

We would like to thank both Reviewers for the valuable feedback, which has helped us to improve the manuscript. Below we added the response to each comment/question as well as the sections of manuscript that were adapted.

REVIEWER 1

Reviewer: Page 1. Title. It seems that the title captures only a part of the accomplishment of this work. As stated in the conclusions, one outcome is the demonstration of the aerosol generation system. Perhaps the title could be revised to reflect this.

Response: Thank you for this comment. We have revised the title to "[Comparing black carbon and aerosol absorption measuring instruments – a new system using lab-generated soot coated with controlled amounts of secondary organic matter](#)".

Reviewer: Page 5, paragraph starting at line 145. One goal of this study seems to be to evaluate new instruments with existing instruments. Since the 'PAS' is a new instrument, it would be good to give more details and a schematic. It seems that operation at such a high frequency (around 23 kHz) would result in a low pass filter response to light absorption, that heat transfer from the aerosol to the surrounding air would not all occur within the acoustic period so that the measurements would be strongly a function of aerosol size. Additional details about the motivation for, and design of, the prototype PAS would be useful, perhaps given in the supplemental section. The additional detail could explain how the instrument 'modulation frequency adapts...' (what measurements or theory are used to accomplish this).

Response: Thank you for pointing this out. We have added a detailed description of the PAS to the Supporting Information in Section S4 as follows:

S4 Description of the PAS

The measurement principle of the PAS is based on the photo-thermal effect of light absorbing particles in an acoustic resonator, as shown in Fig. S3. BC particles in the laser beam absorb light (1), which causes them to heat up (2). This heat is transferred to the carrier gas, releasing a pressure wave from the particle (3). This effect is enhanced if the light is modulated at the right frequency (4), building a standing wave within the resonator which is measured with a sensitive microphone (5). By matching the light intensity modulation frequency with the resonance frequencies of the chamber, the quality factor of the acoustic mode ($Q \sim 1000$ at 22.7 kHz) enhances the signal amplitude. The amplitude corresponds linearly to the amount of absorbed light.

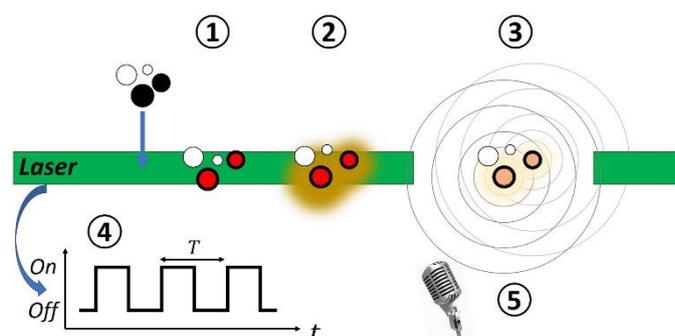


Figure S3. Illustration of the photothermal effect.

The photoacoustic instrument uses a novel resonator chamber with elliptical cross-section to enhance the photoacoustic, three lasers, a microphone, a loudspeaker, an amplifier and a signal-processing unit (lock-in amplifier). The three different wavelengths (445 nm with 700 mW, 520 nm with 300 mW, 638 nm with 300 mW power) of the diode-lasers allow us to measure wavelength-dependent optical properties of the aerosols.

The motivation for the elliptical cross-section is the possibility to separate the attenuation position from the measurement position of the microphone - at approximately the two focal points of the ellipse. The laser beam is guided by mirrors into the aluminium resonator along a focal point, which allows effective excitation of the transversal modes. The microphone is situated in the mid of the 24 cm long resonator case and can be moved transversally into the resonator, as indicated in Fig. S4. The microphone signal is then amplified and demodulated with a Stanford SR850 lock-in amplifier, which allows to measure amplitude and response delay of the photoacoustic signal at the excitation frequency, working currently at around 22.7 kHz. The measurements were performed with an integration time of 1sec therewith the mode can easily build up to a standing wave. The laser intensity modulation frequency is adapted periodically every 10 minutes, with the help of a loudspeaker and a frequency sweep, to match with the resonance frequency of the chamber.

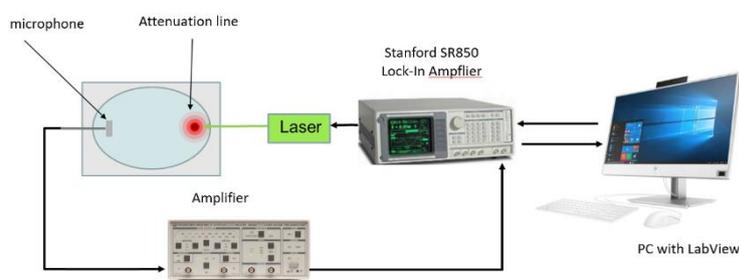


Figure S4: Schematic illustration of the PAS.

A loudspeaker (not shown in Fig. S4) is guided into the ellipse (parallel to the microphone) and attenuates the acoustic modes of the resonator chamber. These modes are independent of the light-absorbing particles in the chamber, but depend on the gas composition, temperature and pressure. The frequency of the loudspeaker is swept within a window where an acoustic resonance occurs. The frequency of the amplitude peak of this spectrum is then determined and its relative shift (to a reference measurement) is added to the laser intensity modulation frequency.

The motivation for the high frequency is on one hand to explore the response of the photoacoustic in the ultrasonic regime, as this was little explored in the past, and on the other hand to eventually measure the aerosol size distribution of the sample. At higher modulation frequencies it is easier to measure the phase delay between different aerosol size distributions, as the delay gets bigger for higher frequencies. At a fixed frequency, larger particle distributions could show a bigger phase delay than small particles, as the heat capacity rises with particle size. This phase delay was not yet measured and is still subject of investigation. Also measurements in the ultrasonic regime are expected to be less influenced by ambient acoustic noise.

Reviewer: Page 6, line 180. It's unclear to me how the babs values were normalized by total number concentration. Total number concentration seems potentially fraught with issues since light absorption of course depends on particle diameter in a complicated way.

Response: The total concentration is shown in the last column of Table 1.

Operation point	GMD_{mob} (nm)	$SS_{APAX,870}$ (-)	$SS_{Aneph/MAAP}$ (-)	AAE^1 (-)	AAE^2 (-)	EC/TC mass fraction ³ (%)	Total concentration ⁴ (cm^{-3})
1 – uncoated	91.7±0.1	0.027±0.001	0.0333±0.0002	1.14±0.01	0.875±0.014	91±7	25900±300
1 – coating 1	86.1±0.1	0.052±0.001	0.0749±0.0003	1.20±0.01	0.984±0.009	65±5	36500±100
1 – coating 2	83.4±0.1	0.12±0.01	0.148±0.001	1.28±0.01	1.05±0.01	48±3	35000±100
1 – coating 3	83.0±0.1	0.18±0.01	0.220±0.001	1.29±0.01	1.06±0.01	39±3	35500±100

GMD_{mob} (see column 2) does not change much throughout the experiment, therefore any artefacts arising from the dependence of light absorption on particle size are expected to be negligible.

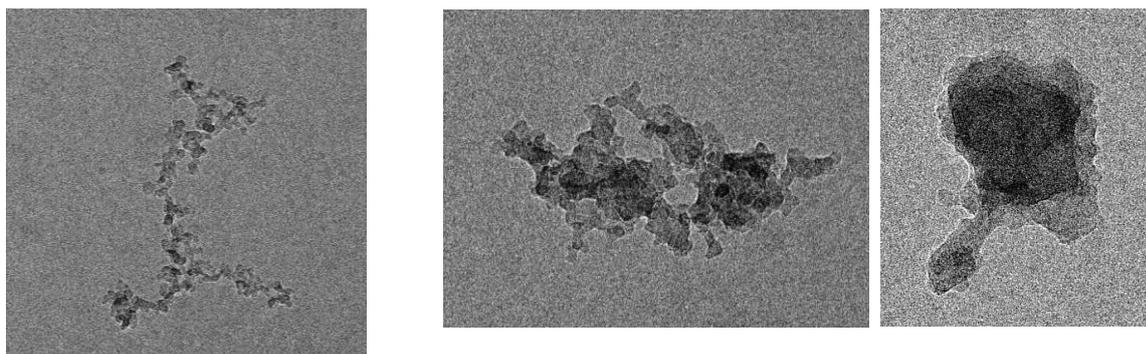
Example:

For "Operation point 1", the concentration of uncoated particles was taken as a reference value. The b_{abs} values of the coated points were adjusted by a correction factor equal to the ratio of the concentrations. For instance, for the first coating, b_{abs} was multiplied by $25900/36500 = 0.71$.

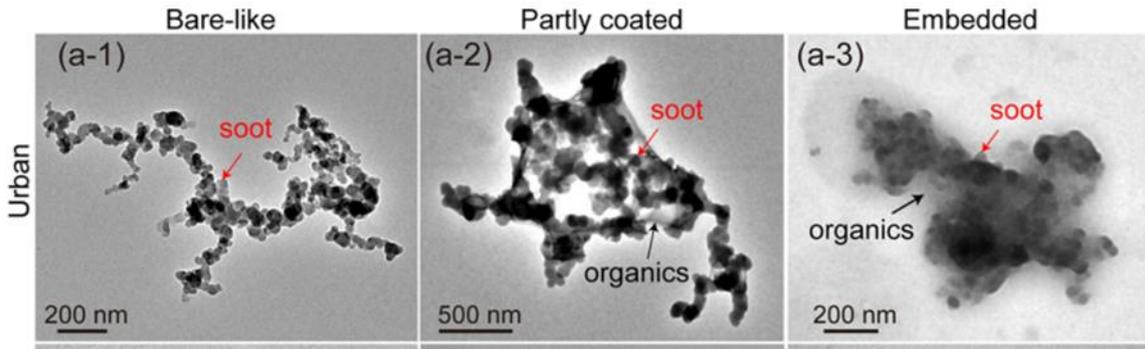
Reviewer: Page 8, line 215. Was electron microscopy performed to investigate soot core restructuring?

Response: This is indeed an important point. Cryo-TEM images of soot and coated soot particles were recorded during the study by (Ess et al., 2021) doi.org/10.1016/j.jaerosci.2021.105820.

The left panel below shows the TEM image of an uncoated (bare) soot particle, the middle panel is the image of a partly coated soot particle and the right panel the structure of a fully coated (i.e. embedded in the organic matter) soot particle. A gradual collapse of the soot core is observed as more SOA condenses on the soot core.



Our findings agree well with those reported by Wang et al. in the study "Fractal Dimensions and Mixing Structures of Soot Particles during Atmospheric Processing". Environmental Science & Technology Letters 2017 4 (11), 487-493, DOI: 10.1021/acs.estlett.7b00418 (see figure below):



Reviewer: Page 9, line 247. It seems that there is a contradiction in this paper to use the AE33 derived AAE to move the babs for other sensors (PAX for example) values all to 532 nm since the AE33 data is also described as being likely incorrect. What is the justification for doing this and what uncertainty enters the conclusions as a consequence?

Response: This is a valid concern. Figures 3 and 4 show b_{abs} at 532 nm. Two of the sensors (MSPT1 and PTAAM) measured already at 532 nm, so no correction was applied. Three other sensors (AE33 520 nm, PAS 520 nm and MAAP 670 nm) measured at a wavelength close to 532 nm, therefore only a very small correction was needed. Any artefacts related to this correction are expected to be negligible compared to the systematic uncertainties of these instruments. The PAX is the only instrument that measured at 870 nm and would require a considerable correction for converting b_{abs} to 532 nm.

In Table 1, two sets of AAE values are calculated based on measurements by the AE33 and by the PTAAM. AAE as calculated based on AE33 is about 20 % higher than the AAE based on PTAAM. This difference is most probably a result of the artefacts related to the interaction between the particles and the filter matrix. Since the sample aerosol features very uniform diameters (Table 1), we believe the reason for this difference is the wavelength dependence of the multiple scattering parameter C in the AE33. The use of the AE33 derived AAE overestimates systematically the extrapolated absorption coefficients (from 870 nm to 532 nm) for PAX. Compared to the calculation using the PTAAM AAE, this overestimation is 10%-14%. We have now revised Figs 3 and 4 of the manuscript by calculating b_{abs} based on AAE values from the PTAAM. We have also revised Tables S2 and S3 accordingly.

Reviewer: Page 9, lines 254-257. Which instrument(s) is (are) being considered reference methods for evaluating the results of these measurements? Which instruments provide the most and least correct measurements and how is that known?

Response: We believe that the AE33 is the instrument that suffers from the largest systematic errors. As discussed in this paper and in many previous publications, aethalometers assume a constant multiple scattering parameter C , which is not correct. C depends on the properties of the filter, design of apparatus and on the properties of the sample aerosol (e.g. particle size and SSA).

In our opinion, the PTAAM is probably the best candidate instrument to serve as reference method since the instrument can be calibrated at 532 nm with NO_2 . The calibration is transferred to the infrared (IR) channel using aerosolized nigrosin and its

relative green-to-infrared absorption ratio determined using a Mie calculation based on size distribution measurements. However, since the PTAAM is a very recent development and the manuscript on the design and calibration of the instrument is still in preparation, we felt it might be premature to make such a strong statement in the current manuscript. Moreover, dedicated inter-comparisons between PTAAM and PAX are needed to understand the deviations between the two instruments. Such studies are already in progress and the results will be presented in due time.

Reviewer: Page 9, line 257, and later in the paper, page 10, lines 264-267. It still is not clear why the PAX 870 nm absorption measurements show no sensitivity to coatings. At what wavelengths are the coatings expected to be light absorbing to various degrees? It seems that the authors could readily evaluate the insensitivity to coating at 870 nm by using the core/shell model for light absorption as a function of wavelength to theoretically evaluate light absorption enhancement at various wavelengths to confirm these results. Mass transfer is known to affect photoacoustic/photothermal measurements when the aerosol (or coating) has a high vapor pressure.

Response: This is an interesting and complex topic. There are two issues: first, the wavelength dependence of absorption of coatings (or coated absorbing cores); second, the “loss” of the latent heat from the photo-thermal or photoacoustic signal due to the volatilization of coatings.

Wavelength dependence

In this study, the soot coatings consisted of secondary organic matter from the oxidation of a biogenic precursor (α -pinene). According to the literature, biogenic SOA does not absorb as strongly as anthropogenic SOA. Light absorption is only observed in the UV and near-UV region:

<https://www.ncbi.nlm.nih.gov/pmc/articles/PMC7372712/>

[https://doi.org/10.1016/S0021-8502\(03\)00361-6](https://doi.org/10.1016/S0021-8502(03)00361-6)

Under certain experimental conditions, SOA from the ozonolysis of α -pinene showed negligible light absorption even at near-UV wavelengths:

<https://doi.org/10.1029/2010JD014387>

Therefore, it is not surprising that the PAX at 870 nm did not show any sensitivity to coatings. We also see the increase of the AAE with increasing thickness of the coating in the PTAAM (Table 1). We interpret this as the increase in absorption at lower wavelengths (532 nm) relative to the higher wavelengths (IR). Additionally, we see with PTAAM an enhancement of absorption at 532 nm of about a factor of 1.3 (Fig. 3) and no enhancement at 1064 nm (Fig. 5).

There are some differences between different modelled results regarding this effect. Virkkula (<https://doi.org/10.5194/amt-14-3707-2021>) states that lensing effect is less pronounced at longer wavelengths using a Mie model. Liu et al. (<http://dx.doi.org/10.1016/j.igsrt.2015.08.005>) show (Figure 9) that the calculated lensing effect differs between the discrete-dipole approximation (DDA) and core-shell Mie models. The DDA model predicts lower lensing at longer wavelengths; the core-shell model predicts a similar magnitude of lensing from the visible to the IR.

The differences between different models can be reconciled with measurements which we present using PTAAM.

Volatilization of coatings

We have evaluated the volatility of our SOA coating using a total carbon analyzer developed by FHNW (manuscript in preparation). We do not have any evidence that the SOA coatings used in our study are more volatile than typical suburban aerosols. Had the coatings we produced been volatile, this would have been seen in the comparison between the PAX and PTAAM.

PTAAM measures absorption at 532 nm and 1064 nm of the same sample (retention time for aerosol in the pump beam is 1000-times longer compared to the modulation interval). In principle, it would be possible for the pump beam of the PTAAM to evaporate the soot coating. Increase of the measured AAE shows, however, that at least part of the coating has not been removed. Estimated pump beam intensity in PTAAM is 2 W/mm² (PTAAM article in preparation – will be submitted shortly).

The recondensation of the possibly volatile coatings would occur during the off-phase of the pumping laser in the PAX and PTAAM. In the PAX, the acoustic wave generated by absorption of light, moves away from the sample signal generation region by the time recondensation occurs. In the photo-thermal interferometer, the signals due to volatilization and recondensation are generated in the region probed by the probe beam, so the recondensation is measured and the latent heat is taken into account properly. We do not expect any absorption by the coating itself and no absorption enhancement by the coated absorbing cores (see arguments above). We would expect to measure the difference in the IR between the PAX and PTAAM if the SOA coatings had been volatile. We saw no differences (Fig. 5) in agreement with our total carbon measurements.

Reviewer: Collectively the results shown in Figures 3b and 4b indicate that common aerosol light absorption measurement methods disagree by a factor of two, even in this case of laboratory-generated aerosol. If the measurements were given error bars that indicate precision and accuracy, would they overlap? It seems that the AE33 results are farthest from the rest, and that may be due to the choice of factors used to convert from filter-based to in situ light absorption. Assuming that the photoacoustic/photothermal measurements are closest to reality, can these measurements provide a suggested correction strategy for the AE33 light absorption calculation as a contribution to a growing literature on the subject?

Response: This is a very good question. Filter-based methods clearly suffer from systematic errors, which are much larger than the statistical ones. At the moment, we cannot give error bars for the combined uncertainties because the systematic errors can only be quantified by comparison to a reference method. Two methods which could serve as reference are the "extinction minus scattering method (EMN)" and photo-thermal interferometry (note that EMN suffers from large uncertainties at high aerosol SSA).

A correction strategy for the AE33 light-absorption calculation is discussed on pages 14-15 (Figure 4). We suggest, as others have done before us, to calculate new values for the aethalometer's C value as a function of SSA using the babs value of the PTAAM (or PAX) as a reference:

$$C_{SSA} = \frac{b_{atn,AE}}{b_{abs,ref}}$$

We have amended the text (Lines 361-371) as follows: "If the SSA of the ambient aerosol is known, then the calculated CSSA values can be used to post-correct the data from the aethalometer. In well-equipped monitoring stations, this can be performed using an integrating nephelometer and a reference absorption measurement at a single wavelength (Yus-Díez et al., 2021), and can be extended using a multi-wavelength reference absorption measurement and a multi-wavelength scattering measurement in a representative campaign. Multi-wavelength absorption corrections and the determination of C can be derived from off-line filter measurements with a time resolution of hours to days (Bernardoni et al., 2021). Moreover, a recent study suggests that "the low-cost and widely used PA monitors can be used to measure and predict the aerosol light scattering coefficient in the mid-visible nearly as well as integrating nephelometers" (Ouimette et al., 2021). By letting a low-cost nephelometer run parallel to the AE33 at monitoring stations, an approximate SSA value (based babs by the AE33 and bscat by the low-cost sensor) can be calculated and the babs of the AE33 can be refined by implementing a new CSSA. Both babs by the aethalometer and SSA can then be refined in multiple steps in an iterative procedure".

Reviewer: Minor issue: Page 15 line 333, 'of' is repeated.

Response: Thank you for spotting this error. We have removed the second "of".

REVIEWER 2

Reviewer: Accurate and precise measurements of aerosol light absorption remain the primary limitation towards reducing the uncertainty in direct radiative forcing brought about black carbon. To this end, the present manuscript brings together well-established instrumentation (AE-33 Aethalometer, single wavelength MAAP and a single wavelength PAX together) with new or prototype in situ measurements (3-wavelength (445 nm, 520 nm, and 638 nm) prototype photoacoustic sensor, a dual wavelength (1064 nm and 532 nm) photothermal interferometer (PTI) and a second single wavelength (532 nm)) to inter-compare and evaluate the performance of all measurement methodologies. Black carbon (BC) aerosol were generated using small burner outfitted with an oxidation chamber to produce secondary aerosols that could, in turn, coat the furnace-generated BC particles thereby enabling a comparison between uncoated and coated BC particles. By and large, the manuscript is clearly written save a few sentences - highlighted below - and the reported findings this manuscript are of value - especially the performance of the photothermal interferometers, which, have not been fully utilized towards the measurement of BC-containing particle light absorption. This said, there are a couple of nagging issues, discussed below, that this reviewer feels needs to be addressed before acceptance for publication can be rendered.

Reviewer: An important caveat with the present work is that there no "direct" measurement of amount of coating mass associated with the soot particles, such as might be procured from a BC particle mixing state analysis using the Single Particle Soot Photometer (SP2) or a measurement conducted on known soot particle diameters and where a mass classifier could

then be used to analyze the amount of coating on the BC particle. Instead, there is only an indirect measure of the coating mass viz-a-viz a EC/TC measurement. This may be a subtle point, but it is quite important as the authors use estimated coating to core mass ratios on polydispersed size distribution to derive conclusions about the observed absorption properties.

Response: Thank you for your comment. The amount of coating mass associated with the soot particles was calculated based on gravimetric measurements by TEOM (direct method). This is explained in line 240: "In Fig. 3b, the absorption coefficient at 532 nm ($E_{abs, 532}$) is plotted as a function of RBC. $RBC = (M_{total} - M_{BC}) / M_{BC}$ is equal to the mass of organic coating over the mass of uncoated soot as measured by the TEOM". Also, in the captions of Figures 3 and 4 it is mentioned that "The total mass to BC mass ratio and RBC are based on TEOM measurements".

The Reviewer is absolutely right that additional measurements with a mass classifier would have been beneficial. The reason why we did not perform such measurements was the lack of availability of an APM or CPMA. Concerning the use of SP2 for deriving rBC mass, we believe that this method suffers from high uncertainties at such small particle sizes (GMD_{mob} 90 nm), which are at the limit of detection. Nevertheless, such measurements have been performed in an experiment conducted by some of the co-authors and are presently being analyzed (and will be featured in a future publication).

Reviewer: A central issue with this manuscript is the attribution of absorption enhancement. A quick back-of-the-envelope Mie calculation, in the core-shell approximation limit, suggests that even a modest coating thickness of 25 nm of a non-absorbing coating on top a 100 nm BC core will lead to absorption enhancements of 1.4 at 550 nm, 1.3 at 870 nm, and even 1.26 at 1064 nm (and even with at 10 nm coating, the enhancements are all on the order of 1.17-1.13). So the observation of no enhancement at the longer wavelengths is a bit puzzling (unless one brings in particle diversity, which the authors do not bring up). Additionally, the authors do not discuss the potential impacts of measurement uncertainty on their analysis (in some cases the discrepancy among the in situ measurements approaches 75%). Instead, the authors suggest that the absence of observed absorption enhancement at 870 nm is consistent with that reported by Cappa et al.. This is a highly-glossed over argument. The Cappa work compared ambient urban emissions with ambient emissions that were passed through a denuder, from which absorption enhancements were derived. Even given the great care the authors exercised in the Cappa work, the authors acknowledged in a later reply, about the possibility that the denuder did not remove all the coating. So the authors are urged to exercise when using Cappa et al. observations to analyze their observations. Additionally, the authors are encouraged to consider the potential impacts of particle diversity on their observations (e.g., see Fierce, et al. Black carbon absorption at the global scale is affected by particle-scale diversity in composition. Nat Commun 7, 12361 (2016)). This said, given the very large discrepancies in the observations, the authors would be well served providing a discussion on the impacts of measurement uncertainty on their conclusions.

Response: We thank the Reviewer for this valuable comment. We were not aware that the work by Cappa et al. might have been biased. We have now removed the sentence in line 262: "~~This is in good agreement with Cappa et al., who reported that BC emitted from large to medium-sized urban centres (dominated by fossil fuel emissions) does not exhibit a substantial absorption enhancement when internally mixed with non-BC material (Cappa et al., 2012). E_{abs} during both field campaigns exhibited minimal dependence on RBC, with $E_{abs, 532nm}$ remaining close to 1 (absorption was measured by photoacoustic spectroscopy)~~".

We amended the text (Line 279) as follows: "A limitation of this study is that the relative humidity of the uncoated soot aerosols when entering the organic coating unit was low to very low (see subsection 2.1). No experiments were performed at high RH due to the presence of homogeneously nucleated SOA particles. It is known that the absorption by BC depends strongly on RH, and may lead to a factor of two increase in absorption at high RH compared with dry conditions (Fierce et al., 2016). This is one of the reasons why GAW (Global Atmospheric Watch) recommends measurements of light absorption at low RH (GAW report no. 226, 2016). Moreover, soot-containing particles generated in the laboratory under controlled conditions might have a more uniform composition compared to the aged soot particles in ambient air. In general, the range of absorption enhancement $E_{\text{abs}} = 1.1-1.3$ calculated based on the PAX and PTAAM agrees very well with (Fierce et al., 2016) who calculated a limited absorption enhancement ($E_{\text{abs}} = 1-1.5$) at low RH, when accounting for particle-level variation in composition of soot-containing particles. The dry conditions during and after ozonolysis of α -pinene may have also had an effect on the phase state of the SOM, leading most probably to a solid-state coating (Saukko, 2012)".

We also amended the section "Conclusions" (Line 390) as follows: "A limitation of this study was that the relative humidity of the uncoated soot aerosols when entering the organic coating unit was below 30%. More studies are needed at higher relative humidity in order to better simulate atmospheric processes".

When replying to a similar question from Reviewer 1, we noted that the enhancement (or lack of it at 870 nm) is dependent on two different issues. We repeat these two arguments here. It is important to note that Liu et al. (2016) ([10.1016/j.jqsrt.2015.08.005](https://doi.org/10.1016/j.jqsrt.2015.08.005)) show a considerably weaker enhancement for the more realistic DDA models than for the core-shell Mie model.

There are two issues: first, the wavelength dependence of absorption of coatings (or coated absorbing cores); second, the "loss" of the latent heat from the photo-thermal or photoacoustic signal due to the volatilization of coatings.

Wavelength dependence

In this study, the soot coatings consisted of secondary organic matter from the oxidation of a biogenic precursor (α -pinene). According to the literature, biogenic SOA does not absorb as strongly as anthropogenic SOA. Light absorption is only observed in the UV and near-UV region:

<https://www.ncbi.nlm.nih.gov/pmc/articles/PMC7372712/>

[https://doi.org/10.1016/S0021-8502\(03\)00361-6](https://doi.org/10.1016/S0021-8502(03)00361-6)

Under certain experimental conditions, SOA from the ozonolysis of α -pinene showed negligible light absorption even at near-UV wavelengths:

<https://doi.org/10.1029/2010JD014387>

Therefore, it is not surprising that the PAX at 870 nm did not show any sensitivity to coatings. We also see the increase of the AAE with increasing thickness of the coating in the PTAAM (Table 1). We interpret this as the increase in absorption at lower wavelengths (532 nm) relative to the higher wavelengths (IR). Additionally, we see with PTAAM an enhancement of

absorption at 532 nm for about a factor of 1.3 (Fig. 3) and no enhancement at 1064 nm (Fig. 5).

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Liu et al. (<http://dx.doi.org/10.1016/j.igsrt.2015.08.005>) show (Figure 9) that the calculated lensing effect differs between the discrete-dipole approximation (DDA) and core-shell Mie models. The DDA model predicts lower lensing at longer wavelengths; core-shell model predicts a similar magnitude of lensing from the visible to the IR.

The differences between different models can be reconciled with measurements which we present using PTAAM.

Volatilization of coatings

We have evaluated the volatility of our SOA coating using a total carbon analyzer, developed by FHNW, during the increase of the sample temperature (manuscript in preparation). We do not have any evidence that the SOA coatings used in our study are more volatile than typical suburban aerosols. Had the coatings we produced been volatile, this would have been seen in the comparison between the PAX and PTAAM.

PTAAM measures absorption at 532 nm and 1064 nm on the same sample (retention time for aerosol in the pump beam is 1000-times longer compared to the modulation interval). In principle, it would be possible to evaporate the coating by the pump beam in PTAAM. Increase of the measured AAE shows that at least part of the coating has not been removed. Estimated pump beam intensity in PTAAM is 2 W/mm² (PTAAM article in preparation – will be submitted shortly).

The recondensation of the possibly volatile coatings would occur during the off-phase of the pumping laser in the PAX and PTAAM. In the PAX, the acoustic wave, generated by absorption of light, moves away from the sample signal generation region by the time recondensation occurs. In the photo-thermal interferometer, the signal due to volatilization and recondensation are generated in the monitored probed by the probe beam, so the recondensation is measured and the latent heat is taken into account properly. We do not expect any absorption by the coating itself and no absorption enhancement by the coated absorbing cores (see arguments above). We would expect to measure the difference in the IR between the PAX and PTAAM if the SOA coatings had been volatile. We saw no differences (Fig. 5) in agreement with our total carbon measurements.

Considering other impacts of measurement uncertainty on the analysis, we provide statistical uncertainties in Figure 3 and 4 as well as the uncertainty of the splitter bias (1%, subsection 2.1). Other possible measurement uncertainties include diffusion losses in the connecting tubes. We have added the following text in subsection 2.1 (Line 110): "The PAX, PTAAM, PAS and AE33 have very similar sampling flows (1-2 L/min) and the difference in diffusion losses was compensated by adapting the length of the connecting tube to the flow of the instrument. For the MSPTI, which has a flow of 0.25 L/min, the connecting tube was kept as short as possible. Possible differences in the internal path length of the instruments (between the aerosol inlet and

measurement cell) were not taken into account. In the case of the MAAP, which was operated at a flow of 12 L/min, it was challenging to compensate for the difference in the diffusion losses. We cannot rule out that the measurements by the MAAP are biased but we estimate that this bias is <5% and therefore much smaller than the systematic uncertainties of this filter-based instrument".

To conclude, we think that systematic measurement uncertainties due to splitter bias and diffusion losses have a small impact on our analysis. The discrepancy in the in-situ measurements seems to be instrument-dependent. Especially the prototype PAS and MSPTI instruments suffer from artefacts arising, for instance, from instabilities in the laser power (see Lines 306-308 and 325-327 for more details). These instruments are still under development. The reason why we included the MSPTI and PAS in the study was not to suggest that these instruments are "mature" enough to be employed for BC monitoring, but to show how the combination of a miniCAST with the OCU can be used to test instruments based on different technologies. Commercialised instruments, such as the PAX and PTAAM, show in most cases a reasonable agreement. Figure 3b shows that the difference between the PTAAM and the PAX in babs is 10-20%, and the two instruments report practically the same Eabs (Figure 3c).

Reviewer: In their analysis described on lines 313-314, the authors used the NIR wavelengths to decouple the lensing effect. If different wavelengths are used, then the size parameters differ, and the amount of lensing changes. Using the NIR wavelengths, the authors propose that absorption enhancement observed at 670 nm and shorter is due to the production of brown carbon from the oxidation of α -pinene. The authors need to back up this assertion. What species do they think is responsible for light absorption all the way out to 670 nm? The authors might want to start by looking at the paper by Song, C., et al., (2013), Light absorption by secondary organic aerosol from α -pinene: Effects of oxidants, seed aerosol acidity, and relative humidity, JGR., 118, 11,741–11,749). Very few organic aerosols exhibit light absorption to such long wavelengths. Additionally, the variability among the in situ measurements of absorption enhancement is surly large enough to warrant a discussion on how this variability impacts the conclusions drawn from the observations as discussed above.

Response: This is a very good comment. We believe that the absorption enhancement reported by the MAAP might be an artefact. We amended the text (Line 270) as follows: "These results suggest that the absorption enhancement observed by the AE33, MAAP, PAS, PTAAM and MSPTI could be due to light absorption by SOM. Biogenic SOM is only expected to absorb light in the UV and near UV region (Song, 2013); thus, it is surprising that the MAAP indicates absorption enhancement at 637 nm. One of the possible reasons could be coating of BC in the filter by SOM or modification of the filter matrix optical properties by SOM (Cappa et al., 2008).

Specific comments:

Reviewer: The authors are strongly encouraged to be very pedantic when discussing the coated soot particles. For example, the authors should explicitly distinguish in their tables uncoated soot particles and soot-containing particles (i.e., coated soot). It took a few rereads for this reviewer to fully appreciate that the reported GMDs were for the polydispersed aerosol exiting the coating/mixing chamber, irrespective of whether the soot was coated or not, and not a more direct and meaningful comparison of "uncoated" soot with "denuded" coated soot. The authors are strongly encouraged to reword some of their sentences to reflect this. For example, on page 8 (lines 230 - 231) the authors write "...The GMDmod of the soot particles increased from 88 nm to 126 nm while the EC/TC mass fraction dropped". A more precise communication would be "The GMDmod of the soot-containing particles increased from 88 nm for uncoated soot to 126 nm following coating in the OCU...." I push

on this because the authors point out because absent explicit distinguishing between soot and soot-containing particles and how the authors report the size distributions, someone could easily misinterpret that the reported GMDs for the coated cases as being derived from denuded soot - which is clearly not the case.

Response: We apologise for the confusion. In Table 1, we do distinguish between uncoated and coated particles (see column 1), but we have now added the following clarification in the text (Line 216) to avoid any misunderstandings: "Note that the uncoated soot particles are "fresh" soot particles generated by the miniCAST burner (and not soot particles that have been coated and denuded)". We have also modified Figure 1 to show that the uncoated soot particles are delivered to the BC-measuring instruments through a line that bypasses the oxidation flow reactor.

We have also modified the text (lines 220 and 239) to make it clear whether soot is uncoated or coated: "The geometric mean mobility diameter (GMD_{mob}) of the soot particles gradually decreased from 92 nm (uncoated soot) to 83 nm (coated soot; coating 3) as shown in Fig. 2a" and "the GMD_{mob} of the soot particles increased from 88 nm (uncoated soot) to 126 nm (coated soot) while the EC/TC mass fraction dropped from ~85 % to ~10 % and the SSA increased up to ~0.7".

We agree with the Reviewer that a comparison of uncoated soot with denuded soot would be meaningful in order to better compare with atmospheric measurements. However, as the Reviewer noted in one of the comments above, denuding coated soot is not as straightforward as it sounds. Some SOA components are not volatile and might not be removed in the thermo-denuder or might be transformed by the heating, thus leaving a thin coating around the soot core. In our study, we made sure to avoid such artefacts by using fresh soot by the miniCAST burner.

Reviewer: It is clear that the filter-based measurements are biased quite high compared to the in situ techniques. Are the authors worried about relying on a filter-based technique to derive AAEs from which measurements at other wavelengths can be adjusted to a single wavelength?

Response: This is a valid concern, which was also raised by Reviewer 1. We have now revised Figs 3 and 4 of the manuscript by calculating b_{abs} based on AAE values from the PTAAM. We have also revised Tables S2 and S3 accordingly.

Reviewer: On line 262 the authors state that $R_{BC}=3.4$ which corresponds to EC/TC of 0.1. From the authors definitions of R_{BC} , and M_{tot}/M_{BC} - which should be $R_{BC}+1$ - then $EC/TC = 1/(R_{BC}+1)$. $R_{BC} = 3.4$ does not give $E/TC = 0.1$. Please check on this.

Response: The values of R_{BC} , and M_{tot}/M_{BC} are based on gravimetric measurements by TEOM as explained in the caption of Figures 3 and 4. It is true that the TEOM measurements do not agree so well with the EC/OC measurements. We believe that this is due to the high measurement uncertainties of the thermal-optical analysis and particularly with the difficulty to define the split point.

Reviewer: While it is likely that the BC particles are fractal-like, to get an SSA of 0.03 (for $m=(2,-1)$) requires a diameter of 50 nm, yet their GMD - mobility diameter - is 90 nm. The authors should explain this discrepancy.

Response: The (uncoated) BC particles with $GMD_{mob} = 90$ nm are fractal-like as shown by TEM analysis (see Ess et al., 2021 main manuscript and supplemental information <https://www.tandfonline.com/doi/full/10.1080/02786826.2021.1901847>).

Reviewer: This reviewer found it interesting that under the more dilute conditions (setup 0.1) the soot-containing particles were more heavily coated - as inferred from the GMDs. Why is that? Is this simply a limitation of the coating chamber in the limit of very high seed aerosol concentrations?

Response: As the dilution only affects the soot concentration and not the concentration of the VOC precursor, we have generally lower soot-to-VOC ratio, resulting in thicker coatings. When we do not dilute the soot aerosols, we have a higher number concentration of soot cores acting as seed particles (and therefore a larger surface area for the SOA to condense on), leading to thinner SOA coatings.

Reviewer: Somewhat related to the above, given the very high concentrations (i.e., 10^7 cc^{-1}) of soot in the micro smog chamber during the Setup 1 experiments, are the authors worried about coagulation? Since coagulation goes as the square of number concentration, it is very likely that coagulation is competitive with condensation and the authors are encouraged to evaluate the impacts, if any, on their observations.

Response: We measured the size distribution of soot upstream and downstream of the micro smog chamber and found no difference when $GMD_{\text{mob}}=90$ nm (even at number concentrations of 10^7 cm^{-3}). We observe, however, coagulation when $GMD_{\text{mob}} < 50$ nm and care must be taken when working with soot in that size range.

Reviewer: Lines 194 - 200. Introducing a filter media before discussing the measurement seems backwards, The authors are encouraged to combine and order the sentences in these two paragraphs.

Response: This is indeed a bit confusing. We have restructured the two paragraphs into a more logical order.

Reviewer: This reviewer found the legends in figures 3 and 4 a bit confusing in that the wavelength listed by the instrument is its operating wavelength, yet the data presented in the plots is adjusted to allow comparison at a single wavelength (532 nm). The authors might want to clarify this a bit better in the figures.

Response: We apologise for the confusion. We have amended the text in the captions of Figures 3 and 4 as follows: "The legend below Figure 3c (Figure 4c) indicates the wavelengths at which the measurements were performed".