We greatly appreciate the review's 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 the review's 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 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 × 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×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 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.

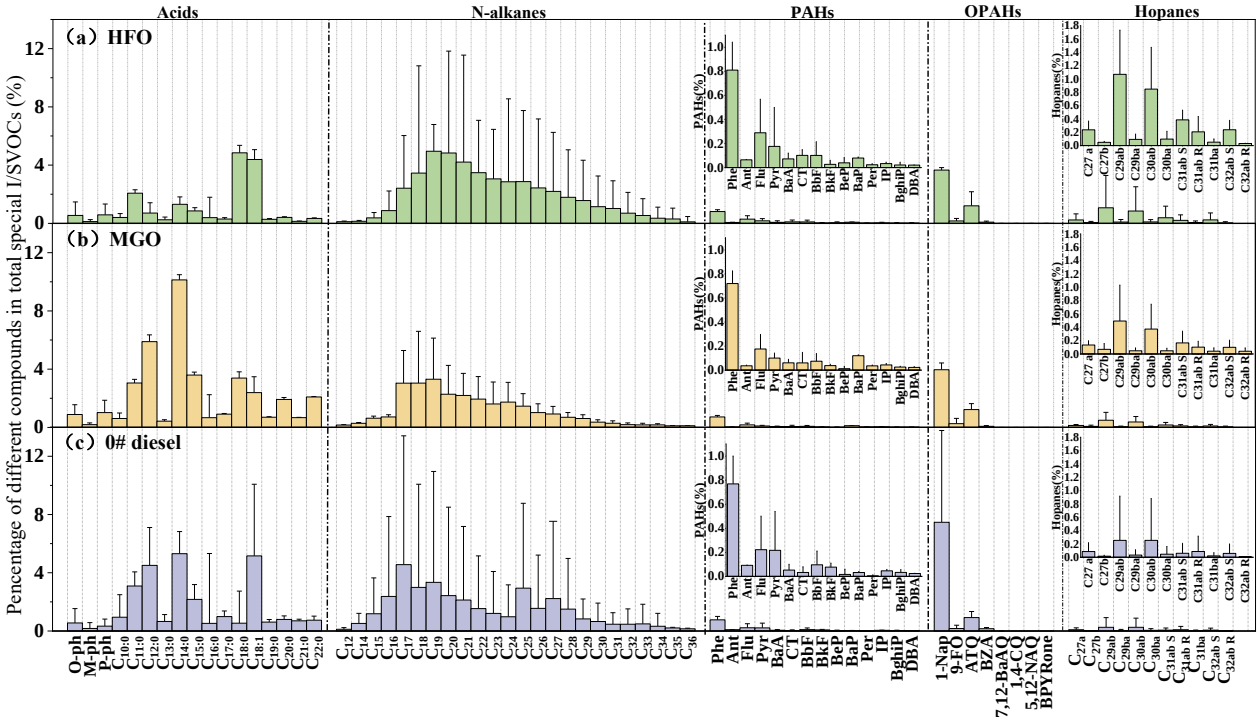

Figure 4 Profiles of I/SVOCs in ship exhausts under different fuels

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.

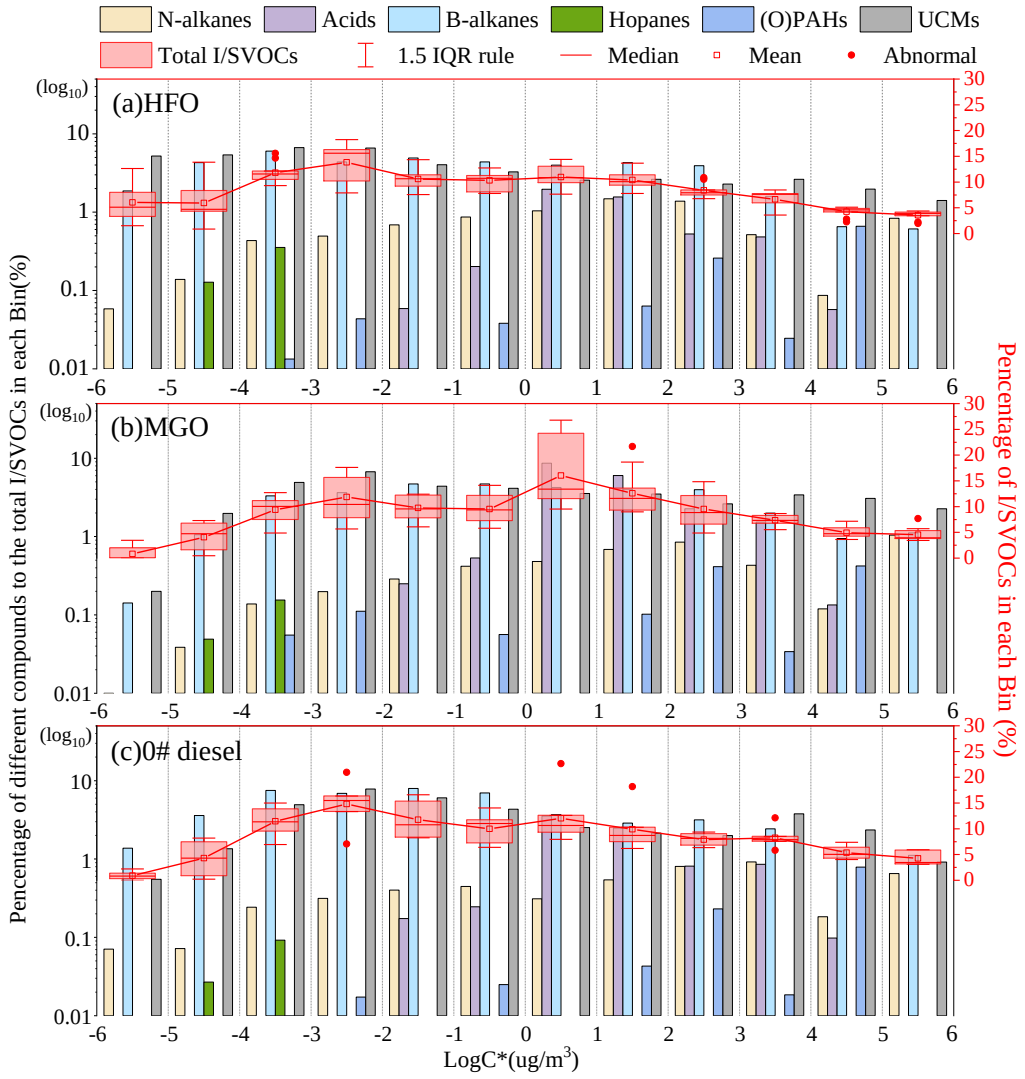

Figure 6 The volatility distributions of I/SVOCs based on the volatility basis set framework from different fuels

(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.

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