

## Anonymous Referee #1, 17 Apr 2025

The study presented here is a valuable contribution to the aerosol community. It provides optical properties, more specifically mass absorption cross section values, for three different soot-like aerosol samples. Two of the samples are based on the combustion of liquid fuels burned in a real diesel engine. The manuscript is well written and the data are well presented. I would suggest publishing it after addressing the following comments:

### Scientific comments

- Provide information on the calibration of the various instruments, especially the PAX.

The SMPS and Giano BC1 instruments undergo factory calibration on an annual basis. The PAX was calibrated following the procedures outlined in the user manual, immediately prior to the start of the experimental campaign. The personal samplers were calibrated using a certified flowmeter before each experiment to verify and adjust the sampling flow, in accordance with the NIOSH Manual of Analytical Methods (NMAM), 5th Edition.

We believe that, while important, these technical details are not essential to include in the main manuscript.

- The EC:TC ratio is quite different for the different samples. The larger amount of OC in some samples could affect the MAC amid coating. However, the authors do not address this issue. Please comment on this.

We thank the Reviewer for this observation. We have addressed this point in the revised manuscript at line 335, where we added the following sentences:

*“MAC parameter values are higher for diesel, indicating more absorbent particulate matter, while HVO and propane show lower MAC values, even above 20%. This behavior is consistent with the EC:TC ratios shown in Table 4: the presence of OC coating soot particles enhances light absorption through the lensing effect (Bond et al., 2006; Lack et al. 2010; Lefevre et al., 2018).”*

We added the relative references in the corresponding section.

*Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, J. Geophys. Res. Atmospheres, 111, 2006JD007315, <https://doi.org/10.1029/2006JD007315>, 2006.*

*Lack, D. A. and Cappa, C. D.: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, Atmospheric Chem. Phys., 10, 4207–4220, <https://doi.org/10.5194/acp10-4207-2010>, 2010.*

*Lefevre, G., Yon, J., Liu, F., and Coppalle, A.: Spectrally resolved light extinction enhancement of coated soot particles, 955 Atmos. Environ., 186, 89–101, <https://doi.org/10.1016/j.atmosenv.2018.05.029>, 2018*

- It has been shown that MISG soot has an average diameter between 200-300 nm. The very different particle diameter compared to the other two samples will strongly affect the MAC values, making these results not comparable. In addition, the large size of MISG-generated soot particles is not representative of engine exhaust soot. Is the use of the MISG still justified?

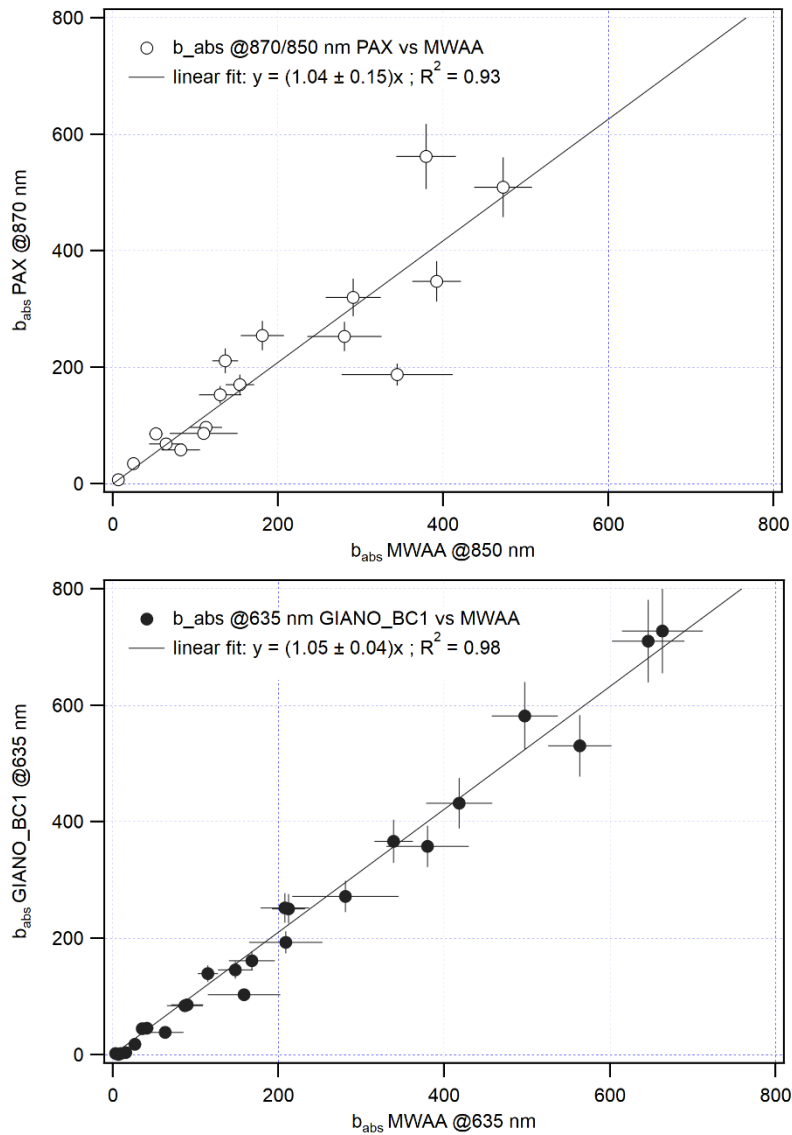
The choice to include the MISG in this study was motivated by its ability to generate almost pure elemental carbon (EC) particles. Inverted-flame burners are widely regarded as ideal soot sources due to their capability to operate under controlled fuel-lean conditions and to produce mature, EC-dominated soot. We adopted this configuration using MISG as a reference source.

To clarify this point, we have added the following sentences to the manuscript:

- Line 121: “The MISG is an inverted-flame burner often considered an ideal soot source due to its capacity to generate almost pure EC particles (Stipe et al., 2005; Moallemi et al., 2019, and references therein).”
- Line 126: “In this study, the MISG, considered a reference EC-dominated soot source, was fueled with propane at a fixed air-to-fuel ratio, following Vernocchi et al. (2022).”

- It would be interesting to see how the different absorption coefficient measurement techniques compare in terms of  $b_{\text{abs}}$ .

We thank the Reviewer for the suggestion. To address this point, we report here the correlation analysis between the  $b_{\text{abs}}$  values obtained from the different instruments used in the study.



The results show good agreement between techniques, with correlation coefficient ( $R^2$ ) above 0.9 and slopes close to unity (within uncertainty). These findings are also in line with previous works, such as Vernocchi et al., 2022 and Caponi et al., 2022, where similar comparisons yielded comparable results, as also referenced in the manuscript.

To keep the main text focused, we decided to not include the correlation plots in the manuscript, but we would be happy to provide them as supplementary material if the reviewer or editor considers it useful.

- In Table 4, it would be valuable to include the MAC values interpolated to 550 nm in addition to those already shown. This would make it easier for the reader to make comparisons with literature values.

We thank the Reviewer for the valuable suggestion. Considering also the related comment below regarding the absorption Ångström exponents, we have added the MAC values extrapolated to 550 nm in Table 4 (now Table 5). These values were calculated using the AAE derived from MWAA measurements, as described in the newly added text at line 308.

- What is the wavelength in Figure 4? Please add it to the axis labels.

The wavelength is 635 nm as reported in the figure caption. We have now added this information in the y-axis label for clarity.

- I would recommend showing the absorption angstrom exponents measured by the MWAA.

As mentioned in our response above, we have now included the absorption Ångström exponents measured by the MWAA. The following sentences have been added at line 308:

*“An average Ångström absorption exponent (AAE; Moosmüller et al., 2011) was calculated for each fuel by aggregating all available  $b_{abs}$  datasets from the MWAA analysis (Table 5). To facilitate comparison with literature data, the MAC value at 550 nm was extrapolated from the MWAA measurement using the relation  $MAC_{550}=MAC_{635} (635/550)^{AAE}$ . All the measured MAC values are summarized in Table 6.”*

- The conclusion section is weak. There is no discussion of the implications of the MAC values found in the study. Please improve.

We thank the Reviewer for this important observation. Since all reviewers suggested to improve the conclusion section at different point, we have revised and expanded it to better summarize the key findings of the study and to discuss their broader implications. The Conclusion Section is now rewritten 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-rich 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  $6.2 \pm 0.5$  to  $9.4 \pm 0.4 \text{ m}^2 \text{ g}^{-1}$  for commercial diesel, from  $5.8 \pm 0.2$  to  $8.4 \pm 0.6 \text{ m}^2 \text{ g}^{-1}$  for HVO, and from  $5.2 \pm 0.5$  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."*

- "Correction factors" are mentioned on page 16. Please elaborate. What corrections are referenced?

Thank you for the comment. We agree that the term "correction factors" was unclear and potentially misleading. We have reformulated the concept as follow:

*"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."*

**Typos**

- Page 11, second paragraph: MISG is misspelled.

Word corrected.