

Anonymous Referee #2, 28 Apr 2025

Measurement report: Investigation of Optical Properties of Different Fuels Diesel Exhaust by an Atmospheric Simulation Chamber experiment by Danelli et al., 2025 report the results of a study conducted to investigate the optical properties of aerosol produced from various diesel fuels. The experiments conducted were well designed and the resulting findings fit the scope of ACP. Results are of interest and demonstrate the effectiveness of using optical properties in evaluating EC; however, there are multiple instances where I could not follow the author's line of thought. The introduction and methods are well-written, with appropriate references to related literature. The results/discussion/conclusion as currently written do not adequately describe the measurements performed or aspects of how these measurements compare with existing literature. I favor publication after major revisions. I encourage the authors to carefully address the comments listed below to produce a revised manuscript for resubmission.

Major Comments

C1. L306 – “The study successfully quantified the EC/OC composition.” Results and explicit discussion of EC:OC is lacking. A figure/table summarizing the analysis of the EC:OC (or EC:TC) ratio for the carbonaceous aerosol produced would strengthen the measurements rather than just a description of the mass concentrations of TC. Are there more results from the Sunset EC/OC analyzer to assist with this that are not currently included?

We thank the Reviewer for the comment. We agree that a more detailed presentation and argumentation of the EC:OC composition would enhance the robustness of the manuscript. We have now added a new table in Section 3.1 (Table 4), which summarizes the EC:OC and EC:TC ratios for each fuel type analyzed. Furthermore, we have expanded the discussion in the Conclusion section to emphasize the variability in carbonaceous aerosol composition across different combustion sources and its implications. The modified Conclusion is reported in C3 answer. We hope this addition provides a clearer picture of the carbonaceous aerosol composition and better supports the interpretation of the optical properties presented in the study.

C2. Section 3.1 – Accordingly, with C1, the inclusion of a more thorough description/quantification for EC:OC is warranted.

As mentioned in our response to Comment C1, we have revised Section 3.1 to include a table summarizing the EC:OC and EC:TC ratios for each fuel. In addition, we have reviewed the Section 3 and especially the Conclusion section to make the discussion on the EC:OC ratio more explicit and to better highlight the implications of these findings.

C3. L77 – It was mentioned that a goal of this study/grant was to evaluate and compare different methods of sampling/analyzing carbonaceous aerosols. The paragraph beginning with 208 explains this well and Figure 2 effectively shows the size-selective samplers performed consistently. I'd encourage a similar description in the conclusions to emphasize this finding

We appreciate the Reviewer's suggestion. In the revised Conclusion section, we have included a dedicated paragraph to underscore this important aspect of the study. Now the conclusion was modified as follows:

“The emissions of three different fuels combustion - propane, conventional fossil diesel, and Hydrotreated Vegetable Oil (HVO)- in terms of particle size distribution, optical properties, and EC/OC concentration in the engine exhaust emissions were investigated using an atmospheric simulation chamber (ChAMBRé). The objective of the study was to evaluate and compare different methods of sampling and analyzing carbonaceous aerosols as well as to characterize the variability in the optical

properties of fresh combustion aerosols as a function of fuel type. Soot particles were generated using a mini-inverted soot generator fuelled with propane, as EC dominant soot source, and a diesel engine running on regular diesel and Hydrotreated Vegetable Oil (HVO).

Different types of size-selective samplers, designed to collect different size fractions of particulate matter for monitoring worker exposure, were tested, showing consistent EC concentrations across different fuels combustion (diesel, HVO, propane). These findings are highly relevant for occupational exposure monitoring, as they show that different size-selective samplers, whether targeting inhalable, respirable, or total fractions, consistently capture ultrafine soot particles across various fuels, including “green diesel” alternatives, which are increasingly adopted, especially in the transport and industrial sectors. This is particularly important given the lack of clear regulatory guidance on particle size cut-offs for diesel exhaust sampling in workplace settings. The demonstrated uniformity in EC measurements confirms that current samplers are reliable for assessing soot exposure, regardless of the sampling convention adopted, helping to ensure consistent and representative data for health risk assessments.

The EC/OC ratios of freshly emitted aerosols varied significantly depending on the combustion fuel and process, and this variability appears to be closely linked to changes in the particles' size distribution and optical behavior. EC-dominated soot was found in the propane emission, according to the fuel-lean condition adopted with the MSIG, while OC-richer combustion particles were observed with diesel and HVO combustion performed with the engine. These results were consistent with previous studies and highlighting once again the influence of several factors such as combustion condition (engine temperature, maintenance, efficiency of the combustion process) and fuel composition on these ratios.

Size distribution measurements provided insights into the particle size distributions for different fuels, showing monomodal log-normal distributions with peaks varying according to fuel type and combustion process. As indicated in previous studies, particle size is influenced by factors such as engine type and load, operating conditions, and fuel properties. In this study, fuel-lean propane combustion produced EC-rich particles with larger diameter in the range of 200-300 nm while diesel and HVO combustion generated smaller particles, with a main peak in the accumulation mode, consistent with the higher OC content and the low-temperature engine's idle operation.

The observed variability in EC/OC ratios and particles size is accompanied by changes in the optical properties of the soot particles. Both the absorption coefficient (b_{abs}) and mass absorption coefficient (MAC) varied significantly depending on the type of fuel. Fresh particles generated by regular fossil diesel combustion were found to be more light absorbing than those produced by propane and HVO, exhibiting higher MAC values. The MAC values, measured at different wavelengths (850/870 and 635 nm), ranged from 6.2 ± 0.5 to $9.4 \pm 0.4 \text{ m}^2 \text{ g}^{-1}$ for commercial diesel, from 5.8 ± 0.2 to $8.4 \pm 0.6 \text{ m}^2 \text{ g}^{-1}$ for HVO, and from 5.2 ± 0.5 to $7.8 \pm 1.1 \text{ m}^2 \text{ g}^{-1}$ for propane. The extrapolated MAC at 550 nm turned out to be $7.0 \pm 0.3 \text{ m}^2 \text{ g}^{-1}$ for propane, $9.2 \pm 0.4 \text{ m}^2 \text{ g}^{-1}$ for HVO and $11.2 \pm 0.4 \text{ m}^2 \text{ g}^{-1}$ for diesel. Finally, it should be noted that different optical analyses performed yielded consistent results in nearly all cases.

In conclusion, the findings underscore the importance of considering several factors in the assessment of carbonaceous aerosols emissions and their optical behavior. The type of emission source (e.g. engine type), the chemical composition of the fuel, and the specific combustion condition (e.g. temperature, efficiency) influence the optical properties of the emitted particles. In particular, the variability of the mass absorption coefficient under different combustion scenarios highlights the importance of a deep characterization of such aspects.

Furthermore, instruments based on the measurements of optical properties, due to their ability to provide continuous and real-time data, represent one of the most promising techniques for monitoring carbonaceous aerosols in both ambient air and workplace environments. However, the significant differences in aerosol optical properties across combustion processes require an accurate source

characterization in order to apply the most appropriate MAC values when interpreting data from optical instruments.

In this context, the presented results provide a valuable framework for describing the diversity of fresh soot emissions from different fuels.”

Minor/Technical Comments

C4. Throughout the manuscript there are odd paragraph breaks that produce a confusing structure when reading. Specific examples are listed below.

C5. Currently, the title does not accurately summarize the contents. Maybe: “Measurement Report: Investigation of the Optical Properties of Carbonaceous Aerosols Produced by Diesel Fuel Combustion in an Atmospheric Simulation Chamber”.

We thank the Reviewer for the suggestion. We have modified the Title in:

“Measurement Report: Investigation of Optical Properties of Carbonaceous Aerosols from the Combustion of Different Fuels by an Atmospheric Simulation Chamber.”

C6. L29 – The use of Indeed would better indicate agreement with the previous statement instead of Actually

Thanks, done.

C7. L30 – Replace clime with climate

Thanks, done.

C8. L35 – The use of Additionally would be a better adverb to include additional information to the previous statement.

Thanks, done.

C9. L79 – The use of alternatively here is confusing.

We have reformulated the sentence as: *“Independent experiments were conducted inside an atmospheric simulation chamber (ASC) connected first to a soot generator and then a commercial diesel engine running on regular diesel and Hydrotreated Vegetable Oil (HVO).”*

C10. L83 – Replace physic-chemical with physicochemical

Thanks, done.

C11. L88-L94 – I would move the citations of the full description of the chamber to the end of the brief description included.

Thanks, done.

C12. L96-L97 – I would try listing “...online and offline gaseous composition and aerosol concentration and properties inside the volume:” differently so the use of and isn’t repeated. Maybe “...online and offline gaseous composition in addition to aerosol concentration and properties inside the volume:”

Thanks, done.

C13. L110-L112 – Confusing use of alternatively. Please reword this sentence to describe the two combustion techniques and their associated fuel type.

Thanks, done

C14. L127-L128 – Paragraph break here is odd

Thanks, done

C15. L128 – Move this sentence to the end of the paragraph ending on L120 so the description of MISG is condensed.

Thanks, done

C16. Sect. 2.2 – All sentences within this section should be one paragraph.

Thanks, done

C17. L159 – Rather than within the results, the method to calculate MAC value would be better placed in the methods.

We thank the Reviewer for this suggestion. We have created a new subsection within the Materials and Methods section, titled “Retrieval of aerosol mass absorption cross section” (Section 2.5), where we have relocated the description of the MAC calculation.

C18. Table 3 – Change Particles in the third column header to Particle

Thanks, done

C19. L209 – “... showed comparable EC concentrations.”

Thanks, done.

C20. Figure 2 – Some x-axis labels overlap.

Thank you, Figure 2 has been revised accordingly.

C21. L237 – The description of data acquisition for the SMPS should be in methods not results.

We thank the Reviewer for the observation. In this case, the sentence in question introduces Figure 3 and contextualizes the data shown. For this reason, we preferred to leave it here.

C22. L246 – “...increased (Vernocchi et al., 2022 and references therein).”

Thanks, done

C23. L254 – Similar to C22 references should replace reference

Thanks, done.

C24. Figure 3 – Specify the fits are for a monomodal log-normal size distribution.

Thank you, done.

C25. L324 – Explicitly list the factors of importance

Thanks for the comment; we have reformulated the paragraph as follows:

In conclusion, the findings underscore the importance of considering several factors in the assessment of carbonaceous aerosols emissions and their optical behavior. The type of emission source (e.g. engine type), the chemical composition of the fuel, and the specific combustion condition (e.g. temperature, efficiency) influence the optical properties of the emitted particles. In particular, the variability of the mass absorption coefficient under different combustion scenarios highlights the importance of a deep characterization of such aspects.

C26. L237 – This sentence could be improved with some restructuring. Needed to read it a couple times to understand it.

We reformulated the sentence as follows:

“The data acquisition started 3 minutes after the end of injection of combustion aerosols; the data reported here are the average of the 4 consecutive minutes time interval”.