Comment on "**Spectral optical properties of soot: laboratory investigation of propane flame particles and their link to composition**" by Johannes Heuser, Claudia Di Biagio, Jerome Yon, Mathieu Cazaunau, Antonin Bergé, Edouard Pangui, Marco Zanatta, Laura Renzi, Angela Marinoni, Satoshi Inomata, Chenjie Yu, Vera Bernardoni, Servanne Chevaillier, Daniel Ferry, Paolo Laj, Michel Maillé, Dario Massabò, Federico Mazzei, Gael Noyalet, Hiroshi Tanimoto, Brice Temime-Roussel, Roberta Vecchi, Virginia Vernocchi, Paola Formenti, Bénédicte Picquet-Varrault, and Jean-François Doussin

General comments:
The manuscript has in my opinion been substantially improved and is now much easier to follow and more clear. The authors have made significant improvements in clarifying the scope and limitation of their work to a satisfactory level which warrants publication in the journal. However, I have requested minor correction to the revised manuscript, since some of the criticism has not been properly addressed. I realize that this might be in part due to me not putting my statements as clearly and bluntly as needed. Below I raise my concerns of the remaining issue with the revised manuscript.

Original referee comment: "P24 Figure 7: I think panels (a) and (b) in Figure 7 are very informative and a great addition to literature and potentially useful for modelers. Having said that, I don't see the point of panels (c) and (d). The SAE and MSC are mainly a function of particle size and has little to do with the OC/EC ratio. Surely the OC/EC ratio will change the refractive index (and density) of the particles and thus also impact the SAE and MSC, but SAE will still be dictated by the size of the particles (and MSC by the density and size of the particles).
P25 L697-P26 L698: "the SAE suggest a potential similar relation between composition and spectral dependence of scattering as observed for the absorption" This is because the particle size changed between the experiments CS1-CS5 (CMD in Table 1). The spectral dependence of scattering (please use SAE) depends primarily on particle size and not the OC/EC ratio."

Authors' reply: In regards to the previous two points: As mentioned above, during the experiments constant particle growth is observed due to coagulation. This growth can be found to result in a consistent increasedecrease in SAE, that based on precision and uncertainty cannot be however considered significant. It can be noted that, independently of the CMD, the SAE of the aerosols differs systematically between the CSs. i.e. if the soot aerosol populations are similar in size the SAE remains different and thus the main cause for difference in the SAE were associated to the 9 refractive index and composition of the particles. Therefore, we prefer to keep the panels c) and d) in Figure 7 (new Figure 6) to provide analysis of the possible relationship between MSC and SAE with CS point/composition so that the analysis is complete.

Original referee comment: P26 L710: MSC is primarily a function of particle size and particle density, and not EC/TC content.
Authors' reply: Considering the precision of measurements, it is not possible to evidence any differences in the MSC between the five CS, even for the much smaller CS5. As a matter of fact, our experiments do not allow to put in evidence significant differences in the MSC related to the size and chemical composition between the different CS points. Considering the limited amount of data in the literature as well, we conclude this aspect could deserve more investigation.

Referee comment on the revised manuscript:
The listed discussion cited above does not address the concerns I raised to a satisfactory level and my opinion on panels c and d (new Figure 6) remains. The authors have tried to justify keeping them in their reply, but the reply is not convincing, nor has it lead to any significant change in the manuscript to highlight that SAE depends to a much greater extent on particle size than the OC/EC ratio, as one would be made to believe based on Figure 6c in the revised manuscript.

On p. 29 L 791 in the revised manuscript it reads: "*The lack of literature data for these values however did not allow to acquire enough information for any significant synthesis. However, while the MSC data do not show a clear dependence, the plot for the SAE suggests a potential similar relation between composition and spectral dependence of the scattering, as observed for the absorption.*"
Again, The SAE is mainly a function of particle size and has little to do with the OC/EC ratio, other than that the OC/EC ratio explains under which miniCAST conditions the aerosols were produced

at. To put it bluntly, plotting OC/EC ratio with SAE without considering the size of the generated particles is misleading. Especially with the quoted statement above that claims that there is a similar relationship between “composition and spectral dependence of scattering, as observed for the absorption”. This is bluntly put not true. Quoting the Hinds (1999, p. 370) textbook “Light scattering provides an extremely sensitive tool for the measurement of aerosol concentration and particle size.”. This can be seen in Figure 1, contrary to what the authors argue in their reply, where the SAE drops as the particles grow through coagulation in the chamber. For atmospheric aerosols, the SAE is in the range of 0 (coarse mode aerosols) to 4 (nucleation mode aerosols & air molecules i.e. Rayleigh scattering regime).

Moreover, light absorption and light scattering are physically two different processes, the latter being, as mentioned, primarily a function of particle size. This is stated in any textbook on the subject and should not be confused with the physical process of light absorption. Furthermore, making a parametrization for modelers for MSC and SAE using the OC/EC ratio can make readers not familiar with aerosol optics adopt the scheme. The results are specific for the miniCAST burner. In their reply, the authors state that

“*It can be noted that, independently of the CMD, the SAE of the aerosols differs systematically between the CSs. i.e. if the soot aerosol populations are similar in size the SAE remains different and thus the main cause for difference in the SAE were associated to the 9 refractive index and composition of the particles.*”

Where in the manuscript is it shown that the CMD and SAE are independent of each other for different CSs? I suggest removing panels c and d from Figure 6 in the revised manuscript.
If the authors are very keen to keep them, move the panels c and d of Figure 6 to the appendix.

AUTHORS REPLY:

First, we want to thanks the reviewer for having provided further comments to ameliorate the manuscript. The authors agree that size is the key property for scattering behaviour of aerosols as it is determined by the underlying physics of aerosol light interaction, as it is well established by the reviewer. We acknowledge that the provided plots (Fig. 7 panels c and d) could be misleading as the proposal of a generalised relation for soot could lead to a neglect of the key property of particle size when modelling the scattering properties. We therefore accept the reviewer’s suggestion and we move panels c) and d) of Figure 7 in the Supplementary Information.

We additionally modified the text in some specific parts to reflect these changes, as clearly identified in the tracked manuscript. Main changed paragraphs are:

Abstract: “In this study, soot aerosols with varying maturity and composition, i.e. elemental-to-total carbon ratio (EC/TC), have been studied systematically in a large simulation chamber to determine their mass absorption, scattering, and extinction cross sections (MAC, MSC, MEC), single scattering albedo (SSA), and Absorption and Scattering Ångström Exponents (AAE, SAE). The MAC, MEC, SSA and AAE show a variability between the different soot with varying EC/TC ratios.”

Paragraph of Sect. 5.2 has been modified as; “An alternative plot relating the MAC and AAE to the also commonly used EC/OC-ratio can be found in the Supplement (Fig. S4). For completeness, also plots for the MSC and the SAE vs EC/TC and EC/OC are shown in Fig. S5 and S6, despite scattering depends primarily on size and relation to composition has limited significance.”

Conclusions: “The dependence of the MAC, MSC, MEC, and SSA for the five generated soot aerosols and their spectral variability, represented by the AAE and SAE, on the soot composition were analysed. While the MSC appears independent of the soot type (0.4-0.5 m^2g^{-1} at 550 nm), all other parameters show a variability associated with the soot and its composition. “

Besides changes based on the reviewer’s feedback, minor changes to the text have been made to adjust phrasing and correct spelling. These changes do not alter the content of the manuscript and can be followed in the tracked manuscript.