Response to Reviewer 2's comments

This paper presents optical properties of fresh smoke measured with a 7-wavelength aethalometer (AE33) from various wood burning devices using different fuels. Emissions from a diesel engine are also included. Additional measurements include the smoke OC, EC concentrations, CO and CO2 to determine the MCE (and NOx). A significant amount of data is presented. The results are generally as expected, when the fuel is wet (or when fresh wood is placed on the fire) the combustion is more smoldering, MCE drops and OC/EC and BrC increases. Many of the graphs need more explanation or improved clarity. More discussion on the effect of coatings over BC on the overall AAE is needed. Possible effects of tar balls or dark BrC is never discussed. Overall, the paper is interesting and the detailed analysis adds to the data base on residential wood heating emissions, but improvements could be made.

We thank Reviewer 2's positive comments and constructive suggestions for improving the manuscript. The responses to the comments are addressed in blue text. The line number refers to the lines in the revised manuscript.

In the revised version, we have made updates to the figures to enhance readability. Additionally, we have incorporated more detailed explanations directly within the text, aiming to provide a clearer and more comprehensive understanding of the content. The other explanations regarding the effect of coatings over BC on the overall AAE and the possible effects of tar balls are discussed below within the specific comments.

Specific Comments;

Line 41-42 & 46. What is small scale residential combustion? Is it different than low-T biomass/biofuel combustion? Is it RWC?

In lines 41-42, it was an alternative wording to the residential wood combustion (RWC) and has now been changed to RWC for consistency. In line 46, the term "small-scale combustion" is used instead of RWC in order to encompass also the possible contribution of residential combustion of other solid fuels, such as coal, to European air pollution.

Line 250, what is evaporation heat of water. I think the term is water latent heat of vaporization.

We have revised the terminology "evaporation heat of water" to "latent heat of vaporization of water," as indicated in the updated text (line 252).

It is unfortunate that lambda has two separate meanings in this paper. Why not use a different variable for the air to fuel ratio (line 246)?

We appreciate your acknowledgment and the issue regarding the use of the same symbol (*lambda*, λ) for two different variables has been addressed. In the updated version, the symbol for the air-to-fuel ratio has been modified to *alpha* (α), as reflected in line 247 and equations 7, 8, and 9 (lines 246, 254, and 255).

Line 260, what is the meaning of the ' in the denominator in equation 12?

This was a comma (,) which is now removed from equation 12 in the updated version (line 260).

Line 178 and line 312 and table S4, are the particle size distribution data for number or mass distributions? Please specify throughout, but it should be stated clearly in line 178 when discussing the electric low pressure impactor.

The manuscript has been updated to specify that the size particle distribution data mentioned in this study refers to number size distributions. This information is now stated in lines 178 and 312, as well as in the list of abbreviations and the Table S4.

Line 332, what exactly is the meaning of indicative? Is the meaning that the magnitude could be biased relative to the real value (whatever that is), but the comparisons between stoves-fuels are ok (ie, like the comparisons shown in Fig 2b)?

Indeed, the term "indicative" (line 333) in this context emphasizes that the MAC_{880nm} presented in this manuscript should not be directly compared with those from other studies utilizing different instrumentation. The variations in measurement techniques and devices require caution in making such direct comparisons. However, the comparability within the manuscript assessing stoves and fuels remains valid, as these comparisons are internally consistent, relying on uniform methods and instruments (aethalometer AE33 measurements) across the specified experiments.

Could the variability in MAC880 also be due to the presence of tar balls or dark BrC which does absorb at the highest wavelengths? The possible role of these forms of BrC should be discussed.

This is a relevant question, as it has been shown that tar balls are emitted from various sources, including biofuel and biomass combustion (e.g., Pósfai et al., 2004), and that they absorb light in the full spectrum from UV to IR wavelengths (Corbin et al., 2019). However, in the present study, we had no means of estimating the contribution of absorption by tar balls to any wavelengths. We have previously carefully characterized the particle morphology for the emissions from the modern masonry heater (Leskinen et al., 2014) and the modern chimney stove (Leskinen et al., 2023) used in this study using electron microscopy and the APM-SMPS method. There were no indications of tar ball -type morphologies in particulate emissions from either of those devices in similar combustion events as covered by this study. However, it is still possible that tar balls or other “dark-BrC” -like organic matter, influencing light absorption in the highest wavelengths, have existed in some of the studied aerosols, particularly those with high OC/EC.

We have now added the following discussion in the results & discussion (line 348 onwards): “MAC_{880nm} might also be enhanced by the presence of organic matter absorbing light also at the higher wavelengths. Specifically, tar ball like morphologies in biomass combustion particles have been associated with a wide absorption wavelength range (Hoffer et al., 2016). In previous studies, tar ball like morphologies were not observed in the emissions of the wood-fired modern chimney stove (Leskinen et al., 2023) or the masonry heater (Leskinen et al., 2014). However, less efficient wood combustion conditions may promote the formation of tar ball like particles (Pósfai et al., 2004).”

References:

Corbin, J. C., Czech, H., Massabò, D., de Mongeot, F. B., Jakobi, G., Liu, F., Lobo, P., Mennucci, C., Mensah, A. A., Orasche, J., Pieber, S. M., Prévôt, A. S. H., Stengel, B., Tay, L. L., Zanatta, M., Zimmermann, R., El Haddad, I., and Gysel, M.: Infrared-absorbing carbonaceous tar can dominate light absorption by marine-engine exhaust, npj Clim. Atmos. Sci., 2, 10.1038/s41612-019-0069-5, 2019.

Leskinen, J., Ihalainen, M., Torvela, T., Kortelainen, M., Lamberg, H., Tiitta, P., Jakobi, G., Grigonyte, J., Joutsensaari, J., Sippula, O., Tissari, J., Virtanen, A., Zimmermann, R., and Jokiniemi, J.: Effective density and morphology of particles emitted from small-scale combustion of various wood fuels, Environ. Sci. Technol., 48, 13298–13306, 10.1021/es502214a, 2014.

Leskinen, J., Hartikainen, A., Väätäinen, S., Ihalainen, M., Virkkula, A., Mesceriakovas, A., Tiitta, P., Miettinen, M., Lamberg, H., Czech, H., Yli-Pirilä, P., Tissari, J., Jakobi, G., Zimmermann, R., and Sippula, O.: Photochemical Aging Induces Changes in the Effective Densities, Morphologies, and Optical Properties of Combustion Aerosol Particles, Environ. Sci. Technol., 57, 5137–5148, 10.1021/acs.est.2c04151, 2023.

Pósfai, M., Gelencsér, A., Simonics, R., Arató, K., Li, J., Hobbs, P. V., and Buseck, P. R.: Atmospheric tar balls: Particles from biomass and biofuel burning, J. Geophys. Res. Atmos., 109, 10.1029/2003jd004169, 2004.

Line 343, GMD of what, number or mass? Isn't mass distribution what matters here?

We refer to the particle number size distributions. This has now been clarified in the text (lines 178, 312, and 343) and the list of abbreviations. The densities were not measured in the experiments, so mass mean diameters cannot be calculated from the data. A reasonable substitute is the volume mean diameter (VMD). For lognormal size distributions, VMD can be calculated using Hatch-Choate equations from VMD = GMDexp(3$\ln^2$GSD) where GSD is the geometric standard deviation of the number size distribution (Hinds, 1999; Baron and Willeke, 2001). Assuming lognormal number size distributions, this equation can be used for estimating the VMD for each of the experiments, with the GMD and GSD results shown in Table S4.

On line 344 it is stated that no correlation was seen between GMD and MAC. The GSDs of the size distributions are approximately 2.0 ± 0.1 which means that VMD ≈ 4.2GMD. In other words, the mean diameter plainly shifts towards larger sizes, and the correlation between the MAC and VMD would be equally poor for VMD as it is with GMD. So, the statement on line 344 would be the same.

Another point about the reviewer's question was that is it reasonable to look for correlations between MAC and number-based GMD. When modeling light absorption by particles, the absorption coefficient babs is calculated as an integral over a number size distribution or in real, discrete size distributions from the sum: babs = sum(Qa(Dp)*((pi*Dp^2)/4)*dN(Dp)/dDp)dDp where Qa(Dp) is the size-dependent absorption efficiency, Dp the particle diameter and dN(Dp) the number of particles in the size bin dDp. Since the MAC of EC is calculated from MAC = babs/EC, where EC is the mass concentration of EC, it is obvious that the number size distribution is an important property influencing MAC values.

References:

Hinds, W. C.: Aerosol Technology – Properties, Behavior, and Measurement of Airborne Particles, John Wiley & Sons, INC., New York, 1999.

Baron, P. A. and Willeke, K.: Aerosol Measurement – Principles, Techniques, and Applications, John Wiley & Sons, INC., New York, 2001.

Fig 2b is not clear. What is the meaning of the overall bar height? Eg, is the MAC with C=3 the sum of the gray and red bars? Needs more explanation.

In Figure 2b, the vertical bars correspond to the MAC_{880nm}. Specifically, the height of the grey bar represents the MAC_{880nm} calculated using the default values in the aethalometer (default C value), while the height of the orange bar illustrates the MAC_{880nm} when the C value is adjusted to 3. To address potential confusion arising from the overlapping bar plots, we have revised Figure 2b in the updated version. The modified figure now features the MAC_{880nm} with C = 3 depicted as orange dots rather than bars, aiming to enhance clarity in the visual presentation of the data.

In Fig 3a the AAE vs OC/EC data looks more correlate then BrC vs OC/EC, opposite to the regression (r2) results given in the plot.

Acknowledging the identified mistake in the equation presented in Figure 3a, we confirm that this error has been rectified in the updated version of Figure 3a and line 491 (when referring to the R^2 value in Figure 3a). The corrected equation now accurately aligns with the intended representation.

Line 396-399. Is this statement correct, ie does lensing change the AAE to a value different than that of pure BC and is this statement supported by references Virkkula et al and He et al. For example, does a clear coating (could be OA, or other species) on BC result in an AAE different from the AAE of pure BC? If the coating is OA containing chromophores, is the AAE different from pure BC AAE only due to those chromophores absorbing light (the BC core has no effect) or does the coating-BC combination change the overall AAE? This needs clarification, possible by adding more details.

This argument is made in many places, so the question also applies to line 429, line 441 and line 489.

Yes, the statement is correct. The lensing effect indeed does change the AAE values compared to pure BC. This has been shown in several studies, e.g., Gyawali et al. (2009), Lack and Cappa (2010), Lack and Langridge (2013), and more recently by Virkkula (2021) and Luo et al. (2023). Already Gyawali et al. (2009) showed that if the absorption by the coating is wavelength dependent, the AAE is different from the AAE of pure BC, and consequently, this effect is a combination of lensing and absorption by the coating and that these effects vary as a function of core size and coating ratio.

We have added a few more references to support our statement in the paper (line 406).

The new citations:

Gyawali, M., Arnott, W. P., Lewis, K., Moosmüller, H., and Moosmüller, M.: Atmospheric Chemistry and Physics In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and non-absorbing organic coatings on spectral light absorption, Atmos. Chem. Phys, 9, 8007–8015, 2009.

Luo, J., Li, Z., Qiu, J., Zhang, Y., Fan, C., Li, L., Wu, H., Zhou, P., Li, K., and Zhang, Q.: The Simulated Source Apportionment of Light Absorbing Aerosols: Effects of Microphysical Properties of Partially-Coated Black Carbon, J. Geophys. Res. Atmos., 128, 10.1029/2022JD037291, 2023.

Fig 4 is not very useful, maybe better to extract the important information and move it to the supplement.

Acknowledging the suggestion, we recognize that Figure 4 could be more suitably placed in the supplement. Accordingly, in the updated version of the manuscript, Figure 4 has been relocated to Figure S12 in the supplementary material.

Line 458 and 459. This line seems incorrect, or at least the meaning is not clear. The contribution of BrC (ie, b(abs) just due to BrC) at 370 nm is always higher than at 470 nm. The issue here is more related to what wavelength ranges are used for the fit. I believe what is being noted here is that when using the lowest wavelength, the fit produces a lower AAE.

The measured relative contribution of BrC to the absorption at 370 nm was in fact often lower than at 470 nm, as evident in Figure 4 and Table S6. To avoid misinterpretations, we have added the word “relative” on line 464. Our results deviate from the anticipated trend where lower wavelengths would typically exhibit a higher contribution of BrC compared to higher wavelengths. Contrary to this expectation, our observations indicate that the BrC contribution is most pronounced at 470 nm for most experiments. Consequently, the AAE fit over the range of 370-950 nm ($AAE_{370-950}$) using all the seven aethalometer wavelengths tends to be lower compared to the AAE calculated using only the two wavelengths ($AAE_{470/950}$).