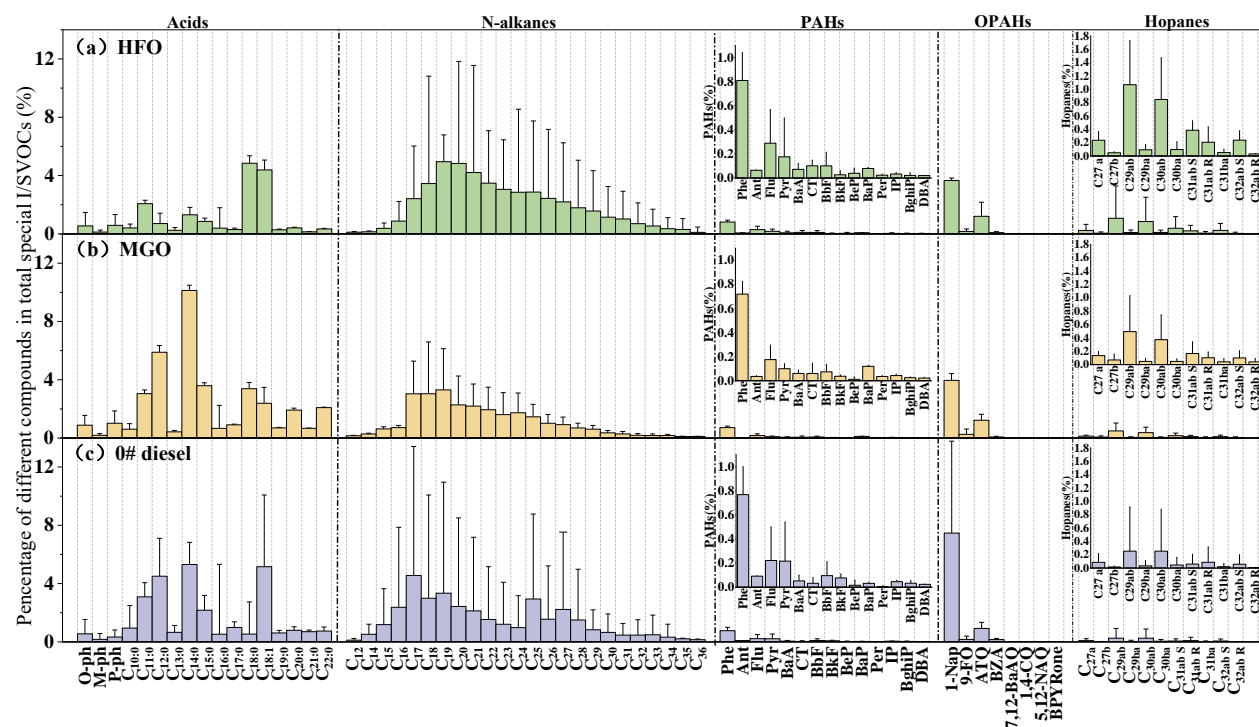


Figure 4 Profiles of I/SVOCs in ship exhausts under different fuels

Figure 6: the word“hapones”should be corrected to“hopanes”.

Reply: We appreciate your attention to detail. In the revised Figure 6, the term "hapones" has been corrected to "hopanes." Moreover, Figure 6 has also been revised as the volatility distributions of I/SVOCs based on the volatility basis set (VBS) framework from different fuels for improved clarity.

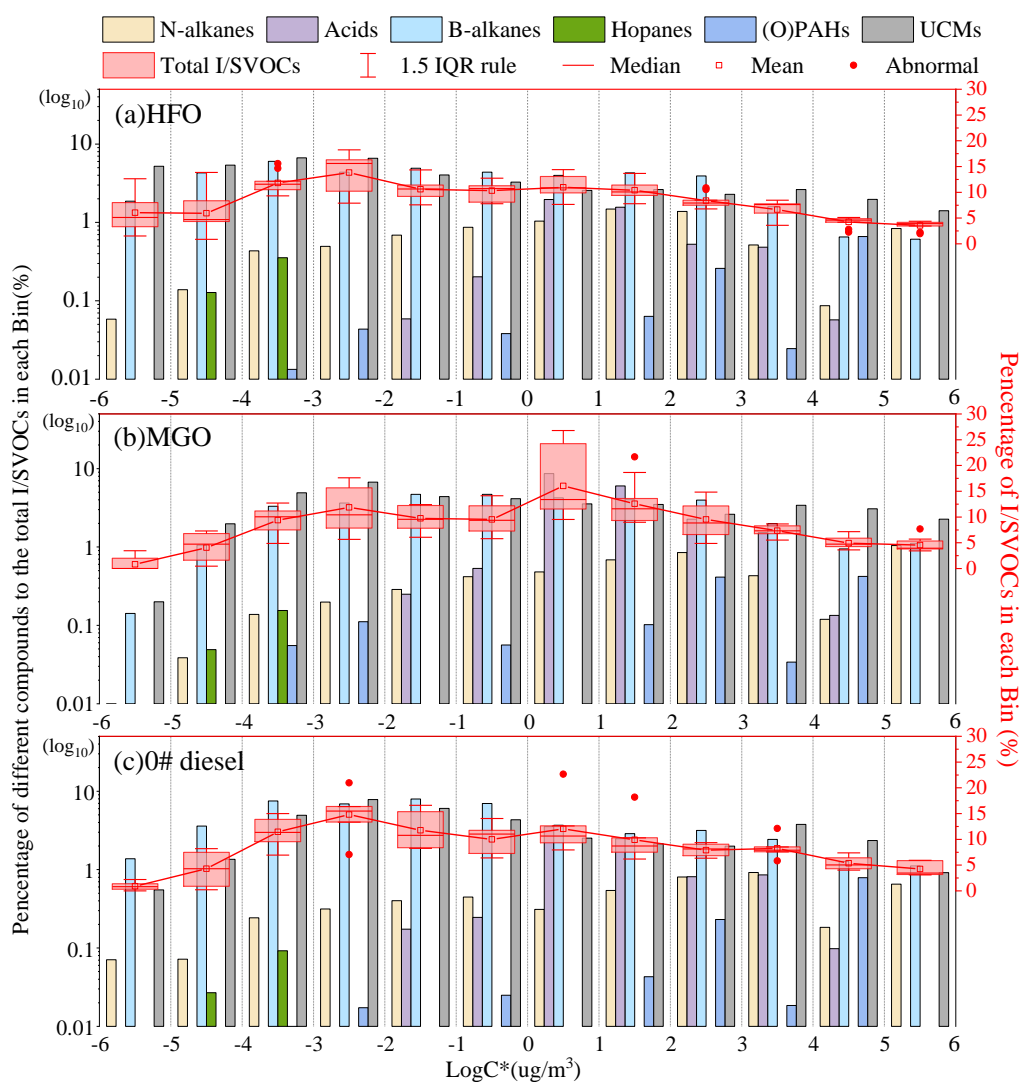


Figure 6 The volatility distributions of I/SVOCs based on the volatility basis set framework from different fuels

(9) The article contains several minor grammatical and expressive errors that require thorough examination and correction to enhance its overall quality.

Line 406-408: "composition" and "compositions" should be unified;

Reply: Thank you for your observation. All instances of "composition" have been corrected to "compositions" for uniformity.

Line 413: "accounting for average of..." is an incorrect expression and should be changed to "accounting for an average of..."

Reply: Thank you for your suggestion. We have revised the expression from "accounting for average of..." to "accounting for an average of..." to ensure correct grammatical structure.

Referee 4#

In this study, the authors evaluated the impact of the transition from high-sulfur to low-sulfur on intermediate/semi-volatile organic matters (I/SVOCs) from ocean-going vessels and inland cargo ships. An increase of I/SVOCs has been found as the sulfur content decreased. This novel finding is very helpful for future oil policies. Although this is an interesting paper, certain clarification and revisions are needed. I recommend the publication after the authors address my comments below.

Major comment:

My major concern lies in the measurement. Section 2 should provide further details about the measurement. What is the clean air used in the sampling system, is it the same as zero air where there is no particle? It is important that the system isolates particles and gases originating exclusively from the ship exhaust, minimizing the potential for contamination from external sources. What measures have been taken to ensure this? Besides, is there a cutoff size of the aerosol or is it total suspended particles (TSP)? What is the uncertainty of the measurement? How long is the sampling time? How many samples were collected? It is beneficial to include a table giving an overview of all the samples, including the ship, engine type, fuel type, and operating mode, similar to Table S2 in Zhang et al. (2024). It can be put in the supplement.

Reply: Thank you for your question. In our sampling system, the clean air refers to ambient air that has undergone filtration through a clean air system equipped with dual particle filters, effectively eliminating particulate matter. This differs from the rigorously defined "zero air," which is purified air devoid of both particulate matter and aerosol precursors. Given the substantial demand for clean air in field sampling and the considerable cost associated with preparing zero air, this study utilized filtered ambient air as an alternative. Despite the potential presence of trace gaseous interferences, preliminary measurements confirmed that their concentrations ($\text{CO} < 1 \text{ ppm}$, $\text{SO}_2 < 10 \text{ ppb}$, $\text{NO}_x < 5 \text{ ppb}$, $\text{VOCs} < 1 \text{ ppb}$) were significantly lower than those in ship exhaust emissions ($\text{CO} > 50 \text{ ppm}$, $\text{SO}_2 > 1 \text{ ppm}$, $\text{NO}_x > 50 \text{ ppb}$, $\text{VOCs} > 50 \text{ ppb}$), with an uncertainty of less than 2%. Consequently, their influence on the experimental results is considered negligible.

To ensure that the sampling system exclusively captures particles and gases from ship exhaust while minimizing interference from external pollution sources, a sampling pipe (about 3 m) was used to route emissions from stack of ships. Then, the probe of a flue gas analyzer was placed into the vessel exhaust pipe to test the gaseous matters directly. Particulate matter was directly sampled after being diluted by the clean air mentioned above. This ensures that the collected gases and particles are exclusively from the ship's exhaust. The sampling tube was designed with a tightly sealed connection to isolate it from the outside air, preventing the entry of external pollutants. Additionally, to prevent interference from external gases, the dilution system is equipped with specially designed closed pipelines and splitters. The system is regularly cleaned and calibrated to guarantee that only ship exhaust is analyzed. These measures guarantee that the collected samples precisely represent the composition of ship exhaust emissions, while simultaneously minimizing interference from external air pollution sources.

In our study, the aerosol collected from the ship exhausts was total suspended particles (TSP), encompassing particles of all size ranges. This has been clarified in lines 174-175 of the revised manuscript. In this study, a total of 64 samples were collected. Detailed information for each sample, including sampling time, vessel type, engine type, fuel type, and operational mode, has been provided in Table S3.

Table S3 Engine type, operating mode, and fuel type of each ship for each measurement

Ship ID	Engine type	Operating mode	Sampling duration	Ship ID	Engine type	Operating mode	Sampling duration
OGV1	Main	20%_MGO ^a	20 min	OGV3	Auxiliary	50%_MGO	20 min
	engine	75%_MGO	20 min		engine	50%_HFO	20 min
	Auxiliary	75%_MGO	27 min			75%_HFO	20 min
	engine	(NCR)					
OGV2	Main	25%_HFO	20 min	ICS1	Main	Maneuvering_0#diesel	20 min
	engine	50%_HFO	20 min		engine	Cruise_0#diesel	20 min

		75%_HFO	20 min	ICS2	Main	Maneuvering_0#diesel	20 min
Auxiliary engine		85%_HFO	20 min		engine	Cruise_0#diesel	20 min
		100%_HFO	20 min	ICS3	Main	Maneuvering_0#diesel	20 min
		50%_MGO	25 min		engine	Cruise_0#diesel	20 min
		50%_HFO	70 min	ICS4	Main	Maneuvering_0#diesel	20 min
OGV3	Main	75%_MGO	40 min		engine	Cruise_0#diesel	20 min
	engine	25%_HFO	20 min				
		50%_HFO	10 min				
		75%_HFO	40 min				
		95%_HFO	40 min				

^a, means percentage of engine load under what type of fuel

Specific comments:

(1) Line 57-58. “shipping emissions are responsible for 25% of fine particulate matter” globally? Please clarify.

Reply: We sincerely apologize for any confusion arising from the typesetting issues in the original manuscript, which led to ambiguity in the reported values. The correct statement should be as follows:

“Results show that shipping emissions are responsible for 2%-5% of fine particulate matter (PM_{2.5})”

(2) Line 358-366. It seems that when analyzing the impact of operating mode (Fig. 2b), all the OGVs and ICSs, fuel types, and engine types are used. Will the result be different if we investigate samples with the same fuel and engine types to exclude their impact?

Reply: We sincerely appreciate the reviewer for raising this important question. Indeed, our current analysis of operating modes (Fig. 2b) encompasses all ship types, fuel types, and engine types, which may introduce variability from these factors. However, upon re-examination of the original data, we discovered that the initial figure (Fig. 2b) displayed trend discrepancies attributable to misclassification errors in the

data. We have now revised both the figure and the manuscript accordingly.

After revision, even if the analysis is limited to a subset of samples with the same fuel type and engine type (See in Fig. 3 (A)), the trend of the impact of operating modes on I/SVOC emissions remains relatively consistent with the comprehensive analysis presented in Figure 2 (b). This study primarily aims to examine the overall impact of various aspects of ships from an integrated perspective. Additionally, due to the insufficient sample size for individual variables, which limits representativeness, we included all OGVs and ICSs in the analysis. Incorporating the aforementioned updates, we have thoroughly revised the manuscript to provide a more comprehensive and robust interpretation of the findings (lines 385-406 in the revised manuscript). We appreciate your valuable suggestions once again, which have significantly enhanced the clarity of our analysis.

It could be seen from Figure 2 (b) that the average emission factors (EFs) of I/SVOCs in this study followed a relatively ascending order across operating modes: $1098 \pm 305 \text{ mg (kg fuel)}^{-1}$ in low, $1542 \pm 465 \text{ mg (kg fuel)}^{-1}$ in medium and $1457 \pm 276 \text{ mg (kg fuel)}^{-1}$ in high operating modes, respectively in this study, revealing significantly elevated emissions at both medium and high loads compared to low-load conditions. This trend was consistent with PM emission patterns reported by Zhang et al. (2021), but notably diverged from the characteristics of ship-emitted IVOCs, which reached its lowest value under medium-load conditions in prior studies (Zhao et al., 2014;Huang et al., 2018b). This discrepancy could be attributed to the dominance of SVOCs over IVOCs in this study. Operating modes affect the combustion state in engines and the air-fuel ratio during the combustion process, thereby influencing exhaust emissions (Shrivastava and Nath Verma, 2020). The air-fuel ratio exhibits a decreasing trend with increasing engine load, which induces more pronounced incomplete combustion within the cylinder, thereby establishing a direct causative relationship with elevated total I/SVOC emissions. (Zhang et al., 2021; Watson et al., 1994). The high operating mode is associated with reduced fuel diffusion and combustion time, leading to a partial oxygen deficiency within the cylinder, thereby

resulting in an increased generation of I/SVOCs (Zhao et al., 2016;Liu et al., 2022). The investigation of methodologies for optimizing engine design and control systems to achieve enhanced combustion efficiency under different operating conditions, thus reducing emissions, is of great significance.

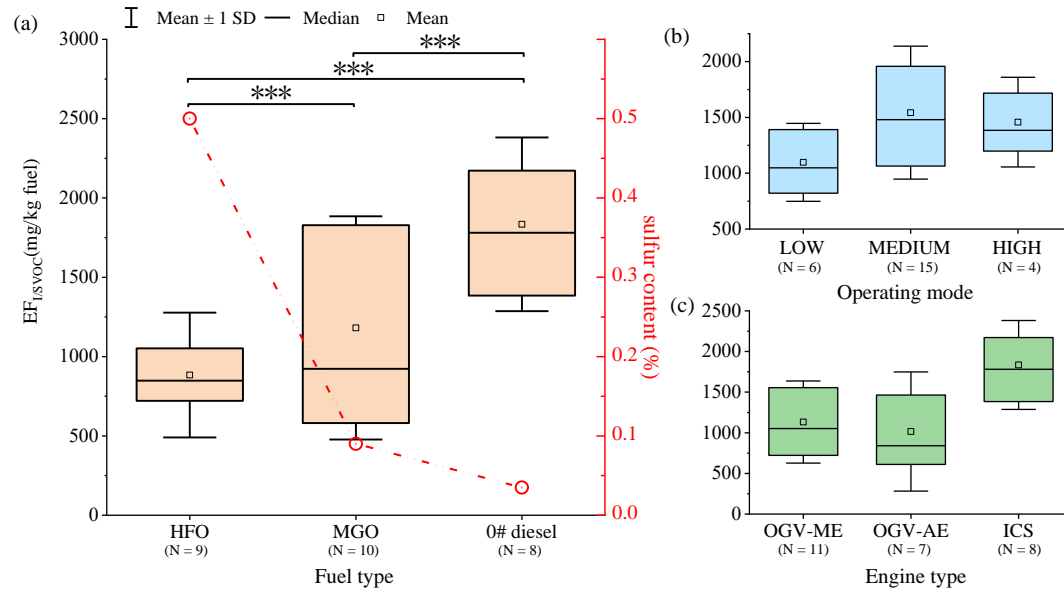


Figure 2 Box-whisker plots of total EFI/SVOCs for the tested ships under (a) different fuel types, (b) different operating modes, and (c) different engine types. N represents the number of samples. Significant differences between samples were determined using an independent samples T-test. The error bars represent the standard deviation of the measured values, while *** indicates a significance level of $p < 0.001$.

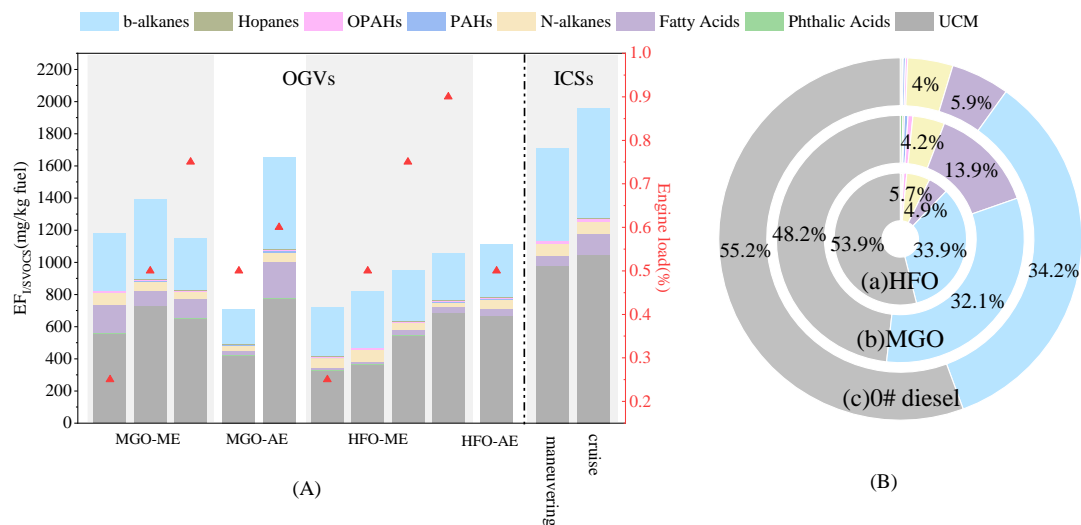


Figure 3. Chemical compositions of I/SVOCs from the tested ships, (A) emission factors (mg (kg fuel)^{-1}) and (B) fractional contributions (%). The red triangles represent the engine load.

(3) Figure 2 in line 402. I would suggest replacing the x-axis label with the figure title.

More details are needed in the figure caption. The elements of boxplots need to be added. Besides, I do not see error bars. By error bars, the authors might mean whiskers. How many samples are there in each box, it will be more robust if the amount of samples could be shown.

Reply: Thank you for your valuable feedback. We have thoroughly reviewed your suggestions and implemented the necessary revisions. Specifically, we have incorporated elements of a box plot and added error bars to enhance clarity. These modifications have been incorporated into the revised manuscript to improve the clarity and accuracy of the figure. Thanks again for your helpful suggestions.

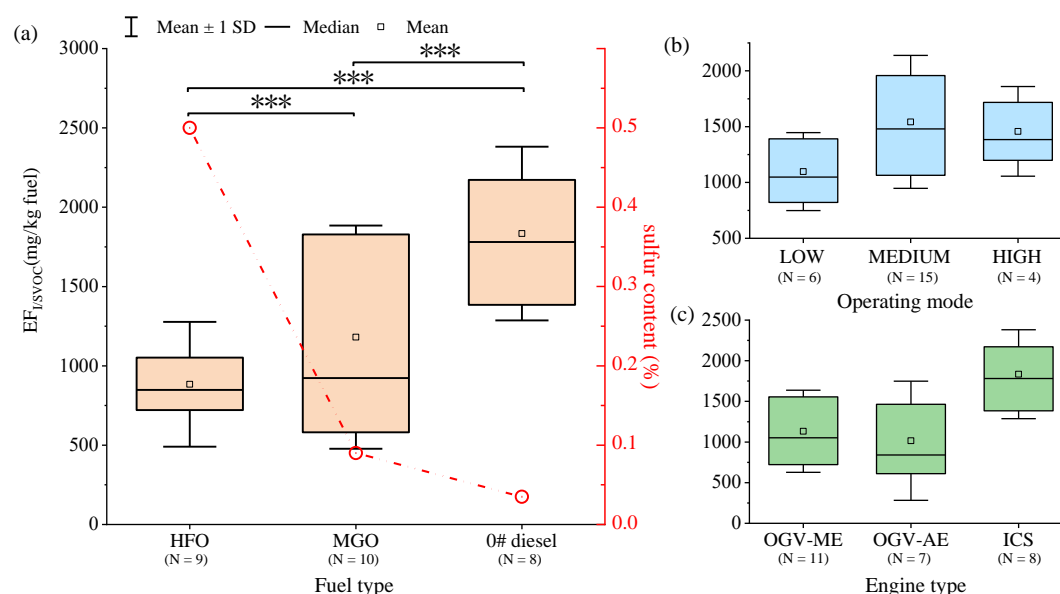


Figure 2 Box-whisker plots of total EFI/SVOCs for the tested ships under (a) different fuel types, (b) different operating modes, and (c) different engine types. N represents the number of samples. Significant differences between samples were determined using an independent samples T-test. The error bars represent the standard deviation of the measured values, while *** indicates a significance level of $p < 0.001$.

(4) 372-374. This sentence needs to be revised. Is an “and” missing?

Reply: Thank you. Indeed, the sentence would benefit from the addition of "and" to enhance clarity and readability. We have revised the sentence in lines 395-397 in the improved manuscript to ensure proper coherence and fluidity.

“Operating modes affect the combustion state in engines and the air-fuel ratio during the combustion process, thereby influencing exhaust emissions.”

Thank you for pointing this out.

(5) Figure 3 in line 453. Why Fig. 3b is called “distributions”? Do the authors mean fractions? I would suggest the same as Fig. 2 that more details need to be added in the figure caption.

Reply: We sincerely appreciate your insightful suggestions. In response, we have revised the figure title from "distributions" to "fractional contributions (%)" to eliminate potential ambiguity. The figure caption has also been improved accordingly.

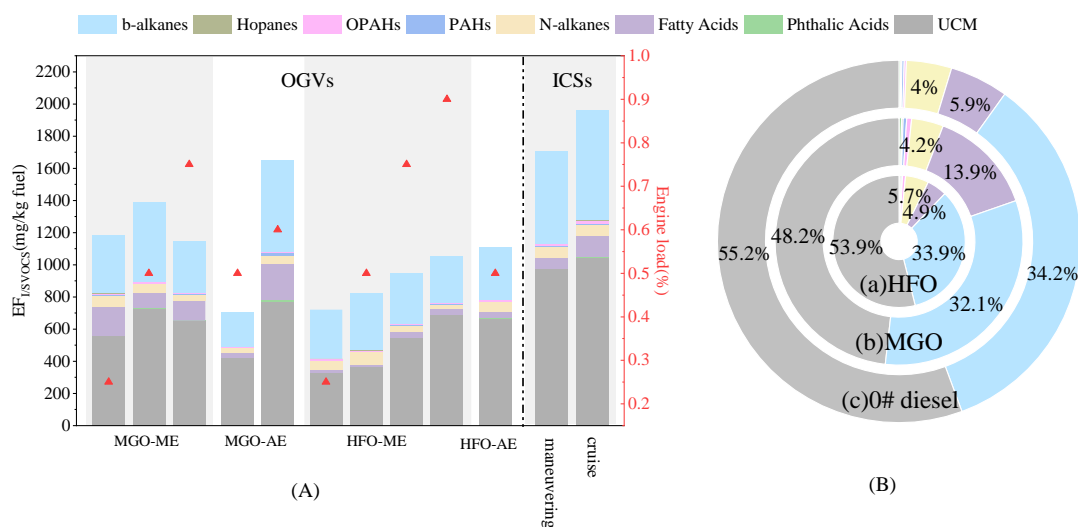


Figure 3. Chemical compositions of I/SVOCs from the tested ships, (A) emission factors (mg (kg fuel)⁻¹) and (B) fractional contributions (%). The red triangles represent the engine load.

(6) Figure 4 in line 545. Please improve the readability.

Reply: Thank you. Figure 4 has been improved as following:

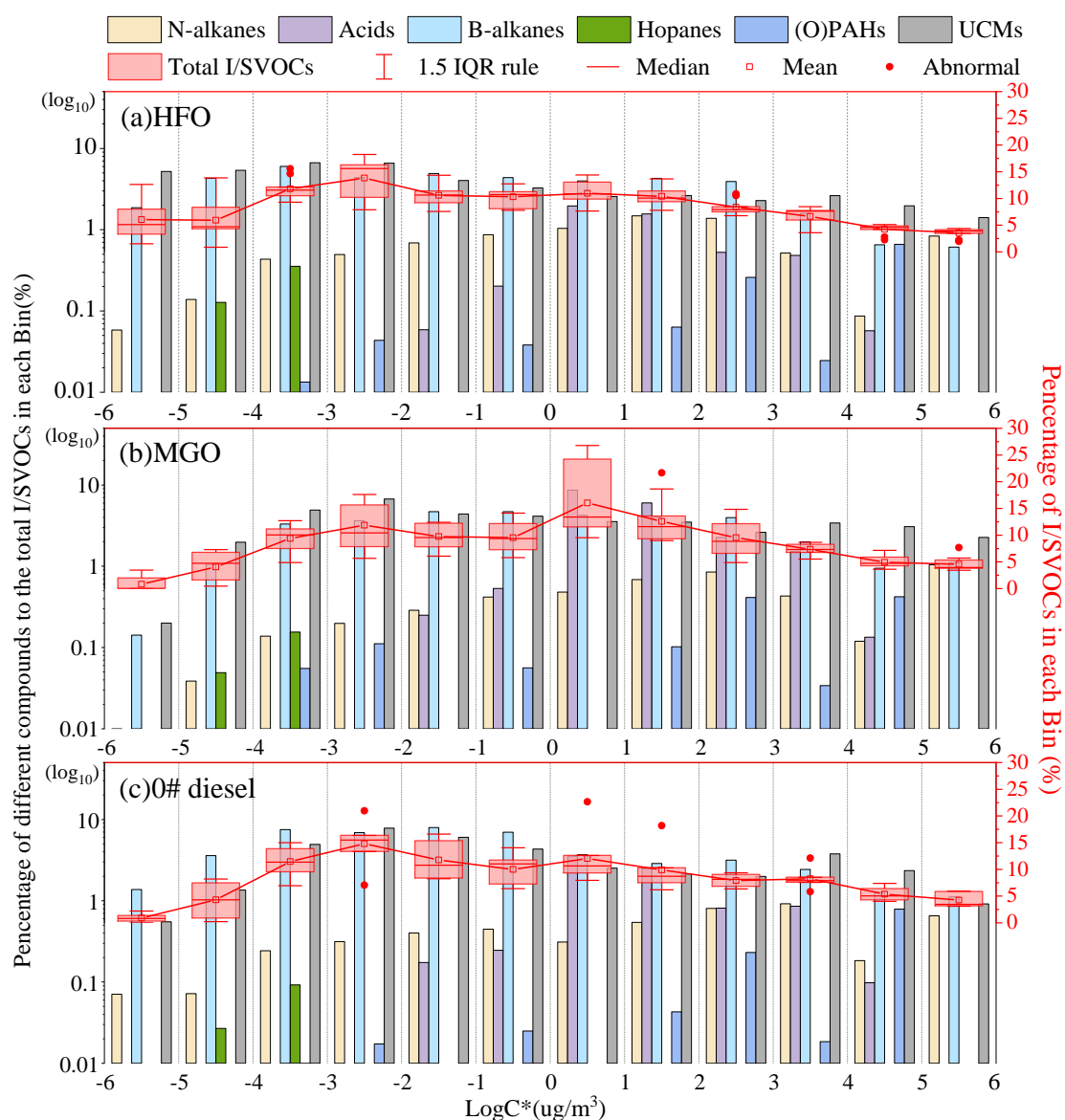


Figure 6 The volatility distributions of I/SVOCs based on the volatility basis set (VBS) framework from different fuels

Figure 6 in line 597. Hapones should be hopanes. It is necessary to improve the readability. The authors can try to use the log scale. More details need to be added in the figure caption as well.

Reply: Thanks very much for your suggestions. The term "Hapones" has been corrected to "hopanes". We recognize the advantages of employing a logarithmic scale for data presentation and have consequently adjusted the Y-axis to adopt a logarithmic scale. Moreover, the figure has been revised to illustrate the volatility distributions of

I/SVOCs based on the Volatility Basis Set (VBS) framework from different fuels, enhancing its clarity and precision.

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