Dear Editor,

Please find attached our detailed responses to the reviewers' comments regarding our manuscript titled "Validation of cantilever-enhanced photoacoustic particle size-resolved light absorption measurement using nigrosin reference particles and Mie-modelling." We greatly appreciate the reviewers' constructive feedback, which has significantly helped us improve the clarity and focus of our work.

Below, we have provided the reviewers' comments in black, followed by our responses in red. Updates to the manuscript are presented in italics beneath each comment, with corresponding line numbers from the revised manuscript where track changes are enabled.

We hope that the revised manuscript meets the reviewers' expectations and look forward to your favorable consideration.

Thank you for your time and effort in handling our submission.

On behalf of all authors,

Joel Kuula

Reviewer 1

Kuula et al. show a new setup using the tandem DMA and 3-wavelength Cantilever-enhanced photoacoustic spectrometer to measure size-resolved aerosol light absorption properties. Overall, the manuscript is well-written, and the setup is novel. I have some minor comments to help authors improve their paper. Please see my comments below.

Major comments:

Why do you use NO_2 not cab-o-jet or other standards? How did you calibrate the CEPAS? Your difference between measured and model values at different wavelengths are not consistent, which could be due to the wrong calibration method. (see "Characterization of light-absorbing aerosols from a laboratory combustion source with two different photoacoustic techniques").

The CEPAS was initially calibrated using NO_2 because it was estimated that a gas-phase calibration would provide the most independent and reliable baseline when transitioning to particle-phase measurements. This approach also mitigates uncertainties associated with particle size-dependent deposition losses. However, we acknowledge the limitations of NO_2 calibration, which may contribute to discrepancies between measured and modeled values. Specifically, NO_2 exhibits photodissociation below 450 nm and has a relatively low absorption cross-section beyond 650 nm, as discussed in Yu et al. (2020). Despite these drawbacks, the wavelengths used in the CEPAS—439.5, 516, and 635 nm—were deemed sufficient for this study. We duly note the reviewer's comment about potential particle-phase calibrations, and this is certainly an area for future improvement during subsequent re-calibrations of the CEPAS.

From a practical standpoint, the initial calibration was conducted by comparing the raw CEPAS photoacoustic signal to various dilution ratios of NO_2 with a known standard concentration (1.17 ppm). Dilution was performed using mass flow controllers and compressed air. Four measurement points were recorded, each with a 10-minute integration time.

In response to this comment, we have expanded the manuscript's discussion on CEPAS calibration. It now provides additional details about the calibration procedure and evaluates the strengths and weaknesses of the chosen approach.

Line 135: "Prior to the laboratory experiments conducted in this study, the CEPAS was initially calibrated using a known standard concentration of NO2. In practice, NO2 (concentration of 1.17 ppm) was diluted using mass flow controllers and compressed air, and four different measurement points were then recorded, each with a 10-min integration time. It was estimated that gas-phase calibration provided the most independent and reliable baseline when transitioning to particle-phase measurements. Furthermore, it mitigated uncertainties associated with particle size-dependent deposition losses. In contrast, the NO2-based calibration suffers from drawbacks as well. For example, NO2 exhibits photodissociation below wavelengths of approximately 420 nm and has relatively low absorption cross-section beyond 650 nm. Further details on the calibration of the CEPAS are also discussed extensively in another study by Karhu et al. (2024)."

The specifics of your Cantilever PAS are unclear to me. I suggest a table summarizing your cell length, resonance frequency, quality type, laser power, mean diameter, detection limit, response time, etc. Moreover, it is also unclear to me in L94-95, where you said the

measurement point is approx. 10-20 S. Does that refer to one data point that will 10-20 s to collect?

We have added a new table outlining the different technical details of the CEPAS. This table is presented below.

Regarding the measurement time, the reviewer is correct. Obtaining a single data point takes approximately 10-20 seconds and involves several sequential steps: sample intake, a brief stabilization period, recording of the photoacoustic signal, and flushing the sample from the cell. Unlike many other aerosol instruments, the CEPAS performs measurements only when the acoustic cell is closed, meaning there is no continuous flow through the cell during measurement. The durations of these actions are adjustable, so we provided a typical range (10-20 seconds) rather than a fixed value. To improve clarity, we have revised the manuscript to explain this in detail.

Table 1. Specifications of the CEPAS photoacoustic instrument.

CEPAS technical specification	
Microphone/detector	*Silicon cantilever whose position is measured using interferometer*
Acoustic cell type and dimensions	*Cylindrical; length 90 mm, diameter 4 mm*
Cell block material	*Aluminum with nickel coating*
Cell window material	*Antireflection-coated fused silica planar windows*
Cell window angle	*35° (with respect to laser beam)*
Acoustic operation mode	*Non-resonant*
Operation frequencies	*105, 110, and 125 Hz*
Laser type	*Multimode continuous-wave diode laser (three pieces)*
Laser wavelengths	*439.5, 516, and 635 nm*
Laser powers	*300, 210, and 130 mW*
Laser beam diameter	*< 4 mm*
Detection limit	*0.0014 Mm^{-1}*
Response time	*10-20 s, adjustable*
Data processing method	*Fast Fourier-Transform (FFT)*

Line 123: "The measurement cell is closed during the measurement (no continuous flow through the cell during measurement), which increases the measurement time. In practice, obtaining a single measurement point takes approx. 10 – 20 s and involves several sequential steps: sample intake, a brief stabilization period, recording of the photoacoustic signal, and flushing the sample from the cell. The time duration of these actions is adjustable."

I feel the discrepancy between your measured and model-predicted light absorption properties could be attributed to the calibration method and multiple-charge particles. Please comment on this.

Our initial assessment attributed the discrepancy primarily to the inaccurate sampling loss function and, to a lesser extent, to CEPAS imprecision (i.e., relatively high measurement standard deviation). This conclusion was based on the challenges encountered during the sampling loss characterization process, which forms the focus of our separate study (Grahn and Kuula, 2024; https://doi.org/10.3390/pr12122827). The complexity of accurately defining

the loss function and the lack of unequivocal results are discussed further in the minor comments. Supporting this interpretation is the observation that the initial particle size-dependency analysis revealed a size-dependence, and subsequent adjustments to the loss function improved the comparability between measurements and model predictions.

While the NO_2-based calibration may have contributed to the discrepancy, we believe its impact was relatively minor compared to that of the inaccurate loss function. Studies suggest that photodissociation of NO_2 occurs mainly at wavelengths below 420 nm, with the 398-420 nm range contributing "quite limited" effects (see e.g., Wang et al., 2020, https://doi.org/10.1016/j.atmosenv.2020.117559; Guan et al., 2023, https://doi.org/10.5194/acp-23-10413-2023). As the blue wavelength used in CEPAS was 439.5 nm, the potential contribution from photodissociation is limited. Additionally, the smaller absorption cross-section of NO_2 at the red wavelength likely had a negligible impact, as this wavelength exhibited the best accuracy among the three tested.

Multiple-charge particles are another potential source of discrepancy, but their influence was likely limited due to the use of a pre-impactor, as applied in similar studies. While the second reviewer suggested verifying the presence of multiple-charge particles with a tandem DMA setup, this was not feasible due to the unavailability of a spare DMA.

As the second reviewer pointed out, the refractive indices used for nigrosin may have also contributed to the observed discrepancy. The used indices were originally determined for wavelengths of 450, 532, and 633 nm, which differ from the wavelengths used in the CEPAS by +10.5 nm (+2.39%), +16 nm (+3.10%), and -2 nm (-0.31%) for blue, green, and red, respectively. To address this, we decided to adopt a more sophisticated way of defining the refractive indices used in this study. Essentially, a 2nd degree polynomial fit was applied to the original data, allowing extrapolation and interpolation of the refractive indices for the CEPAS wavelengths. Additionally, as stated in the source material, an uncertainty of ±3 % was also added to the imaginary part of the refractive indices to account for potential inaccuracies in its definition. The imaginary part of the refractive index drives the attenuation of light within the particle and therefore essentially defines the particle's light absorption. The effect of this more sophisticated approach on the re-calculated results was found to be negligible. The updated results are shown in the manuscript.

Several changes:

Line 135: Calibration (same as above).

Line 190 Sampling losses: "The root cause for this offset was estimated to be small flow channels and tube fitting sizes (inner diameter 2 mm), which resulted in significant under-pressure within the system. When sampling through the CEPAS, the CPC indicated a pressure of approximately 0.50 atm compared to 0.90 atm during bypass measurements. Literature suggests that CPCs may undercount particles under low-pressure conditions (Bauer et al., 2023). Since the current photoacoustic cell is a commercial component originally designed and manufactured by Gasera Ltd. for trace gas measurement, redesigning and optimizing the flow channels for aerosol-phase measurements was beyond the scope and resources of this study. While the hypothesis of low pressure as the root cause of the transmission offset remains to be fully validated, the study's results are considered useful and reliable within its context. The sub-optimal flow channels will be addressed in the future work."

Line 231 Refractive indices: "Notably, Drinovec et al. provided values for wavelengths of 450, 532, and 633 nm, which differ slightly from the CEPAS laser wavelengths of 439.5, 516, and 635 nm. To account for this, a 2nd degree polynomial fit was applied to the original data, allowing extrapolation and interpolation of the refractive indices for the CEPAS wavelengths. Additionally, as stated in the source material, an uncertainty of ± 3 % was also added to the imaginary part of the refractive indices to account for potential inaccuracies in its definition. The imaginary part of the refractive index drives the attenuation of light within the particle and therefore essentially defines the particle's light absorption. Nigrosin is often used in light absorption instrument testing as there is literature available describing its optical properties and because it forms spherically shaped particles when aerosolized and dried (Drinovec et al., 2022; Lack et al., 2006). Particle sphericity is an assumption of Mie theory."

Results section updated with new figures and re-calculated correlation and accuracy metrics.

Minor comments:

L126-127, "For a full … checks)." It is not clear what the 12 steps mean here. Are you referring to 12 different particle sizes? If so, could you provide these sizes?

Yes, the 12 steps here refer to the 12 different particle size bins measured. These sizes were 30.0, 38.0, 48.0, 60.8, 76.9, 97.4, 123, 156, 197, 250, 316, and 400 nm. We have added this information to the manuscript Table 2.

Line 168: "For a full scan comprising 12 steps (i.e., 12 different particle sizes, see Table 2 for the exact particle sizes), the total measurement duration was 4 minutes and 40 seconds (12 measurements and two zero-background checks)."

L144-145, "The observed … modeling results." This is not very clear to me. Typically, measurements is more reliable than models. And what artifact are you referring to here? Does that mean your measurements are not reliable at all?

Our interpretation of the experimental results is that, while useful and largely consistent with the modeling results, there appeared to be a constant offset or bias we could not fully resolve. We estimated that this artifact was most likely caused by the significant under-pressure induced by the small flow channels and fittings in the CEPAS system. Literature indicates that CPCs may undercount particles when exposed to such conditions (see Bauer et al., 2023; https://doi.org/10.5194/amt-16-4445-2023). Despite this offset, we believe the measurements remain reliable within the study's context.

Line 190 Sampling losses (same as above).

L145-147, "The root … the CEPAS." Why did you use this small tubing? I do not quite understand why smaller tubing is the reason for that. Your CEPAS should always be under pressure since it is connected to a pump. Did you measure the pressure? And if you think this is the reason for the artifact, why didn't you fix that?

The small tubing was necessitated by the thread sizes of the photoacoustic cell, requiring correspondingly small fittings. This design caused a significant pressure-drop across the

CEPAS sampling system compared to typical aerosol instruments. During our loss characterization study (a separate manuscript currently under review), we measured pressure levels at the CPC under different conditions. The pressure was approximately 0.50 atm when sampling through the CEPAS and 0.90 atm when bypassing it.

Evidently, addressing this issue would require redesigning and fabricating a new photoacoustic cell with flow channels better optimized for aerosol-phase measurements. Unfortunately, this was beyond the scope and resources of this study. The current photoacoustic cell is a commercial component originally designed and manufactured by Gasera Ltd. for trace gas measurements.

To clarify this point, we have revised the manuscript to better explain the sampling losses and their implications.

Line 190 Sampling losses (same as above).

L182-183, "The reference … 1:10." What's the RH after the dryer? RH can affect the PAS measurements.

It is true that water content in particles may affect the photoacoustic signal. In our case, the relative humidity was within 10-30 % (measured at the DMA) throughout the measurements. We have added this information to the manuscript.

Line 238: "The reference aerosol generated with a model ATM 226 aerosol generator (Topas GmbH., Germany) was first dried with a silica gel dryer (relative humidity measured at the DMA was 10-30 % throughout the measurements) and subsequently diluted with varying ratios of 1:5, 1:7.5, and 1:10."

Table 1, what are the references for nigrosine refractive index.

The original study, from which the refractive indices of nigrosine for different wavelengths were retrieved from, was by Drinovec et al. (2022). The full citation is shown below. This citation is also displayed in the Table caption.

Drinovec, L., Jagodič, U., Pirker, L., Škarabot, M., Kurtjak, M., Vidović, K., Ferrero, L., Visser, B., Röhrbein, J., Weingartner, E., Kalbermatter, D. M., Vasilatou, K., Bühlmann, T., Pascale, C., Müller, T., Wiedensohler, A. and Močnik, G.: A dual-wavelength photothermal aerosol absorption monitor: design, calibration and performance, Atmos. Meas. Tech., 15(12), 3805–3825, doi:10.5194/amt-15-3805-2022, 2022.

L209-211, "With respect … Mie-modelled values." It is unclear to me why you observed that inconsistent discrepancy. I would like to see your explanation.

After considering and addressing – to the best of our ability – the impacts of calibration method, sampling losses, multiple-charge particles, and uncertainty of refractive indices on discrepancies, we do not have a clear explanation. To take some action, systematically investigating and experimenting with different calibration approaches would probably be worthwhile.

We have added a general note about the unresolved discrepancies to the manuscript.

Line 406: "Furthermore, despite calibrating the CEPAS prior to the experiments, the measured absolute absorption levels for the blue and green wavelengths deviated from that of the Mie-predicted values. Therefore, re-calibration is necessary before deploying the system to outdoor measurements, for example."

Reviewer 2

The paper describes the performance of a three-wavelength cantilever-based photoacoustic spectrometer by comparing absorbing particles from the nebulization of nigrosine ink with Mie calculations. The cantilever technique has great potential, and I think it holds great promise for the future, for that reason I loosely followed the development of this new technology over the past years with interest, and I was excited to have to review the paper. However, I found the paper to be misleading on some key aspects (as discussed below) as well as very dismissive with regard to extensive work done in this field in the past and fully available in the literature. It should not be too difficult to correct these issues, but before the paper can be accepted, a serious effort must be made to address these problems.

General Comments

- The introduction and motivation of the work focus mostly on the aging, citing, and absorption enhancement of black carbon particles. However, the paper presents nothing on any of these topics. The paper shows measurements on nigrosine (not black carbon), spherical (not aggregates), and not coated. I do not doubt that the technique can be applied to study the aging, coating, and absorption enhancement of black carbon in the future. Still, the motivations and introduction provided in this work do not match what has actually been done here. The abstract, introduction, and conclusion should make this point clear.

We agree that there is a mismatch between the broader context and future applications discussed in the introduction and the specific scope of the current study. The focus of this paper is indeed on developing and validating a new methodology using nigrosine particles as a model system, rather than directly addressing black carbon aging, coating, or absorption enhancement.

To address this, we have revised the abstract, introduction, and conclusion to clarify that while black carbon aging and coating are key applications motivating the broader development of this methodology, the present study focuses on nigrosine as a simplified test case for demonstrating the validity and feasibility of the approach. We now explicitly state that black carbon-related investigations remain a future application and that this work represents a critical first step toward such studies.

Changes to the Abstract, Introduction, and Conclusions. (To save space and maintain readability, we have not copied these longer sections here. Please refer to the track changes version of the manuscript for the detailed revisions.)

- The paper, including the introduction and especially Section 3.3, almost completely ignores (with just a couple of exceptions) many other previous photoacoustic experiments (some more than a decade old) that have been carried out in the past including with size selection discrimination and at multiple wavelengths. For example, the work done by Lack et al., Cross et al., Schnaiter et al., Arnott et al., Sharma et al., Smith et al., just to mention a few groups (but more are out there). Therefore, statements like those in lines 304-306 are certainly incorrect. A web search will return a few studies done in the past using photoacoustic even with size selection. In addition, the paper (and especially section 3.3) fails to recognize other types of measurements performed in this field, for example, with the extinction minus scattering

approach, or using photothermal interferometry, which even if with their own challenges, are an important contribution to the field as well.

We thank the reviewer for highlighting the omission of several important studies and for pointing out areas where the discussion in Section 3.3 and elsewhere can be improved. To address this, we have extended the literature review and added more citations especially to those studies presented in the comment. We have removed the statement in line 304-306 to avoid confusion. The initial intention of the statement, which was poorly worded, was to point out that we are not aware that there is a similar, fully integrated and independently operated size-resolved absorption measurement instrument developed. It is certainly the case that size-resolved measurements, where a DMA or similar size classifier is placed upstream in the sampling line and then a fleet of various instruments, including photoacoustic spectrometers or filter photometers, are connected to it afterwards, have been performed in different studies previously. We have also included discussion about photothermal interferometry and extinction-minus-scattering methods for measuring light absorption. While these are not the primary focus of our study, we have added a brief acknowledgment of their contributions to aerosol absorption measurements and the challenges they address, as well as a few references to studies in these areas.

Section 3.3.

Currently, the most technologically advanced and commercially available devices for measuring size-resolved light-absorbing particles include the Single Particle Soot Photometer (SP2, Droplet Measurement Technologies LLC., USA) and the Soot Particle Aerosol Mass Spectrometer (SP-AMS, Aerodyne Research Inc., USA). These instruments have proven vital for aerosol research, particularly in the study of carbonaceous particles (e.g. Chen et al., 2018; Liu et al., 2019; Schwarz et al., 2006). The SP2 measures the number and mass size distributions of refractory black carbon (i.e., vaporizes only at very high temperatures) and its coating thickness using techniques based on light scattering and incandescence (Baumgardner et al., 2004; Stephens et al., 2003). The SP-AMS, on the other hand, measures the size-resolved chemical composition of particles using mass spectrometry and ion concentration quantification (Onasch et al., 2012). Despite the utility of the SP2 and SP-AMS, they do not explicitly measure particle light absorption, which is an essential aerosol parameter when considering the impact of light-absorbing particles on climate.

More conventional light absorption measurement instruments have been employed for size-resolved light absorption measurements in various ways. For example, a method involving sample collection using a multi-stage impactor followed by optical or chemical analysis (e.g., measurement of transmission or methanol/water extraction and spectrophotometer measurement) has been utilized in multiple studies (Feng et al., 2023; Gao et al., 2015; Horvath, 1995; Lei et al., 2018; Liu et al., 2013; Wu et al., 2020). Although this is a valid method, it suffers from poor temporal resolution (typically >1-hour) and does not produce real-time data. Moreover, depositing particles on a filter may cause morphological changes and thus alter their optical properties, as discussed in the introduction. A more autonomous method, similar to the DMA-CEPAS, is to couple a particle sizer such as a DMA or an Aerodynamic Aerosol Classifier (AAC) with a filter photometer. This approach has been employed in several studies using various instrument configurations (Baxla et al., 2009; Ning et al., 2013; Stabile et al., 2012; Tunved et al., 2021; Zhao et al., 2019, 2022). The challenges associated with the filter photometers are mostly the same as those of the manual sample collection: the deposition of particles on a filter as well as the temporal resolution arising from the sensitivity and speed of

the measurement. Comparatively, similar studies where a photoacoustic spectrometer has been used instead of a filter photometer are also available (Chakrabarty et al., 2007; Forestieri et al., 2018; Slowik et al., 2007a, 2007b). The focus of these studies has been in the investigation of particle ageing, coating, and absorption enhancement in a laboratory setting. To the degree that can be interpreted from the experimental descriptions, the DMA (used in these cases) has not been integrated with the spectrometer with respect to instrument control and data processing. Although this deviates from the DMA-CEPAS design, the end result is practically the same. The EMS method discussed in the introduction has also been used in particle size-resolved absorption measurements (Khalizov et al., 2009). With respect to fully integrated devices, a photoacoustic instrument capable of indicative size-selectiveness was recently developed by Ajtai et al. (2023). The size-selection is based on the measurement of phase shift between the modulation of the light and the resulting photoacoustic signal. The key benefit compared to other discussed systems is that the size-selection is essentially performed within the domain of the photoacoustic instrument itself; it does not rely on "external" means of classifying particle sizes and, therefore, many of its design and operation characteristics remain fairly simple (e.g. no data inversion required). However, until further improvements are made, the size-resolving power of this method remains limited in comparison traditional DMAs or AACs.

To date, there appears to be no established method for the measurement of particle size-resolved light absorption. In comparison to the previous implementations, the main advantages of the DMA-CEPAS are its high level of instrument integration, sensitivity as well as the aerosol-phased measurement. As noted in the introduction, Karhu et al. (2021) demonstrated noise equivalent absorption coefficient (1σ) of the CEPAS to be 0.013 Mm^{-1} (= 1.3 × 10^{-10} cm^{-1}) in 20 s integration time. Nevertheless, the DMA-CEPAS requires more development and testing. For example, its adaptation to field measurements may require re-configurations in system running parameters, although the standalone version of the CEPAS without the DMA has been used successfully in the field (Karhu et al., 2024). Additionally, re-visiting the instrument calibration using both gas- and particle-phase reference would be beneficial.

- Finally, I have some concerns with the "correction" scheme developed and presented in the paper, as discussed in the specific comments.

Specific Comments

- Section 2 should report more details on the specification of the lasers, in particular, the power and the operation mode (CW or pulsed)

- Also, section 2 should clarify if the cylinder where the sample is analyzed is acoustically resonant or not.

- What material is the cylinder made of?

- Provide specifications about the windows as well as the lens.

We have combined the response to these first four specific comments together as they revolve around the same subject.

In conjunction with the comment made by the first reviewer, we have added a new table to the manuscript, which outlines the technical details of CEPAS photoacoustic instrument. The new table is shown below.

Table 1. Specifications of the CEPAS photoacoustic instrument.

CEPAS technical specification	
Microphone/detector	*Silicon cantilever whose position is measured using interferometer*
Acoustic cell type and dimensions	*Cylindrical; length 90 mm, diameter 4 mm*
Cell block material	*Aluminum with nickel coating*
Cell window material	*Antireflection-coated fused silica planar windows*
Cell window angle	*35° (with respect to laser beam)*
Acoustic operation mode	*Non-resonant*
Operation frequencies	*105, 110, and 125 Hz*
Laser type	*Multimode continuous-wave diode laser (three pieces)*
Laser wavelengths	*439.5, 516, and 635 nm*
Laser powers	*300, 210, and 130 mW*
Laser beam diameter	*< 4 mm*
Detection limit	*0.0014 Mm^{-1}*
Response time	*10-20 s, adjustable*
Data processing method	*Fast Fourier-Transform (FFT)*

- What's the rationale for the specific choice of multiplexing frequencies, where they optimized somehow?

The frequencies were chosen such that no overlap with external noise sources were present. Noises typically result from mechanical vibrations such as nearby pumps. We have added a note about this to the manuscript.

Line 130: "The laser source (RGB laser module by Opt Lasers, Tomorrow's System Sp. z o.o., Poland) uses three different wavelengths (measured at 439.5, 516, and 635 nm) and they are multiplexed at modulation frequencies of 105, 115, and 125 Hz. These frequencies were chosen to avoid spectral overlap with external acoustic noise and mechanical vibrations, such as those caused by nearby gas pumps. The different technical details of the CEPAS are summarised in Table 1."

- Some more detail on the NO2 calibration procedure would be useful. For example, was photodissociation being accounted for?

This is something that the first reviewer suggested as well. To address this, we have extended the discussion about calibration, and it now provides additional details about the calibration procedure and evaluates the strengths and weaknesses (incl. photodissociation) of the chosen approach.

Line 135: "Prior to the laboratory experiments conducted in this study, the CEPAS was initially calibrated using a known standard concentration of NO2. In practice, NO2 (concentration of 1.17 ppm) was diluted using mass flow controllers and compressed air, and four different measurement points were then recorded, each with a 10-min integration time. It was estimated that gas-phase calibration provided the most independent and reliable baseline when

transitioning to particle-phase measurements. Furthermore, it mitigated uncertainties associated with particle size-dependent deposition losses. In contrast, the NO2-based calibration suffers from drawbacks as well. For example, NO2 exhibits photodissociation below wavelengths of approximately 420 nm and has relatively low absorption cross-section beyond 650 nm. Further details on the calibration of the CEPAS are also discussed extensively in another study by Karhu et al. (2024).”

- Deposition losses were calculated assuming the particles are spherical. That’s reasonable, but whether that is the case or not it should be made clear.

We have added this notion to the manuscript.

Line 201: “The particles were assumed to be spherical, although their morphology was not explicitly verified.”

- Is the LabView code for the DMA going to be made openly available?

All data, including the LabVIEW code for the DMA, are available upon request.

- It would have been useful to check the size selected distribution with an additional sizer downstream of the DMA system to verify that indeed the effect of multiple charges was minimized.

We agree, but we did not have access to a spare DMA.

- A sensitivity of the Mie calculations to the assumed index of refraction would have also been valuable. Could it be that the particle-phase index of refraction differs enough in the experiments carried out here from that found in the literature to explain at least part of the discrepancies?

We decided to adopt a more sophisticated way of defining the refractive indices used in this study. Essentially, a 2nd degree polynomial fit was applied to the original data, allowing extrapolation and interpolation of the refractive indices for the CEPAS wavelengths. Additionally, as stated in the source material, an uncertainty of ±3 % was also added to the imaginary part of the refractive indices to account for potential inaccuracies in its definition. The effect of this more sophisticated approach on the re-calculated results was found to be negligible. The updated results are shown in the manuscript.

Line 231 Refractive indices: “Notably, Drinovec et al. provided values for wavelengths of 450, 532, and 633 nm, which differ slightly from the CEPAS laser wavelengths of 439.5, 516, and 635 nm. To account for this, a 2nd degree polynomial fit was applied to the original data, allowing extrapolation and interpolation of the refractive indices for the CEPAS wavelengths. Additionally, as stated in the source material, an uncertainty of ± 3 % was also added to the imaginary part of the refractive indices to account for potential inaccuracies in its definition. The imaginary part of the refractive index drives the attenuation of light within the particle and therefore essentially defines the particle’s light absorption. Nigrosin is often used in light absorption instrument testing as there is literature available describing its optical properties and because it forms spherically shaped particles when aerosolized and dried (Drinovec et al., 2022; Lack et al., 2006). Particle sphericity is an assumption of Mie theory.”

Results section updated with new figures and re-calculated correlation and accuracy metrics.

- I might have missed some key details regarding the development of the correction scheme, but it seems to me that the correction was developed from the divergence between measured and Mie estimates, and then used again to show that after the correction the size-resolved absorption matched the Mie calculations. If that was indeed the procedure, the better agreement in figures 8 and 9 is obviously not surprising, but also of relatively low value without further independent validation that the correction is indeed "correct". For example, if the issue is an incorrect index of refraction for particle phase nigrosine, then the "correction" would actually make the measurements less correct with respect to the real absorption value.

To make this simpler, we did not include the regression correction to Figures 8 and 9, but only the re-formulated loss function. This way the effect of the loss function on the results is clearer. In case the sample loss analysis is still considered useless, we are willing to remove it. However, we want to point out that its effect is fairly significant, especially compared to that of the refractive indices, for example.

- Line 322: "However, this discrepancy was ultimately resolved." As mentioned earlier, I do not feel like an empirical adjustment of the data to the theory can be considered as a resolution of the discrepancy between the same measurements and the same theoretical values. Further blind validation of the correction scheme would need to be carried out to test if indeed the correction is effective or not.

We have deleted this sentence.

Technical Comments

- Lines 38-40: Not all particles emitted by combustion are black carbon and therefore not all are agglomerates.

We have rephrased this as "Black carbon particles…".

- Line 44: These increases are indeed predicted, if anything, experimental studies have shown enhancement lower than predicted by Mie calculations (several published papers are available on this topic). The enhancement is defined as the absorption cross-section of the combination of the absorbing particle and coating to that of the absorbing particle alone.

We have rephrased "higher-than-predicted" as "increase in".

- Line 52: The main key factor is probably the heterogeneity of coating thickness on different particles, which is related to the size but not uniquely.

We agree. We have made minor changes to the manuscript to address this.

Line 63: "The underlying reason for this is that the coating accumulation and particle mixing-state depend on coagulation and condensation, which are both, in part, driven by particle size. For instance, it has been hypothesized that climate models' unrealistic approximation of uniform distribution of coating material across different sized particles is a source of discrepancy, and that in reality, the growth rate of a particle is nonlinear and dependent on,

among other things, particle size (Fierce et al., 2020).”

- Line 62: This assumes there is no phase change (evaporation of coating) during the exposure to the laser.

Added this note to the manuscript.

Line 75: “Photoacoustic spectrometers may not be as common as filter photometers, but they measure absorption directly in the aerosol phase without disturbing the sample (assuming no phase change when exposed to the laser).”

- Line 72: remove “a have”

Removed.

- Line 111: “air flow controls were replaced with aftermarket components” what was the reason to do so?

The controls were replaced to improve the usability of the system. We have clarified this in the manuscript.

Line 152: “The high voltage and sheath air flow controls were replaced with aftermarket components for practical purposes, and the system (encompassing both the CEPAS and DMA) was operated using a custom LabVIEW program.”