

Interactive comment on “Molecular Characterization of Polar Organic Matters in Off-road Engine Emissions Using Fourier Transform Ion Cyclotron Resonance Mass Spectrometry (FT-ICR MS): New Direction to Find Biomarkers” by M. Cui et al.

Anonymous Referee #1

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Overall comments: The study focused on the elemental compositions of particulate matters emitted from the typical off-road engines in China. The chemical structures and molecular information of major emissions are inferred in some cases, which should potentially benefit the research community in terms of the tracers indicating off-road engine emissions. Differences were clearly demonstrated between the emissions from diesel engines and those fueled by heavy fuel oils, particularly the degree of unsaturation and oxidation. While the paper will likely be a significant contribution, revisions and

clarifications are needed at this moment. Firstly, the English should be substantially improved before I can comprehensively evaluate the quality and value of the paper. The language and writing skills in the present manuscript seriously hinder the transferring of knowledge to the readers, as well as the objective evaluation on the work. I would recommend the manuscript to be edited by an editing company. Then, the significance of the paper should be more clearly demonstrated. The knowledge gaps illustrated in the introduction include (1) the unavailability of unique tracers for separating the on-road and off-road engine emissions; and (2) the challenge in detecting the large molecule and polar markers by the conventional GC-MS. However, I do not think the study filled the gaps sufficiently. How will the elemental compositions of off-road engine emissions contribute to the identification of these emissions in airborne particulate matters? Obviously, the molecular information of the tracers inferred in this study is inadequate. How about the differences in elemental compositions between the on-road and off-road engine emissions? Can the findings in this study be directly used in the concurrent source apportionment techniques, such as the filter based and AMS based source apportionment? In other words, there should be a section in this paper showing the implications of the study.

Specific comments: 1. Improve the English throughout the manuscript. Lines 13-15, page 3, line 1, page 4, lines 10-13, page 4. Too many grammatical errors, and I cannot list of them here. 2. Methodology: How many samples were collected in total and in each scenario? How did you combine the samples? What was the purpose of combining the samples given the expected enough loading of PM for chemical analysis? How to consider the variations among the samples collected in the same scenarios? The representativeness of the samples should be discussed. 3. Off-road and non-road are alternatively used. Keep consistent throughout. 4. Lines 5-8, page 12: Do you mean the number of peaks for CHO compounds? The similarly inaccurate descriptions appeared many times in the manuscript, which need to be double checked and clarified. 5. Lines 8-12, page 12: What are the ranges of number of peaks detected in biomass and coal combustions, and the references? 6. Lines 1-5, page 14: References must be

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provided to support the interpretations on the regions in Figure 2. 7. Lines 9-13, page 16: How does kinematical viscosity account for the high oxygen content in HFO-fueled vessel emissions? 8. Lines 4-6, page 17. I do not get the point why the discussions only focus on the excavators under the working mode and vessels using HFO, given that the CHON structures were different even among the excavators as mentioned in the previous sentence. 9. Lines 8-20, page 17: The inferences on the structures of CHON must be illustrated in more details. Was any of the proposed compounds ever reported in previous studies? 10. Figure 3: I do not think the structures of dinitrophenol and methyl dinitrophenol have been correctly presented in Figure 3, same for nitronaphthol and methyl nitronaphthol. Please clarify. 11. Lines 1-6, page 20. What are the bases that the conclusions can be drawn? For example, "The most of S-containing compounds emitted from off-road diesel engines were aliphatic with long chains and sulfate fraction". 12. Lines 9-11, page 20: Descriptions should be more accurate. I suppose you mean that organic sulfates were the most important S-containing compounds emitted from off-road engines. 13. Line 12, page 20: This expression " $O^*(O-3)$ " will mislead the readers. Change it to O^* ($O^* = O - 3$). 14. As mentioned earlier, the implications of this study should be summarized and clearly demonstrated, rather than a simple summary of the findings. 15. English and writing skills must be substantially improved. Otherwise, it will be impossible for this paper to be published on ACP.

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