

We thank the editor and reviewers for the timely handling of our manuscript, particularly during this difficult pandemic year. Please find below our point-by-point responses to each of the reviewer comments, including descriptions of the modifications we have made to the manuscript. Reviewer comments are in black text and our responses are given in blue text.

Referee #1: Timothy Onasch

General comments:

Overview: This paper investigates the errors associated with the extinction, scattering, and absorption measurements of the recently commercialized CAPS PM_{2.5} instrument on deployment as a field-based instrument. The paper focuses on the method and errors associated with the scattering truncation and scattering calibration (to the extinction measurement) and how these relate to the absorption measurement. Field data cases are shown that highlight the strengths and weaknesses of the CAPS PM_{2.5} absorption measurement. The authors build a truncation model for the CAPS PM_{2.5} that explicitly includes the glass tube inside the scattering sphere and build a full error model for the EMS-based (Extinction Minus Scattering) absorption measurement. Finally, a list of recommendations is given to ensure accurate absorption measurements in the field using the CAPS PM_{2.5} instrument. I find this manuscript well written and organized. In general, I have very little argument with their structure, methods, and conclusions. This paper has a relevant and timely topic for the Atmospheric Measurement Techniques (AMT) journal, specifically, and the atmospheric community, in general. In particular, the manuscript addresses the critical need for field-based absorption measurements on freely-floating particles (i.e., not filter-based measurements). This article deserves to be published in AMT and is almost ready for direct publication. I have a few specific comments that should be addressed prior to publication.

We thank Dr. Onasch for his careful review and constructive comments. These have helped to improve the paper by clarifying some important points. We have addressed each of the comments as explained in the detailed responses below.

Specific comments:

1.) Terminology: Filter-based instruments do not measure absorption. In general, the authors are very careful about this issue; however, it is worth restating. First sentence on the last paragraph of page 2, for example, states: “aerosol light absorption has been measured by detecting the attenuation of light transmitted through aerosol samples deposited on filter substrates.” Perhaps a slightly more accurate portrayal would be to state: “aerosol light absorption derived by measuring the attenuation of light transmitted through aerosol samples deposited on filter substrates.”

Agreed. We have modified the sentence in question as suggested. It now reads: “Traditionally, aerosol light absorption has been derived by measuring the attenuation of light transmitted through aerosol samples deposited on filter substrates (e.g. Rosen et al., 1978)”

2.) The Multi-Angle Absorption Photometer (MAAP) instrument is highlighted by the authors as an improved filter-based method for deriving aerosol light absorption from light attenuation and scattering from an aerosol loaded filter substrate. The MAAP is indeed an impressive filter-based system; however, it is still a filter-based attenuation (and scattering) technique and therefore susceptible to filter substrate-based issues, in addition to potential failures in the 2 stream radiative transfer model approximation from which the absorption is derived. The MAAP is not an absorption standard in any sense and this should be acknowledged. The authors describe some of the errors associated with the MAAP on page 16, but do not include this in the analysis of the instrument comparison in section 6.2. It is understandable that these errors are not emphasized in the comparison, as the focus of the work is on the CAPS PM_{ss}a absorption measurement. However, in this case the authors should either state that their analysis of this data make the implicit assumption that the MAAP data has no uncertainties for this comparison, or acknowledge the inherent errors in the MAAP by including them in the comparison. For example, all of the differences between the CAPS PM_{ss}a absorption and the MAAP absorption measurements are all explained as potential issues with the CAPS, but no discussion is given about potential issues with the MAAP measurements..

We agree the MAAP should not be considered as a true absorption standard. Our intention is rather to use the MAAP as a common reference point, since the instrument unit-to-unit variability for the technique has proven to be very good. Thus, by comparing our CAPS PM_{ss}a absorption coefficients against those measured by a MAAP we expect future studies will also be able to use MAAP measurements as a way to link to the CAPS PM_{ss}a results presented in this work.

We also agree that these points should be stated more explicitly in the main text. We have added additional explanation to Sect. 3.1.1 and Sect. 6.2 (also in line with reviewer comments 14 and 16) in order to highlight that we do not consider the MAAP measurements to be a true absorption standard, to mention that its ability to measure super-micrometer particles requires further investigation, and finally to include discussion of MAAP errors alongside the CAPS PM_{ss}a error discussions.

The updated text in Sect. 3.1.1 is the following:

“During the RAOS campaign (Petzold et al., 2005), MAAP absorption coefficients were observed to have no relationship with aerosol SSA. However, at extremely high SSA values the absorption coefficient measurements from the MAAP can be biased high. It is also important to consider that – to the best of our knowledge – no dedicated study has yet been performed to assess the precision and accuracy of MAAP measurements of samples containing a large fraction of super-micrometer particles. To quantitatively compare the MAAP and CAPS PM_{ss}a absorption coefficients during the Cabauw field campaign, both coefficients were adjusted to standard temperature (273.15 K) and pressure (1 atm). It should be stressed that in this comparison we do not consider the MAAP to be a true reference standard for measuring aerosol absorption coefficients. Rather, the value of the instrument for the present study lies in the fact that it displays very low instrument unit-to-unit variability, which means it can provide a common and stable reference point against which CAPS PM_{ss}a absorption measurements can be compared.”

The updated text in Sect. 6.2 is the following:

“In this section we use an example dataset from the Cabauw campaign to make a direct absorption instrument-to-instrument comparison between the CAPS PM_{ss}a and the MAAP. This comparison is

performed in order to gain insight into the ability of the CAPS PM_{ss}a to measure absolute aerosol absorption coefficients. As discussed in Section 3.1.1, the MAAP was used for this comparison because it provides stable and reproducible absorption measurements. However, it should be reiterated that the MAAP is not a true absorption reference standard: this instrument is associated with its own measurement uncertainties, which are not well characterized for aerosols with large contributions of super-micrometer particles. Despite these issues, our focus here is only on the uncertainties related to the CAPS PM_{ss}a absorption measurements, including detailed characterization of the scattered light truncation effect. The CAPS630b was the PM_{ss}a unit operated during the Cabauw campaign. The unit ran autonomously, continuously, and stably over the one month of operation, which we have found to be typical for CAPS PM_{ss}a units operated at stationary field sites.”

Additionally, the following sentence has been added to Sect. 6.2.4:

“Another possibility is that the MAAP absorption coefficients were systematically overestimated (uncertainties of up to 7% can be expected based on previous studies, see Section 3.1.1)”

3.) On page 6 line 175, the authors state that the mirror purge flow is “drawn” continuously over the mirrors, whereas the purge flow is actually pushed (i.e., positive flow)..

Thanks for the clarification. We have changed ‘drawn’ to ‘pushed’.

4.) On page 6 line 182, the authors state that the extinction channel signal is monitored by a vacuum photodiode. The one exception for this is the 780nm wavelength system which uses a PMT detector for greater sensitivity at the longer wavelengths.

Thanks for the clarification. We have changed the sentence accordingly (“...is monitored by a vacuum photodiode or, in the case of the 780 nm unit, a photomultiplier tube (PMT)”).

5.) Page 7 line 221 and Figure 2, the scattering channel is normalized to the amount of light circulating in the optical cell during the LED off phase using the reported “signal” levels and is a function of the cell loss, which controls the rate of decay of light in the cell. Thus, the authors are incorrect when they note that the total extinction is used in the scattering channel ($b_{sca, sample}$) calculation. Extinction is related to the loss, but is itself a differential measurement of the loss measured with sample from the loss measured without sample. This needs to be corrected..

Thanks for this clarification. We have changed the relevant labels in Fig. 2 from ‘extinction’ to ‘optical loss’.

6.) Page 9 line 260 (and elsewhere), the authors note that the CAPS PM_{ss}a firmware automatically applies a geometry correction factor of 0.7. The manufacturer’s geometry correction factor, as noted in Onasch et al. (2015), is 0.73 for CAPS PM_{ss}a monitors. It has been noted that early instruments may have dropped the second digit in the data files, even though the internal value was always 0.73.

Thanks for this clarification. We indeed took the figure of 0.7 from the header of the data acquisition files where the 2nd digit has been dropped. We have changed the text in all relevant places from 0.7 to 0.73, in order to represent the actual internal value. It was also necessary to reprocess the data displayed in Figs. 4 and 8 using the value of 0.73 rather than 0.7. As is to be expected with such a small change, the resulting changes to the final plots are very minor and do not alter our final conclusions.

7.) Page 9 line 273, “N2” should read “NO2”

The ‘N2’ was deliberate and meant to refer to the low span calibration point. But we agree that that doesn’t really fit with the sentence and causes confusion. We have simply deleted the N2 to prevent any possible confusion.

8.) Page 9 line 275, the authors state “we have observed that the instrument can take a long time (~hours) to adjust and stabilize when filled with different gases.” Note that this may be true for the CAPS PMssa monitor due to the purge flows that have large area filters and very slow flows, but is not true of the CAPS NO2 systems, for example, that do not have the more complicated flows. Thus, this sentence should be modified to reflect the actual or presumed reason for this observed behavior. There is nothing mysterious about this issue. Further, this issue will be more of an issue for “sticky” gases, such as water vapor.

We have added a qualifier to the end of the sentence to indicate that this behavior is expected. It now reads: “...we have observed that the instrument can take a long time (~hours) to adjust and stabilize when filled with different gases (as expected due to the low flows and large filter areas in the purge flow setup)”

9.) Page 15 line 468, remove “anyway” from “anyway small relative”

The “anyway” has been removed as suggested.

10.) Page 19 line 597, the authors write “A number of repeat experiments were performed for some of the aerosol types, as indicated in Table 3.” Table 3 lists the “Dates of experimental repeats”. That said, the actual dates are not the important information here, rather the number of measurements (i.e., repeats) and the time between is the useful information. Suggest modifying Table 3 to address.

Agreed. We will modify the table so that the number of repeat measurements can be easily determined.

The modified version of Table 3 now looks like the following (including the updated refractive index value for ammonium sulphate, as suggested in the next comment 11.):

Table 3: Test aerosols used to measure scattered light truncation in the CAPS PMssa as a function of particle diameter and the values of parameters used in the corresponding model calculations. All particles were size classified by AAC.

Test aerosol	Refractive index (wavelength)	Geometric standard deviation of the size distributions	Number of repeat experiments at each wavelength (dates)
PSL spheres	1.59 + 0i (at 450, 630 & 780 nm)	1.05 and 1.1	2 at 450 nm, 3 at 630 & 780 nm (between Apr. 2018 & Jan. 2020)
DEHS (Di-Ethyl-Hexyl-Sebacat)	1.46 + 0i (at 450 nm); 1.45 + 0i (at 630 & 780 nm)	1.1	1 at 450, 630 & 780 nm (Jan. 2019)
Ammonium sulphate	1.52 + 0i (at 450, 630 & 780 nm)	1.1	3 at 450 nm, 4 at 630 nm, & 2 at 780 nm (between Apr. & Aug. 2018)

11.) Table 3 – why did the authors chose to use 1.5+0i for ammonium sulfate refractive index? Toon et al. 1976 suggests a value closer to 1.53 for VIS. Presumably this minor difference makes little to no effect.

This was a typo, thanks for picking this up. The ammonium sulphate calculations were actually performed with a refractive index of 1.52, in line with a number of chemical information databases and closer to the Toon et al. value. We have modified Table 3 to fix this typo.

12.) Page 22 lines 676 to 688 and figure S6. In Figure S6 and in this paragraph, the authors discuss an issue they observed in a field study that is not discussed in the paper except for this single example. The example shown comes from a field study where the highly reflective mirrors on the CAPS PMssa monitor deployed became significantly contaminated. Under these conditions, the instrument is operating outside of its intended parameter space, though to be fair, the intended parameter space on dirty mirrors is not defined by a set threshold, but rather by operator decision making. The data in the figure and discussed in the text notes that as the mirrors get dirtier, the extinction measurement increases relative to a CAPS PMex system and a nephelometer. As the extinction channels in the CAPS PMssa and the CAPS PMex are nearly identical (SSA slightly shorter than EX), the increase in the mirror contamination could have readily happened in either system. In other words, there is nothing here specific to the CAPS PMssa compared with the CAPS PMex for the extinction measurement. The authors discuss this issue as if it were related to a non-linearity at high extinction levels, though later decide that this does not fit the observations. They do not note that the mirrors become dirty when either (a) there is a significant pressure burst that can lift sediment from walls and deposit on the mirrors or (b) the purge system in the CAPS PMssa is failing, causing particles to access the purge regions. The latter explains both the increase in the baseline loss of the cell due to dirty mirrors and the observed increase in the extinction coefficient as the sample pathlength increases as particles cross the light beam inside

the purge regions that are normally particle free (i.e., the geometry parameter decreases). The authors conclude that they (i.e., the operators) must be vigilant and keep the mirrors clean. In essence, this paragraph and supplemental figure are hardly relevant to the current discussion of errors in the CAPS PM_{ss}a instruments measure of particulate absorption, as purge failure is a potential issue for any CAPS PM_{ss}a or PM_{ex} system. At worst, this section/figure could cause readers to think that the CAPS PM_{ss}a fails under high loadings, which is not true. Basically, while this section/figure aren't incorrect, it does not add anything useful to the discussion of errors at hand and could potentially cause confusion. I would suggest this section/figure be removed, or at least, qualified by noting that the issue here were likely caused by a purge system failure that went undetected during a field study, which is not a minor contamination issue, and under such out-of-tolerance conditions the CAPS PM_{ss}a, like any instrument, struggles to meet specifications. For example, the authors use this information to make the statement, "the CAPS method is effectively 'calibration free' (apart from the geometry correction factor, as discussed in Sect. 2.2.1, as well as potential non-linearities at high baseline losses)" on page 9. I would suggest that this over emphasizes an issue that was caused by user fault (failure of purge flow), rather than potential measurement fault.

While we agree that this example is somewhat separated from the rest of the paper we think it is important to show for the following reasons. 1) As noted there is no set threshold above which a user can be sure mirror contamination is affecting the measurements. Although this will vary from unit to unit, we still think it is useful to show an example of the behavior that might be expected for these types of increases in the baseline optical loss (we are often asked this type of question by new users). 2) The fact that the extinction measurement became biased as the contamination increased while the scattering measurement remained more stable implies the instrument's cross calibration constant varied due to the contamination event. In this sense, we think the result is important to show and fits nicely within the context of the section in question, Sect. 4, which is focused on the stability of the cross calibration constant over time.

Having said that, we agree that more emphasis should be placed on qualifying the fact that this is an example of when the instrument is operating outside of its intended operation range, as well as the reasons this could occur (purge flow failure or pressure burst). We will modify the revised manuscript accordingly as suggested. We will also explicitly state that this type of instrument failure isn't directly related to measuring high aerosol loads to try and prevent readers drawing this incorrect conclusion.

Finally, we will remove the two sentences on L684 of the AMTD version of the manuscript relating to the non-linear regime hypothesis and then the ruling out of this hypothesis. However, we note that this example was not the only reason behind the statement on L267 about "...potential non-linearities at high baseline losses)." We have observed this type of behavior in laboratory experiments with both CAPS PM_{ss}a and PM_{ex} systems. Therefore, we elect to keep this particular statement as is.

The updated paragraph in Sect. 4 now reads:

"In addition to continual drifts in β_{Rayleigh} over time, it is also interesting to note how β_{Rayleigh} can change following known instrument-malfunction events such as contamination of the PM_{ss}a optical cavity. Fig. S6 displays CAPS PM_{ss}a measured b_{ext} (left panel) and b_{sca} (right panel) at 450 nm against independent measurements of these quantities (CAPS PM_{ex} for b_{ext} , nephelometer for b_{sca}) during a field campaign at the rural background site of Melpitz, Germany. Throughout the duration of these measurements the optical cavity of the 450nm PM_{ss}a unit became contaminated, moving the instrument outside of its

intended range of operation, with average baseline optical loss varying from 758 to 1248 Mm^{-1} . Such contamination events can occur due to large pressure fluctuations in the aerosol sampling line or failure of the instrument's purge flow system. They do not occur just by measuring high aerosol loads. In this case, the instrument-malfunction contamination event caused an increase in the bias of the b_{ext} measurement relative to the corresponding PM_{ss} measurement from 5 to 17%. However, over the same period, the bias of the b_{sca} measurement with respect to the corresponding nephelometer measurement was unchanged, which implies that $\theta_{Rayleigh}$ did change. Therefore, as specified by the manufacturer, we recommend that the CAPS PM_{ss} baseline should be monitored continuously throughout measurement campaigns for signs of mirror contamination, and that contaminated mirrors are cleaned promptly."

13.) Page 10 line 297 add "to" in "... we refer to the calibrated scattering coefficient..."

Thanks for picking this up. The "to" has been added.

14.) Coarse mode. The authors include significant discussions in this paper relating to the errors in b_{sca} and b_{abs} by the CAPS instrument (Sections 6.2.2, 6.2.3) and during the instrument correlation with the MAAP in section 6.2.4 when measuring coarse mode (i.e., PM₁₀) particles. While these discussions are important, the authors need to qualify these discussions with the facts that neither the CAPS nor the MAAP have been extensively tested with coarse mode particles (to my knowledge). For example, the discussion of errors in the MAAP on page 16 mention the RAOS study and the Mueller et al. (2011a) study, but neither of these studies included coarse mode particles (i.e., particles greater than 1 micron in diameter). Thus, while including these discussions here is important and relevant to the ambient results, the authors need to acknowledge that neither technique has been extensively studied for coarse mode particles (to my knowledge), which limits the quantitative aspect of these discussions. Furthermore, a large fraction of the discussions on Figure 8 and the subsequent analysis relies on the coarse-mode and full dataset (which is highly biased by a coarse-mode). The authors should at least qualify these results with respect to the lack of coarse mode quantification in both instrument techniques.

This is a good point and we generally agree: also to our knowledge there has been no extensive and dedicated studies on the ability of the MAAP to measure absorption coefficients for super-micrometer aerosols. We do not think this point affects the majority of the discussion in Sect. 6.2, which is focused on how the CAPS PM_{ss} measurements responds to the presence of aerosols with strongly forward-focused scattering phase functions. However, we agree that it is an important qualification to highlight.

We added explicit statements to both Sections 3.1.1 and 6.2 to highlight this issue. The modified text is included above in the response to the Reviewer comment number 2).

15.) Page 25 line 765, very nice correlation for high time resolution data. What does the correlation plot (or histogram of the ratios) of CAPS PM_{ss} b_{abs} to SP2_rBC look like for the data presented in Figure 7? Since you explicitly state not to look at the quantitative value, I did that. The apparent ratio is 8 m^2/g ,

which is very high for a wavelength of 780nm. What are the SSA values for these samples? The authors point out that the one unknown (i.e., not measured) component during this study is the scattering truncation (i.e., size distributions). For scattering truncation uncertainties, the SSA would have to be high to attempt to account for these apparently high MAC values. What were the size distributions of rBC measured by the SP2? What about the scattering measurements by SP2? Could the SP2 be saturating at these high rBC levels? What about absorption enhancement due to coated rBC particles (would be $\sim 1.5x$ assuming a $1/\text{wavelength}$ from 550nm 7.5 m²/g MAC, which might not be unreasonable)? Perhaps, it would be either better to (a) remove one or both axes if considered non-quantitative or (b) provide a potential rationale for this apparent issue.

As well as the uncertain truncation correction constant the other main quantitative issue in this figure is related to the relatively small size of the freshly-emitted rBC cores, which means the SP2 was unable to detect an unknown fraction of the total rBC mass (this issue is discussed at length by Pileci et al. 2020, including with reference to this particular field campaign). Underestimated total rBC mass from the SP2 is the most likely reason for the high apparent MAC value. In combination with the unknown truncation correction constant, this is also the reason we don't think there is any value in analyzing or constraining the quantitative aspects of this result any further. However, we agree it is important to add further rationale about this for readers who are as curious as the reviewer. In the revised manuscript we will add statements in the main text and in the caption of Fig. 7 that the absolute rBC mass concentrations should also be treated with caution, since the SP2 was unable to detect all of the rBC mass present due to the small size of the rBC cores.

The modified text in Sect. 6.1 now reads:

“Although Fig. 7 demonstrates the responsiveness of the CAPS PM_{ss}, it must be stressed that the absolute values of the plotted b_{obs} measurements are still uncertain and should not be used quantitatively (the SP2 measurements of rBC mass are also underestimated and should not be used quantitatively, since the instrument was unable to detect an unknown fraction of the total rBC mass due to the small size of the freshly-emitted BC cores; Pileci et al., 2020b)”.

And in the caption of Fig. 7:

“In addition, the SP2 measurements have not been corrected for missing mass and should also not be interpreted quantitatively (Pileci et al., 2020b).”

Pileci, R. E., Modini, R. L., Bertò, M., Yuan, J., Corbin, J. C., Marinoni, A., Henzing, B. J., Moerman, M. M., Putaud, J. P., Spindler, G., Wehner, B., Müller, T., Tuch, T., Trentini, A., Zanatta, M., Baltensperger, U. and Gysel-Beer, M.: Comparison of co-located rBC and EC mass concentration measurements during field campaigns at several European sites, *Atmospheric Measurement Techniques Discussions*, 1–32, doi:<https://doi.org/10.5194/amt-2020-192>, 2020b.

16.) Section 6.2. This section analyzes a comparison of the CAPS absorption measurements compared with the MAAP absorption measurements. As noted above in points 2 and 14, both instruments have associated errors for submicron and potential unknown errors for supermicron. The authors state (page 25, line 784) that this was done to “perform a full quantitative assessment of the ability of the CAPS PM_{ss} to measure absolute aerosol absorption coefficients...”. In reality, this section is a direct

instrument to instrument comparison, rather than a quantitative assessment for the measurement of absolute aerosol absorption coefficients. It is likely that the errors in the MAAP are smaller than the errors in the CAPS b_{abs} , but they should still be included. What MAC value was assumed for the MAAP to derive the b_{abs} values? On page 28, when discussing the ~20% discrepancy between the CAPS b_{abs} and MAAP b_{abs} , the only errors discussed are those of the CAPS geometry correction factor and truncation factor, which are only on the order of 2-9%. The MAAP uncertainties stated on page 16 indicate similar orders of magnitude (5-7%), but are not discussed.

Again, this is a good point and we generally agree. We will adopt the language accordingly, including:

- Rewording the phrase "...a full quantitative assessment of the ability of the CAPS PM_{ss} to measure absolute absorption coefficients" to something like: "a direct instrument to instrument comparison in order to gain insight into the ability of the CAPS PM_{ss} to measure absolute absorption coefficients".
- Mention of MAAP-related uncertainties in Section 6.2 (specifically in the discussion on page 28 as suggested). We note that we refrained from plotting MAAP-related error bars in Fig. 8 because they are too small and overly complicate an already busy plot. However, we will add a statement to the caption of Fig. 8 that MAAP-related uncertainties are not displayed for visual clarity.

The manufacturer-specified MAC value of 6.6 m²/g was used to convert the eBC concentrations reported in the output of the MAAP firmware to absorption coefficients. We will include this information in Section 3.1.1

Update: the following changes were made in the manuscript.

Section 3.1.1. now includes the following statement:

"A mass absorption cross section value of 6.6 m² g⁻¹ was used to convert the equivalent BC mass concentrations reported in the firmware output of the MAAP to absorption coefficients (as specified by the manufacturer)."

The relevant modified text in Sect. 6.2 is included above in the response to the Reviewer comment number 2).

The caption of Fig. 8 now includes the following statement:

"Uncertainty in MAAP b_{abs} of up to 7% can be expected based on previous studies (Section 3.1.1): this uncertainty is not displayed for visual clarity."

17.) Figure 8. This figure shows a lot of useful information tightly packed into one figure. The problems of using log-log plots for instrument intercomparisons is that it (a) blows up the noise at low signals and (b) removes any potential negative values from the comparisons. My first question here is how were the fits done for this figure? For example, the fit in the center plot does not go through the data shown at all. Thus, it is not a linear fit of the data in linear space and placed on log-log axes. Further, the fits assume that there is no y-intercept (or x-intercept). If it is a power law fit to the data, then there must

be a lot of data not shown that is highly biasing the power fit. How do the fits account for the negative values? More information is warranted on the fitting approach and why it was chosen.

The decision to display these results on a log-log scale was a deliberate one. We believe it is important to show the level of agreement across the full range of measurements. In contrast, display on linear axes would visually preference those measurements performed at the highest b_{abs} values. Although these particular scatterplots don't show much evidence of random noise at low signal levels for the given averaging level of 1 hour, we believe that even if they did this would rather be an argument for showing the results on a log-log scale, i.e., we aim for maximum transparency, and prefer that readers have the opportunity to judge for themselves at what signal levels noise starts to become an issue.

The major drawback with our choice is that, as suggested, the negative CAPS PM_{ssa} measurements cannot be displayed. In this instance however, we believe the most pertinent piece of information is that negative measurements were sometimes present, not how those negative measurements actually looked like. Therefore, we added text boxes indicating the fraction of the CAPS PM_{ssa} measurements that were negative in order to get around this issue.

For these reasons, we prefer to keep these measurements displayed as they currently are on log-log axes.

18.) Figure S15 caption indicates that the fit lines in Figure 8 are not fits, but rather mean ratios.... – this information needs to get into the main text and main figure captions!!! By taking the mean ratio, the resulting values are heavily influenced by noise in the smaller measurements. This would explain why the orange lines do not pass through the majority of the data points, especially at higher values of b_{abs} and lower values of SSA, where one would expect better agreement. Another potential approach that could be taken would be to estimate a conservative limit (for example 50 Mm⁻¹ was chosen for this purpose when looking at the geometry correction factors above) and then look at the histograms of the ratios (again similar to the geometry correction factor analysis). This approach might better define the mean, variance, and skewness of the resulting histograms, where the skewness could be used as a measure of how well the truncation models are being applied, rather than the vague (though true) color trends.

Firstly, we apologize for the omission of details concerning the mean ratio calculation in the original submission. This will be corrected in the revised manuscript with appropriate description added to both Sect. 6.2 and the caption of Fig. 8.

The decision to display lines with gradient of mean ratio rather than standard linear fits was taken to avoid biasing the fits towards the measurements at high b_{abs} values. One key difference between the measurements shown in Fig. 8 and those used to calculate the cross calibration constants (where, as suggested, a conservative limit of 50 Mm⁻¹ was used, e.g. Figs. S3, S4, S10 and S11), is that the measurements in Fig. 8 are hourly averages, not 1 second measurements, and therefore they are less affected by noise at very low signal levels (a good example of how averaging to 1 hour removes the random measurement noise at low signal levels is shown in Fig. S9). Therefore, noise at low signal levels does not bias the mean ratios displayed in Fig. 8 substantially.

The fact that the lines of mean ratio do not pass through the majority of the points plotted in Fig. 8e is rather to do with the fact that this subplot contains a sizeable fractions of negative CAPS PMssa measurements that are not displayed on the log-log axes. These negative values are the result of over-corrected truncation, not noise at low signal levels, as we discuss at length in Section 6.2.

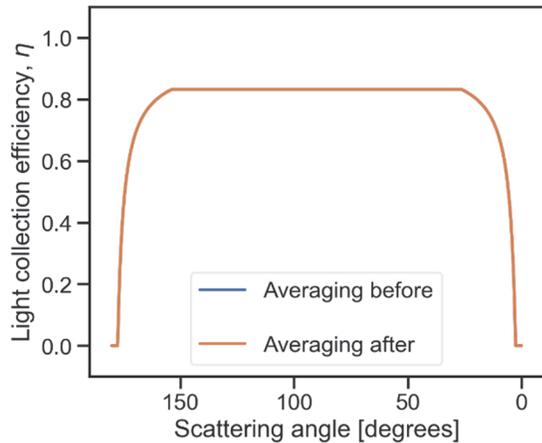
Finally, we experimented considerably with more quantitative measurements for assessing the performance of different truncation approaches (e.g. histogram skewness, bias relative to the MAAP as a function of SSA). However, our goal is not optimization of the parameters in our Mie-based truncation correction model. I.e., we're not looking to identify a single best set of parameters for this particular dataset. The reason is that this optimized set of parameters would not be generally applicable to other studies, so there would be little value in reporting it. Rather, our aim is to show the sensitivity of the CAPS PMssa b_{abs} measurements to different truncation correction scenarios, to provide general insight and intuition into the effects of the correction on the final results. For this reason we think it is more appropriate to rely on qualitative metrics such as the color change rather than specific quantitative metrics.

Update: the following text has been added to Sect. 6.2.4 to explain that the orange lines in Fig. 8 are mean ratios, and why mean ratios were chosen rather than linear fits:

"The uncertainty envelopes are plotted around straight lines (shown as solid orange lines) representing the mean ratios of the CAPS to MAAP b_{abs} values displayed in each subplot. Mean ratios were chosen rather than standard linear fits to avoid biasing the plotted lines towards the highest measured values, and because averaging to 1 hour effectively removed the random noise at the lowest measured values (e.g. as can be seen in Fig. S9)."

19.) Equation A11 takes the probably, R , which is an average of the s and p polarizations (see Eq A9), and takes it to the power of the number of estimated reflections for a light ray to exit the glass tube. As s and p polarizations have significantly different reflection probabilities as a function of incident angle, equation A11 is an approximation. Ideally, one would calculate R_s and R_p separately for the two polarizations and then average. Not sure how much of a difference this would make, but it would be more accurate.

We tested this by calculating light collection efficiency function curves with the reflection probabilities for the different polarization states averaged before and after the raising to the power of the number of reflections. The result is displayed in the following figure. It is seen that averaging before or after has no noticeable different on the calculated efficiency curves (the 2 curves overlap very closely). Therefore, we decided to make no changes to Eq. (A11).



Referee #2: Anonymous

General comments:

This study provides a detailed characterization and thorough discussion on the performance of commercialized CAPS PM_{2.5} in determining aerosol light absorption using the extinction minus scattering method and the associated uncertainties with this method. While the paper is well written in many aspects, I am a bit concerned that the key points and many useful information may be missed by the readers because of the extraordinary length of the manuscript. In general, I feel that Section 2 and 3 can be combined and shortened to make the experimental method clearer and to the point. Specifically, in section 2.1 and 2.2, Onasch (2015) was heavily cited and many of the description of the instrument is repetitive. Section 2.3 and the sub-sections of Sect 2.2 are also mixed with extensive discussion, which can either to be moved to later sections or to Appendix. Also, the results from the Bologna campaign seems less relevant to the main focus of this study: identifying and quantifying uncertainties in the absorption measurement due to the scattered light truncation effect and cross calibration constant. I recommend publication of this manuscript after the above points are considered.

We thank the reviewer for his/her review and comments regarding the manuscript length. While we acknowledge that the manuscript is lengthy, we believe that this is necessary since it is only by considering all the specific aspects of the instrument and data processing that one is able to fully understand the experimental results displayed in Sections 4, 5 and 6.

Section 2.1 indeed contains some instrument details that are already reported by Onasch et al., (2015). However, we have endeavored to restrict this information to only those details that are pertinent to understanding this study. Since this Section only consists of 4 short paragraphs, we believe there is not much to be gained by further cutting it down. In contrast, retaining these details makes the manuscript self-contained and prevents readers from constantly having to switch between 2 different papers.

Regarding Sections 2.2. and 2.3, most of these details have not been previously discussed at length before in the literature and, as mentioned, they are necessary to understand the experimental results that follow. Therefore, we believe the extensive discussion in these sections is necessary.

Finally, although the Bologna result cannot be interpreted as quantitatively as the rest of the results we believe they are extremely important to show. They demonstrate a key advantage of the CAPS PM_{ss}a relative to filter-based absorption photometers: the ability to measure at very high time resolution. This key advantage has not been demonstrated extensively in the literature yet, which is why we believe the results are pertinent and worth including in this paper.

Detailed characterization of the CAPS single scattering albedo monitor (CAPS PMssa) as a field-deployable instrument for measuring aerosol light absorption with the extinction-minus-scattering method

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Abstract. The CAPS PMssa monitor is a recently commercialized instrument designed to measure aerosol single scattering albedo (SSA) with high accuracy (Onasch et al., 2015). The underlying extinction and scattering coefficient measurements made by the instrument also allow calculation of aerosol absorption coefficients via the extinction-minus-scattering (EMS) method. Care must be taken with EMS measurements due to the occurrence of large subtractive error amplification, especially
20 for the predominantly scattering aerosols that are typically found in the ambient atmosphere. Practically this means that although the CAPS PMssa can measure scattering and extinction coefficients with high accuracy (errors on the order of 1 – 10%), the corresponding errors in EMS-derived absorption range from ~10% to greater than 100%. Therefore, we examine the individual error sources in detail with the goal of constraining these as tightly as possible.

25 Our main focus is on the correction of the scattered light truncation effect (i.e., accounting for the near-forward and -backward scattered light that is undetectable by the instrument), which we show to be the main source of underlying error in atmospheric applications. We introduce a new, modular framework for performing the truncation correction calculation that enables the consideration of additional physical processes such as reflection from the instrument's glass sampling tube, which was neglected in an earlier truncation model. We validate the truncation calculations against comprehensive laboratory
30 measurements. It is demonstrated that the process of glass tube reflection must be considered in the truncation calculation, but that uncertainty still remains regarding the effective length of the optical cavity. Another important source of uncertainty is the cross calibration constant that quantitatively links the scattering coefficient measured by the instrument to its extinction

coefficient. We present measurements of this constant over a period of ~5 months that demonstrate that the uncertainty in this parameter is very well constrained for some instrument units (2 – 3%), but higher for others.

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We then use two example field datasets to demonstrate and summarize the potential and the limitations of using the CAPS PM_{ss} for measuring absorption. The first example uses mobile measurements on a highway road to highlight the excellent responsiveness and sensitivity of the instrument, which enables much higher time resolution measurements of relative absorption than is possible with filter-based instruments. The second example from a stationary field site (Cabauw, the Netherlands) demonstrates how truncation-related uncertainties can lead to large biases in EMS-derived absolute absorption coefficients. Nevertheless, we use a subset of fine-mode dominated aerosols from the dataset to show that under certain conditions and despite the remaining truncation uncertainties, the CAPS PM_{ss} can still provide consistent EMS-derived absorption measurements, even for atmospheric aerosols with high SSA. Finally, we present a detailed list of recommendations for future studies that use the CAPS PM_{ss} to measure absorption with the EMS method. These recommendations could also be followed to obtain accurate measurements (i.e., errors less than 5 – 10%) of SSA, and scattering and extinction coefficients with the instrument.

1. Introduction

Light absorbing aerosols such as black carbon (BC; Bond et al., 2013), brown carbon (BrC; Laskin et al., 2015), tar balls (Corbin and Gysel-Beer, 2019), anthropogenic iron oxide (Moteki et al., 2017), and mineral dust (Sokolik and Toon, 1999) redistribute radiant energy in the Earth’s atmosphere as heat. This perturbs the Earth’s radiative balance directly (Haywood and Shine, 1995), and semi-directly through alteration of atmospheric circulation and cloud cover (Koch and Del Genio, 2010). Currently, large discrepancies exist between global climate model simulations of column-integrated aerosol absorption (absorbing aerosol optical depth, AAOD) and sun photometer measurements of the same quantity taken within the AERONET network (Bond et al., 2013; Samset et al., 2018). The uncertainty resulting from this discrepancy feeds into radiative forcing estimates for absorbing aerosols, contributing to the large and stubborn uncertainty in quantitative estimates of aerosol-radiation climate effects (Myhre et al., 2013). One element that is required to improve this situation and validate both the model simulations and sun photometer measurements is accurate and wide-spread measurements of atmospheric aerosol absorption coefficients (b_{abs}). This activity requires sensitive, field-deployable and robust in situ aerosol instrumentation for measuring absorption (Cappa et al., 2016; Lack et al., 2014; Moosmüller et al., 2009).

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Traditionally, aerosol light absorption has been ~~measured-derived~~ by ~~detecting-measuring~~ the attenuation of light transmitted through aerosol samples deposited on filter substrates (e.g. Rosen et al., 1978). A number of online (i.e., continuously measuring), field-deployable instruments have been developed based on this principle, including the aethalometer (Hansen et al., 1984), the particle soot absorption photometer (PSAP; Bond et al., 1999), and the continuous light absorption photometer

65 (CLAP; Ogren et al., 2017). An important further development of this class of instruments is the multi-angle absorption
photometer (MAAP; Petzold and Schönlinner, 2004), which additionally measures the light backscattered from aerosol-laden
filter samples at two separate angles and processes the resulting measurements with a simplified radiative transfer model in
order to improve the accuracy of the retrieved aerosol absorption coefficients. Collectively, these instruments are referred to
as ‘filter-based absorption photometers’.

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While the popularity of filter-based absorption photometers has provided critical insights into the optical properties of
atmospheric aerosols over the last decades, the limitations of the technique are becoming more problematic as research efforts
progress even further. Filter-based light absorption measurements are subject to large positive artefacts due to the effects of
multiple scattering from the filter material and the deposited particles, and they are sensitive to aerosol loading, humidity and
75 aerosol single scattering albedo, SSA (Moosmüller et al., 2009). An additional concern is that the commercial production of
some important filter-based instruments has recently been discontinued (e.g. the PSAP by Radiance Research, and the MAAP
by Thermo Fisher Scientific).

Motivated by the limitations in the filter-based techniques, instrumentation development efforts have recently focused on
80 methods for measuring light absorption by aerosols in their natural, suspended state. These techniques include photoacoustic
spectroscopy (Arnott et al., 1999; Lack et al., 2006), photo-thermal interferometry (Moosmüller and Arnott, 1996; Sedlacek,
2006), and extinction-minus-scattering (EMS) methods. Here we focus on the EMS method. The EMS method is comprised
of two separate underlying measurements: one of the aerosol extinction (b_{ext}) and one of the aerosol scattering coefficient (b_{sca}).
The aerosol absorption coefficient b_{abs} is then obtained by subtracting b_{sca} from b_{ext} :

85

$$b_{abs} = b_{ext} - b_{sca} \quad . \quad (1)$$

Traditionally, EMS measurements have been performed by two separate instruments (e.g. an integrating nephelometer for
aerosol scattering, and a separate extinction monitor). Additionally, the use of EMS measurements has mostly been limited to
the laboratory where high absorption signals are easily achievable, and artefacts (e.g., due to the scattered light truncation
90 effect) can be avoided. In such a laboratory setting, EMS measurements are considered as a primary standard for measuring
aerosol absorption thanks to the traceability of the underlying b_{ext} and b_{sca} measurements (e.g. Bond et al., 1999; Schnaiter et
al., 2003; Virkkula et al., 2005).

The continued development of sensitive techniques for measuring b_{ext} using multi-pass optical cavities (e.g. cavity ring-down
95 spectroscopy, Moosmüller et al., 2005; and cavity attenuated phase shift spectroscopy, CAPS, Keabian et al., 2007) has
created the possibility of extending application of the EMS technique more broadly to different types of atmospheric and/or

test bench (i.e., emissions) measurements. This endeavor poses several challenges: i) subtractive error amplification in EMS-derived b_{abs} can become very large when b_{sca} is close to b_{ext} (i.e., as $SSA \rightarrow 1$), which occurs very commonly throughout the Earth's atmosphere (Dubovik et al., 2002), ii) artefacts such as the scattered light truncation effect in integrating nephelometer measurements of b_{sca} are generally unavoidable and more difficult to quantify for ambient aerosols (which are typically complex mixtures of particles of varying size, composition and morphology), iii) it is more difficult to ensure thorough and regular instrument calibrations in a field versus a laboratory setting, and iv) it is usually more difficult to control sampling arrangements in the field to ensure that b_{ext} and b_{sca} are measured under the same (or at least well-known) environmental conditions.

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Despite these challenges, the possibility of performing EMS measurements of atmospheric aerosol absorption has recently been boosted by the development and commercialization of the cavity attenuated phase shift SSA monitor (CAPS PMssa) by Aerodyne Research Inc. (Billerica, MA, USA; Onasch et al., 2015). The CAPS PMssa monitor combines measurements of b_{ext} and b_{sca} in a single instrument and sample volume, following in the tradition of earlier combined extinction-scattering instruments (Gerber, 1979; Sanford et al., 2008; Strawa et al., 2003; Thompson et al., 2008). Its direct precursor instrument – the Aerodyne CAPS extinction monitor (CAPS PMex) – uses the CAPS technique to measure b_{ext} values with high sensitivity in a compact optical cavity and overall instrument unit (Massoli et al., 2010; Petzold et al., 2013). The CAPS PMssa is based on the same optical cavity, but additionally includes an integrating sphere reciprocal nephelometer around the cavity for measurement of b_{sca} .

115
The CAPS PMssa was originally designed to measure SSA (i.e., the ratio of b_{sca} to b_{ext}), a quantity which is not subject to the same subtractive errors as b_{abs} . However, its design addresses two of the key challenges of atmospheric EMS measurements that were listed in the paragraph above, which makes it an attractive candidate for performing such measurements. Specifically, by simultaneously measuring b_{ext} and b_{sca} for the same volume of air, there is no need to account for possible differences in environmental conditions or sampling losses that could affect these two coefficients. Additionally, this feature allows the cross calibration of one coefficient against the other using white test aerosols (non-absorbing, i.e., where $b_{ext} = b_{sca}$), which facilitates the development of relatively simple field calibration procedures (in practice, b_{sca} is cross calibrated against b_{ext} in the CAPS PMssa). Nevertheless, great care must still be taken when performing EMS measurements with the CAPS PMssa to ensure that errors in the underlying b_{ext} and b_{sca} measurements are minimized and that very large subtractive error amplification is avoided. This essentially reduces down to the following problem: errors that may be acceptable if one is interested in measuring b_{ext} , b_{sca} , or SSA (say on the order of 5 – 10%), are substantially magnified – perhaps to over 100%, as we will show below – when using the very same measurements to derive b_{abs} . Therefore, the user must be concerned about errors on the order of only a few percent if they wish to use the CAPS PMssa to reliably measure atmospheric aerosol absorption coefficients.

130 One of the key sources of uncertainty that must be considered for the CAPS PM_{ss}a (and integrating nephelometry in general) is the scattered light truncation effect (e.g. Moosmüller and Arnott, 2003; Varma et al., 2003). Integrating nephelometers seek to detect light scattered in all possible directions. In reality, a fraction of near-forward and near-backward scattered light is always lost due to unavoidable physical design limitations. As a result, b_{sca} measurements are biased low and need to be corrected. The required correction factor depends on a particular instrument's geometry, as well as the angular distribution of
135 light scattered from an aerosol sample, which is a function of the optical wavelength, and the size distribution, composition, mixing state, and morphology of the particles in that sample.

Onasch et al. (2015) presented a simple model for calculating truncation correction factors for the CAPS PM_{ss}a based on Mie theory calculations with inputted particle size distributions. However, this model does not consider an important physical
140 process that serves to increase scattered light truncation: reflection of scattered light from the inner surface of the glass sampling tube within the integrating nephelometer. Liu et al. (2018) developed a more sophisticated truncation model based on solution of the radiative transfer equation (RTE) configured specifically to the PM_{ss}a optical system. As well as allowing for the treatment of non-spherical particles (which is not possible with Mie theory), the RTE approach also allows for the treatment of additional physical processes (e.g. multiple scattering from the aerosol and glass tube reflection). CAPS PM_{ss}a
145 truncation values calculated with these models have so far been validated against only a limited dataset of experimental measurements (Onasch et al., 2015). Furthermore, there is a lack of systematic analyses that aim to determine the sensitivity of EMS-derived b_{abs} values to changes in calculated truncation (e.g. for ambient aerosol samples).

Despite the many unresolved uncertainties, the CAPS PM_{ss}a has already been used as an instrument for measuring b_{abs} in a
150 number of different ambient field campaigns (Chen et al., 2018; Han et al., 2017; Xie et al., 2018), emissions testing experiments (Corbin et al., 2018; Zhai et al., 2017), and soot characterization experiments (Dastanpour et al., 2017; Forestieri et al., 2018; Perim de Faria et al., 2019).

In this study, we present a compilation of theoretical calculations, novel laboratory measurements, and example field
155 applications that all serve a common purpose: to improve the truncation correction approach and to determine the extent to which the CAPS PM_{ss}a can be used to measure aerosol absorption coefficients via the EMS method.

In Sect. 2 we present a theoretical description of the instrument, including the introduction of a new truncation model that includes the process of glass tube reflection and is suitable for application to large field datasets. This Section culminates in
160 the presentation of a detailed b_{abs} error model, which is used to demonstrate why it is so critical to constrain errors in the truncation calculations and instrument cross calibration constant. This finding motivates the experimental work described in the remainder of the paper. Section 3 details the experimental methods used. Section 4 then presents some regular measurements of the CAPS PM_{ss}a cross calibration constant in order to assess its precision and stability. In Sect. 5 we compare

the results of novel and comprehensive laboratory truncation measurements with calculated values from a range of different
165 truncation models. Synthesizing all of these issues together, Sect. 6 then presents two example field datasets that demonstrate
both the potential and the limitations of using the CAPS PMssa to measure atmospheric aerosol absorption. Finally, in the
concluding Sect. 7 we present a list of recommendations for future CAPS PMssa studies.

2. Theoretical description of the CAPS PMssa monitor

2.1 General introduction

170 The CAPS PMssa monitor is described in detail previously in the original technical paper by Onasch et al. (2015). A schematic
diagram of the instrument is shown in Fig. 1. Briefly, the instrument consists of an optical cavity formed by two high
reflectivity mirrors (reflectivity ~ 0.9998), creating a long effective optical path length ($\sim 1 - 2$ km). Aerosol samples are
drawn continuously through this cavity at a flow rate of 0.85 litres per minute (light blue arrows in Fig. 1) with no size selection
performed at the instrument inlet, meaning that the samples generally contain both sub- and supermicrometer particles.
175 Smaller, particle-free purge flows of ~ 0.025 litres per minute are ~~drawn~~ pushed continuously over the high reflectivity mirrors
to prevent their contamination (green arrows in Fig. 1). The purge and sample flows are generated from the same double-
headed membrane pump.

The input light source to the cavity is provided by a single light emitting diode (LED). Units are available from the
180 manufacturer Aerodyne Research, Inc. with LEDs centred at wavelengths of 450, 530, 630, 660 and 780 nm. The intensity of
the LED input light is square-wave modulated (typically at 17 kHz), and the intensity of light leaking through one mirror is
monitored by a vacuum photodiode or, in the case of the 780 nm unit, a photomultiplier tube (PMT). The intensity of the light
circulating in the cavity increases exponentially during the LED on-phase and decreases exponentially during the LED off-
phase, with a timescale dependent on the reflectivity of the mirrors and optical loss in the cell (Lewis et al., 2004). The
185 introduction of a scattering or absorbing species to the cell enhances this optical loss, resulting in a shorter optical lifetime in
the cavity and a phase shift of the output signal relative to the input signal. This phase shift is measured by the vacuum
photodiode using a quadrature signal integration method (Kebabian et al., 2007). This is the technique for measuring extinction
coefficients known as cavity attenuated phase shift spectroscopy (CAPS) and its application in the CAPS PMssa is referred to
as the ‘extinction channel’ of the instrument.

190 The second light detector in the instrument is a ~~photomultiplier tube (PMT)~~ that is used to measure the integrated aerosol
scattering coefficient (Fig. 1). It is referred to as the ‘scattering channel’ of the instrument. The PMT is placed on the integrating
sphere that surrounds the centre of the optical cavity. The integrating sphere has an inner diameter of 10 cm. The inside of the
integrating sphere is coated white to form a Lambertian reflector (reflectivity = 0.98), which functions to maximize the amount
195 of scattered light detected by the PMT and to minimize any bias between light collected from different scattering angles.

Onasch et al. (2015) calculated that the variation in the angular sensitivity of the sphere as a function of scattering angle is less than 1%. The integrating sphere does not contain a baffle as described by Onasch et al. (2015). A glass tube with inner diameter of 1 cm passes through the centre of the integrating sphere in order to encapsulate the aerosol flow along the central axis of the optical cavity.

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In this study we define the central axis of the optical cavity as the z -dimension and the centre of the integrating sphere to be at position $z = 0$ cm. A particle lying along the central z -axis scatters light in polar directions at scattering angles θ defined with respect to the z -axis (two limiting examples for forward- (θ_1) and back-scattered (θ_2) light are shown in Fig. 1), and azimuthal directions at scattering angles φ (not shown in Fig. 1).

205

2.2 Data processing and important correction and calibration factors

The data processing chain applied by the CAPS PM_{ss}a instrument firmware to calculate aerosol extinction and scattering coefficients from the measured photodiode and PMT signals is displayed in Fig. 2 (Onasch et al., 2015). The instrument has two modes of operation where data are collected: sample and baseline measurements. The sample and baseline measurements

210 are achieved by a controlled three-way valve that directs the sampled air either directly into the optical cavity or first through a filter that removes all particles. The instrument firmware allows the baseline measurements to be repeated automatically at a frequency and duration set by the user. Typically during field operation baseline measurements are performed for 1 minute every 5 or 10 minutes.

215 In the extinction channel, the sample and baseline measurements are first treated by subtracting out a constant factor that accounts for extinction due to Rayleigh light scattering from the aerosol carrier gas. The subtraction term is corrected using temperature and pressure measurements taken by the instrument to account for possible variations in these quantities between sample and baseline periods.

220 Full treatment of the PMT scattering signals is given by Onasch et al. (2015). The scattering signals are counted during the LED off-phase when only highly collimated light is circulating in the cavity in order to minimize the contribution of light scattered from interior surfaces of the instrument. Consequently, the average intensity of circulating light during the LED off-phase must be accounted for in the scattering calculation, as illustrated by the dot-dashed lines in Fig. 2 and described in detail in Onasch et al. (2015).

225

Following these initial data treatment steps, uncorrected aerosol extinction and uncalibrated scattering coefficients ($b_{ext,uncorr.}$ and $b_{sca,uncalib.}$, respectively) are obtained by taking the difference between the sample-mode coefficient measurements

(which we term $b_{ext,sample}$ and $b_{sca,sample}$) and the interpolated baseline-mode coefficient measurements ($b_{ext,baseline}$ and $b_{sca,baseline}$). By default, the instrument firmware uses a step function to interpolate the baseline values between each baseline period (i.e., the mean value of a baseline period is assumed to stay constant until it is replaced by the mean value of the next baseline period). However, the data output files from the instrument also provide sufficient information for the user to apply custom methods for calculating the interpolated coefficients $b_{ext,baseline}$ and $b_{sca,baseline}$ (e.g. linear or cubic spline interpolation; Pfeifer et al., 2020).

Following the sample-baseline difference calculations, one extinction correction factor (the geometry correction factor, α) and two scattering correction factors (cross calibration, β , and truncation factors, γ) are multiplicatively applied to the respective signals in order to obtain the calibrated and corrected aerosol coefficients b_{ext} and b_{sca} . The α and β factors are applied automatically by the instrument firmware, while γ must be applied manually by the user in post-processing. All three correction factors are discussed in detail in the Sections below. The aerosol absorption coefficient is then obtained as:

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$$b_{abs} = b_{ext} - b_{sca} = \frac{1}{\alpha} \cdot (b_{ext,sample} - b_{ext,baseline}) - \frac{\gamma}{\beta} \cdot (b_{sca,sample} - b_{sca,baseline}) \quad . \quad (2)$$

2.2.1 Geometry correction factor (α)

The purge flows that protect the high reflectivity mirrors in the CAPS PMssa shorten the effective optical path length of the cavity and may slightly dilute the instrument sample flow at the cavity inlet. Therefore, a correction factor must be applied to the measured extinction coefficients in order to account for these changes (Massoli et al., 2010; Onasch et al., 2015; Petzold et al., 2013). We refer to this correction factor as the geometry correction factor, α , which can be determined by external calibration, i.e., by comparing CAPS PMssa measurements against independently measured or calculated b_{ext} (e.g. Mie-calculated b_{ext} values for spherical, monodisperse test aerosols, Petzold et al., 2013; or measured b_{ext} values for non-absorbing test aerosols obtained with a reference nephelometer, Pfeifer et al., 2020).

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Onasch et al. (2015) applied the Mie calculation approach to measurements of polystyrene latex (PSL) spheres of varying diameter to determine an α value of 0.73 for a CAPS PMssa unit operating at 630 nm. This is lower than the general value of 0.79 quoted by Onasch et al. (2015) for CAPS PMex monitors, which they note is expected due to small differences in the cavity geometries. The CAPS PMssa units used in this study (Table 2) participated in European Center for Aerosol Calibration (ECAC; <http://www.actris-ecac.eu/>) workshops (CAPS630b in August 2016; and CAPS450, CAPS630a, CAPS780 in January 2017) where their geometry correction factors were determined against reference instrumentation (CAPS PMex, nephelometer) using ammonium sulphate test aerosols. The units were determined to have α values of 0.78 (CAPS450), 0.71 (CAPS630a),

0.7 to 0.73 (CAPS630b), and 0.78 (CAPS780). Therefore, it appears that α is instrument-unit-dependent. The stability of α over time is still an open question. However, regular and frequent measurements of α in CAPS PMex monitors performed at the ECAC suggests that it does not drift by more than 3% over the period of a year. By default, the CAPS PMssa firmware automatically applies an α factor of 0.73 to calculate b_{ext} (Fig. 2).

2.2.2 Scattering cross calibration factor (β)

The scattering cross calibration factor (β) is used to relate the PMT-measured scattering signal of the CAPS PMssa to an absolute aerosol scattering coefficient. The value of β can be determined by cross calibrating the uncalibrated aerosol scattering coefficient $b_{sca,uncalib}$ against b_{ext} measured by the extinction channel (Onasch et al., 2015). This approach is possible because the scattering and extinction coefficients are measured simultaneously for the same air sample, and b_{ext} measured using the CAPS method is effectively ‘calibration free’ (apart from the geometry correction factor, as discussed in Sect. 2.2.1, as well as potential non-linearities at high baseline losses). Amongst other factors, β depends on the PMT detector response, which can vary over time. Therefore, regular cross calibrations should be performed.

Non-absorbing test samples are required to perform the cross calibration and to determine a value for β (i.e., purely scattering samples for which $b_{ext} = b_{sca}$, or SSA = 1). In principle, the calibration can be performed with gases or aerosol particles. In practice, we performed all calibrations in the present study with particles because readily available calibration gases such as ~~N₂~~ and CO₂ span a much smaller range in b_{sca} than is achievable with aerosols of different concentrations, additional corrections are required to account for the changes in optical path length and dilution with the purge flows for different gases (see Sect. 2.2.1), and we have observed that the instrument can take a long time (~hours) to adjust and stabilize when filled with different gases (as expected due to the low flows and large filter areas in the purge flow setup).

When using the particle-based calibration method, non-absorbing aerosol particles with size parameters x in the Rayleigh light scattering regime should be used to ensure well-defined scattered light truncation, since the scattering phase function is independent of particle size in the Rayleigh regime. We term cross calibration constants derived in this specific manner as $\beta_{Rayleigh}$. The size parameter x relates the aerosol particle diameter D_p to the wavelength of light λ through the expression $\pi D_p / \lambda$. The Rayleigh regime is defined by the condition $x \ll 1$. In practice, there is a trade-off between selecting particle sizes that are small enough to lie within or near the Rayleigh regime limit but large enough to generate scattering and extinction signals with sufficiently high signal-to-noise ratios. This means particles with diameter less than approximately 150 nm should be used to determine $\beta_{Rayleigh}$ in the 450 nm CAPS PMssa, while slightly larger particles (e.g. $D_p \sim 200$ nm) can be used with 630 or 780 nm CAPS PMssa instruments.

Formally, the Rayleigh-regime, particle-based cross calibration approach can be expressed as:

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$$\beta_{Rayleigh} = \frac{b_{sca,uncalib.}^{non-abs,Rayleigh}}{b_{ext}^{non-abs,Rayleigh}} = \alpha \cdot \left(\frac{b_{sca,uncalib.}^{non-abs,Rayleigh}}{b_{ext,uncorr.}^{non-abs,Rayleigh}} \right) = \alpha \beta'_{Rayleigh} \quad (3)$$

where $b_{ext}^{non-abs,Rayleigh}$ and $b_{sca,uncalib.}^{non-abs,Rayleigh}$ are the extinction and uncalibrated scattering coefficients, respectively, for a population of non-absorbing particles with size parameters in the Rayleigh regime. The right-hand side of Eq. (3) is obtained by substitution of the relationship $b_{ext} = 1/\alpha b_{ext,uncorr.}$ into the left-hand side ratio. From this substitution, it can be seen that $\beta_{Rayleigh}$ (and β , generally) is directly proportional to the geometry correction factor α , which is required to measure b_{ext} accurately as discussed in Sect. 2.2.1 (the remaining fraction of the cross calibration constant is termed $\beta'_{Rayleigh}$ to distinguish it from $\beta_{Rayleigh}$). Thus, Eq. (3) demonstrates how the cross calibration approach quantitatively links b_{sca} to b_{ext} in the CAPS PMssa. Following application of $\beta_{Rayleigh}$, we refer to the calibrated aerosol scattering coefficient corrected for the truncation of Rayleigh scattered light as $b_{sca,Rayleigh}$. This is to recognize the fact that the cross calibration approach represented by Eq. (3) implicitly corrects for the truncation of light scattered from the calibration aerosol, which has been chosen specifically to have the well-defined phase function corresponding to Rayleigh light scattering.

Onasch et al. (2015), demonstrated that the linearity shown by Eq. (3) is valid up to extinction coefficients of $\sim 1000 \text{ Mm}^{-1}$, which is higher than typical ambient aerosol extinction coefficients, excluding perhaps coefficients in heavily polluted urban environments. The precise limit of linearity should be examined for individual instrument units if it is relevant for a particular experiment. For very high aerosol loadings above the limit of linearity the CAPS PMssa cross calibration approach can still be used. However, this requires the addition of empirically-derived higher order terms in $b_{ext}^{non-abs,Rayleigh}$ to Eq. (3). In addition, the potential occurrence of multiple scattering effects needs to be considered at very high aerosol loadings (Wind and Szymanski, 2002).

2.2.3 Truncation correction factor (γ)

The final quantitative correction factor that must be applied to the scattering coefficients measured with the CAPS PMssa is the truncation correction factor, γ . The truncation correction factor γ is applied to $b_{sca,Rayleigh}$ to compensate for the light scattered in near-forward and near-backward directions that is not measured by the instrument due to geometric restrictions. The truncation correction factor γ depends on both the instrument properties as well as the angular distribution of light scattered from the aerosol sample being measured (referred to in short as the ensemble scattering phase function, S_p), which depends on the aerosol size distribution, morphology, mixing state, and composition (refractive indices). The existing methods for calculating the CAPS PMssa truncation correction factor γ either do not include the process of scattered light reflection from the inner surface of glass sampling tube (Onasch et al., 2015), or are computationally expensive (Liu et al., 2018) and not well-

suiting to calculate time-resolved truncation factors for large datasets (e.g. as required for the example Cabauw dataset in Sect. 6.2). Therefore, we present here a new truncation calculation framework that overcomes both of these limitations.

The new calculation framework is presented visually as a flow chart in Fig. 3. The full set of details and equations is given in Appendix 1. Briefly, we define γ as the normalized ratio of the true integrated scattering coefficient, $b_{sca,true}$ to the truncation-affected scattering coefficient that is actually accessible to measurement, $b_{sca,meas}$. The true scattering coefficient $b_{sca,true}$ represents the coefficient that would be measured by an ideal integrating nephelometer capable of collecting light scattered in all possible directions. The ratio requires normalization by a factor $k_{Rayleigh}$ to represent the fact that some scattered light truncation is already included implicitly in the cross calibration constant, due to the way in which it is measured. For the recommended case of cross calibration with Rayleigh scatterers according Eq. (3), $k_{Rayleigh}$ represents the truncation of the Rayleigh scattered light from the calibration aerosol. That is,

$$\gamma = \frac{b_{sca,true}}{b_{sca,meas}} \cdot k_{Rayleigh} = \frac{b_{sca,true}}{b_{sca,meas}} \cdot \left(\frac{b_{sca,meas}^{Rayleigh}}{b_{sca,true}^{Rayleigh}} \right). \quad (4)$$

Defined in this manner, γ equals 1 for aerosols in the Rayleigh regime. For aerosols containing larger particles or non-spherical particles that produce more forward-focused light scattering, γ is always greater than 1.

The equations for calculating the integrated scattering coefficients in Eq. (4) are detailed in Appendix A1. These equations have been given in several previous publications (Anderson et al., 1996; Heintzenberg and Charlson, 1996; Moosmüller and Arnott, 2003; Müller et al., 2011b; Peñaloza M, 1999). The novel aspect of our formulation is that we explicitly define a function representing the efficiency with which an integrating nephelometer is able to collect scattered light, $\eta(\theta, \lambda)$, which is a simple function varying between 0 and 1. Values of 0 indicate that a nephelometer collects no light of wavelength λ at some scattering angle θ , while values of 1 indicate that a nephelometer collects all the light scattered at angle θ . Considering $\eta(\theta, \lambda)$ explicitly has a number of advantages: i) it allows transparent representation of an instrument's truncation angles (i.e., by setting η equal to 1 between two truncation angles, and 0 beyond them), (ii) it allows for the simple and explicit introduction of additional physical processes into light scattering calculations (e.g. reflection from the glass sampling tube can be considered by combining the Fresnel equation for reflection probability with η , as shown in Appendix A1), (iii) it provides a clear and intuitive way to compare the abilities of different nephelometers to collect scattered light, and (iv) it emphasizes the modular nature of the truncation calculation.

One of the important characteristics of integrating sphere type reciprocal nephelometers like the CAPS PM_{ss} is that truncation is a function of position along the central axis of the optical cavity (which we denote as the z -dimension, Fig. 1). This

350 characteristic is represented by the small subplots in Fig. 3 that show light collection efficiency curves (termed η_{spot} in Appendix A1) for six different z positions in the CAPS PMssa, including for two positions at 1 cm outside of the integrating sphere (i.e., $z = -6$ and 6 cm). Positions outside of the integrating sphere must be considered since it is possible for particles outside the sphere to scatter light into the sphere (e.g. Varma et al., 2003), even if only through a narrow range of scattering angles. We term the extra length that needs to be considered outside the sphere's boundaries as the l parameter. The geometrical
355 limits for the l parameter are 0 (i.e., no extra path length considered) and 4.7 cm (the distance between the integrating sphere and the sample inlet and outlet ports to the optical cavity). Onasch et al. (2015) and Liu et al. (2018) both used $l = 1$ cm in their calculations (i.e., they considered a z -range from -6 to 6 cm). The η_{spot} subplots in Fig. 3 also demonstrate the effect of glass tube reflection: between a sphere's truncation angles, reflection decreases the probability of light collection from 1 to some value less than 1. Therefore, glass tube inner surface reflection serves to increase scattered light truncation. A single, integrated
360 light collection efficiency function for the CAPS PMssa can be generated by integrating η_{spot} over all possible z positions (Eq. A13). CAPS PMssa integrated η functions are shown in Fig. 3 for the two cases of without and with glass reflection.

It is important to stress the implications of the modularity of truncation calculation. This modularity means that once the $\eta(\theta, \lambda)$ and angular sensitivity functions are known for a particular instrument, they can be combined with any measured or
365 calculated ensemble scattering phase function in order to calculate γ . In the present study, we used Mie theory and co-located particle size distribution measurements to efficiently calculate hourly-resolved S_p functions and γ values for a month-long field campaign (Sect. 6.2; Fig. S12). This Mie calculation method assumes spherical, homogeneous particles. If one wished to consider more complex particle morphologies, a more sophisticated optical model could be used to calculate the scattering phase functions S_p . Or, if co-located polar nephelometer measurements of the scattering phase function were available (e.g.
370 Espinosa et al., 2018), these could be input directly into the truncation calculation.

2.3 Absorption error model for the CAPS PMssa and discussion of the sources and effects of uncertainties in β and γ

It is critical to carefully consider and understand the sources of errors in EMS-derived b_{abs} values, since these can be very large when taking the difference of two potentially larger numbers – b_{sca} and b_{ext} – that each carry their own uncertainties. Based on the data processing framework presented in the previous Sect. 2.2, an error model can be constructed for CAPS PMssa
375 absorption coefficients by considering the uncertainty in each of the individual parameters in the right-hand side of Eq. (2) and applying the standard rules of error propagation, including consideration of potential covariance of the errors in b_{sca} and b_{ext} . The explicit equations for such a model are given in Appendix A2. Table 1 lists the individual parameters in the error model along with realistic estimates of their uncertainties. In general, we consider two sources of uncertainties: uncertainty due to the limited precision with which a particular parameter can be determined during calibration or measurement, and uncertainty due
380 to possible drift of a parameter between available calibrations or measurements (e.g. baseline drift between two subsequent baseline measurements). For a given parameter, these two sources of errors are independent and they can be added in

quadrature, or if one of the errors is much larger than the other, this larger error can simply be used in error propagation calculations.

385 Many of the individual uncertainty estimates given in Table 1 are taken from previous studies and will not be discussed in great detail here. However, the uncertainties in the b_{sca} correction factors γ and β are still poorly constrained and require further investigation. We refer to these uncertainties as $\delta\gamma$ and $\delta\beta$, respectively. Onasch et al. (2015) showed that β can be measured with high precision for a 630 nm PM₁₀ unit, but the obtainable precision at other operation wavelengths as well as the stability in β over time have not been fully explored. Therefore, the overall $\delta\beta$ is still not well characterized.

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The uncertainty in γ is more difficult to quantify. At the highest level it can be categorized into uncertainties related to the instrument properties (e.g. should glass tube reflection be considered, and an appropriate l value), and those related to knowledge of the scattering phase functions of the aerosol samples being measured. Regarding uncertainties in the latter category, these can be further characterized depending on how the angularly resolved light scattering information is obtained.

395 In the best case scenario, the scattering phase functions would be obtained directly from co-located polar nephelometer measurements, in which case $\delta\gamma$ would depend on the accuracy of these measurements (and possible extrapolation of those measurements beyond a polar nephelometer's truncation angles). Since polar nephelometer measurements are rarely performed in measurement campaigns, it is more likely that scattering phase functions will be calculated with an optical model (e.g. Mie theory) using co-located size distribution measurements (covering both sub- and supermicrometer size fractions) as input. In this case, $\delta\gamma$ will be a function of the accuracy of the input size distribution measurements, as well as the representativeness of the optical model and its inputs (e.g. complex refractive index, particle morphology if the optical model includes treatment of this). In the worst case scenario, which is expected to occur frequently in field work, there might be no information available to constrain the scattering phase function. In this case, γ values would need to be assumed. For example, a user might simply assume that γ equals 1, which is equivalent to assuming that all particles in the sample are Rayleigh light scatterers. In this case, $\delta\gamma$ should reflect the possible consequences of that assumption. In Table 1 we provide some estimates for both $\delta\gamma$ and $\delta\beta$ that are based on the results of the present study. These estimates and results are discussed in specific detail below in Sections 4, 5, and 6.

For now, we use our error model to assess the possible impacts of $\delta\gamma$ and $\delta\beta$ on the relative uncertainty in EMS-derived b_{abs} , regardless of where the uncertainty in these two parameters actually comes from. Indeed, we generalize this analysis even further by considering the relative uncertainty in the combined b_{sca} correction factor γ/β , given by the equation:

$$\frac{\delta(\gamma/\beta)}{\gamma/\beta} = \sqrt{\left(\frac{\delta\gamma}{\gamma}\right)^2 + \left(\frac{\delta\beta}{\beta}\right)^2} \quad (6)$$

This approach is motivated by the fact that $\delta\gamma$ and $\delta\beta$ have equal impacts on the uncertainty in EMS-derived b_{abs} , and it is
 415 justified because $\delta\gamma$ and $\delta\beta$ are independent of one another.

The relative uncertainty in b_{abs} calculated with our error model can be interpreted as the precision with which b_{abs} can
 theoretically be determined for a given set of error model inputs. It should be noted that in addition to this precision-based
 uncertainty, the absolute accuracy of b_{abs} will also depend directly on the accuracy of the geometry correction factor α if the
 420 instrument is cross calibrated as recommended in Sect. 2.2.2. This is because in the same manner as with b_{sca} , the cross
 calibration serves to define b_{abs} with respect to α , which can be seen by substituting the right-hand side of Eq. (3) into Eq.
 (2):

$$b_{abs} = \frac{1}{\alpha} \cdot \left(b_{ext,uncalib.} - \frac{\gamma}{\beta'_{Rayleigh}} \cdot b_{sca,uncalib.} \right) . \quad (7)$$

In the present study we do not explicitly consider the α -related uncertainty in b_{abs} , though it is important to keep this in mind.
 Specifically we note that the errors in α cause covariant errors in b_{ext} and b_{sca} . Hence, the relative error in α propagates 1-to-1
 to corresponding relative error in EMS-derived b_{abs} , independently of SSA. This is not the case for errors in $\beta'_{Rayleigh}$, for
 example, which lead to error in b_{sca} that is independent of errors in b_{ext} , and therefore relative errors in b_{abs} that do vary with
 SSA. In practice, the uncertainty due to α can only be determined by comparison of CAPS PM_{ss}a measurements against an
 430 independent reference. It is also worthwhile to note that the uncertainty in SSA measured by CAPS PM_{ss}a does not depend
 on the uncertainty in α , since this factor simply cancels out when taking the ratio of b_{sca} to b_{ext} . This is one of the key design
 features of the cross-calibrated instrument (i.e., the relative error in α makes identical and covariant contributions to the
 errors in b_{ext} , b_{sca} , and b_{abs}).

Focusing on the precision-related uncertainty in b_{abs} that is quantified by our error model (Eq. A16), Fig. 4 displays this variable
 as a function of the combined relative uncertainty in β and γ for a range of different atmospheric conditions (two different
 aerosol loadings and 4 different SSA values). The curves in this figure were generated using the following model inputs
 designed to represent the CAPS630b instrument characteristics during the Cabauw field campaign (Sect. 6.2): [$\alpha=0.73$, $\delta\alpha=0$,
 435 $\beta=0.81$, $\gamma=1.04$, $\delta b_{ext,sample} = \delta b_{sca,sample} = 1/\sqrt{3600}$ Mm⁻¹, $b_{ext,baseline}=512$ Mm⁻¹, $\delta b_{ext,baseline}=0.35$ Mm⁻¹, $b_{sca,baseline}=50$ Mm⁻¹,
 440 $\delta b_{sca,baseline}=0.66$ Mm⁻¹]. Parameter $\delta\alpha$ was set to 0 to reflect the fact that the accuracy of α is not considered in the simulation,
 as well as the assumption that α does not vary between subsequent cross calibration measurements. Fig. 4 can be interpreted
 as follows: taking an uncertainty of 5% for γ and 2% for β (which we will show later to be realistic estimates), the relative

uncertainty in the combined b_{sca} correction factor equals 5.4% based on Eq. (6). This example corresponds to vertical blue dashed line in Fig. 4. Two other realistic examples are also shown in the Figure as vertical dashed lines.

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Several important and general features are apparent in Fig. 4. Firstly, it is seen that the precision-related uncertainty in b_{abs} increases dramatically with small increases in uncertainty in either β or γ . As a result, small uncertainties in β or γ can result in large uncertainties in b_{abs} . The relative uncertainty in b_{abs} is also a strong function of SSA due to the large subtractive error amplification that results from taking the difference of two large and uncertain numbers. Taking these two points together and
450 considering the example case demonstrated by the vertical red dashed line, a combined uncertainty of only 10.2% in γ and β leads to precision-related uncertainties in b_{abs} of over 80% at SSA greater than 0.9. Such large SSA is very common for atmospheric aerosols, which highlights why it is so critical to minimize uncertainties in β and γ when using the CAPS PM_{ss} to measure atmospheric aerosol absorption with the EMS method.

455 The divergences between the corresponding dashed and solid grey lines in Fig. 4 represent the effects of the errors in both the extinction and scattering baseline signals. These errors can be important under very clean atmospheric conditions (represented by the case $b_{ext} = 10 \text{ Mm}^{-1}$), since the absolute differences between sample-mode and baseline signals are then small. However, these sources of uncertainty are quickly overwhelmed as uncertainties in β and γ increase, resulting in the convergence of the pairs of dashed and solid grey lines moving from left to right across the figure. For the high aerosol load case (represented by
460 $b_{ext} = 100 \text{ Mm}^{-1}$), it is interesting to note that for 0% uncertainty in β and γ , the relative uncertainty in b_{abs} is still SSA dependent, even though b_{sca} has been defined with respect to b_{ext} by the cross calibration and $\delta\alpha$ set to 0 in the simulation. This is because $b_{ext,sample}$ and $b_{sca,sample}$ still carry independent uncertainty due to random noise, even if this is relatively small (i.e., 1 Mm^{-1} at 1 sec temporal resolution).

465 The b_{abs} uncertainty values displayed in Fig. 4 were simulated to represent 1-hour averaged measurements. Fig. S1 indicates that the equivalent values representing 1-minute averaged measurements are practically equivalent to those shown in Fig. 4, while those representing 1-second measurements are only greater for low values of uncertainty in β and γ . This is because of all the uncertainties listed in Table 1, only the uncertainties in $b_{ext,sample}$ and $b_{sca,sample}$ are related to random noise, and hence
470 can be reduced by signal averaging. Since these error components are ~~anyway~~ small relative to the other error components in the model, averaging for 1 min or 1 hour has only a minor effect on the calculated uncertainty in b_{abs} .

3. Experimental methods

3.1 Instrumentation

3.1.1 Instruments for measuring aerosol light absorption and black carbon concentrations

In this Section we detail the experimental methods that we applied to investigate and characterize the ability of the CAPS PM_{ss} to measure atmospheric aerosol absorption coefficients. A total of four different CAPS PM_{ss} monitors were used in this study: one operating at 450 nm, two at 630 nm, and one at 780 nm. The four units are listed in Table 2 along with their relevant specifications.

A multi-angle absorption photometer (MAAP; Thermo Fisher Scientific, Waltham, MA, USA) was used during the Cabauw field campaign (Sect. 3.4.1) to measure absolute aerosol absorption coefficients at a wavelength of 637 nm (Petzold and Schönlinner, 2004). As discussed in the Introduction, the MAAP is a filter-based absorption photometer that incorporates additional measurements of back-scattered light and a two-stream radiative transfer scheme in order to constrain aerosol absorption coefficients more tightly than is possible with simple light attenuation measurements. The MAAP is a well-known and well-characterized instrument for measuring light absorption by atmospheric aerosols. The accuracy of MAAP absorption coefficients was investigated against laboratory reference EMS absorption measurements in the Reno Aerosol Optics Study (RAOS) and the two methods were found to agree within 7% for a range of different black carbon containing aerosols (Petzold et al., 2005). Müller et al. (2011a) demonstrated that the unit-to-unit variability between six different MAAP instruments was less than 5%. These authors also showed that the true operation wavelength of the instrument was 637 nm, not the nominal value of 670 nm. Assuming an absorption Ångström exponent of 1.02, a 5% correction factor should be applied to the firmware output of the MAAP to account for this wavelength difference (Müller et al., 2011a). This correction factor was applied in the present study. A mass absorption cross section value of 6.6 m² g⁻¹ was used to convert the equivalent BC mass concentrations reported in the firmware output of the MAAP to absorption coefficients (as specified by the manufacturer).

During the RAOS campaign (Petzold et al., 2005), MAAP absorption coefficients were observed to have no relationship with aerosol SSA. However, at extremely high SSA values the absorption coefficient measurements from the MAAP can be biased high. It is also important to consider that – to the best of our knowledge – no dedicated study has yet been performed to assess the precision and accuracy of MAAP measurements of samples containing a large fraction of super-micrometer particles. To quantitatively compare the MAAP and CAPS PM_{ss} absorption coefficients during the Cabauw field campaign, both coefficients were adjusted to standard temperature (273.15 K) and pressure (1 atm). It should be stressed that in this comparison we do not consider the MAAP to be a true reference standard for measuring aerosol absorption coefficients. Rather, the value of the instrument for the present study lies in the fact that it displays very low instrument unit-to-unit variability, which means it can provide a common and stable reference point against which CAPS PM_{ss} absorption measurements can be compared.

505 A single particle soot photometer (SP2; Droplet Measurement Technologies, Longmont, CO, USA) was used to measure black carbon mass concentrations at high time resolution from a mobile laboratory deployed during the Bologna field campaign (Sect. 3.4.2). The SP2 measures the mass of individual black carbon particles on a single-particle basis using the principle of laser-induced incandescence. The instrument has been described in detail previously (Schwarz et al., 2006; Stephens et al., 2003). Due to its very high sensitivity and responsiveness, its specific purpose in the present study was to provide a high time resolution reference time series of relative absorbing aerosol concentration. Its configuration during the present study is 510 described by Pileci et al. (2020).

3.1.2 Particle size classifiers applied for cross calibration and truncation measurements

Two different types of aerosol size classifiers were used to generate monodisperse test aerosols for the purposes of measuring cross calibration constants and scattered light truncation: an aerodynamic aerosol classifier (AAC; Cambustion Ltd, Cambridge, UK) and a differential mobility analyzer (DMA; custom built version of same design as the TSI Model 3081 long-column DMA, TSI Inc. Shoreview, MN, USA). The correct operation and sizing of both types of classifiers was confirmed 515 throughout all the experiments by measuring nebulized PSL particles of different diameters (i.e., by operating the classifiers in scanning mode with downstream concentration measurements performed by a condensation particle counter).

The AAC classifies particles based on their relaxation time under the action of a centrifugal force generated in the annular gap 520 between two rotating coaxial cylinders (Johnson et al., 2018; Tavakoli and Olfert, 2013). The particle relaxation time is related to aerodynamic equivalent diameter in a straight-forward manner. In the context of highly size-dependent optical measurements, the major advantage of such a classification method is that it does not depend on particle electrical charge (unlike the DMA), which means the AAC can produce truly monodisperse distributions of particles (i.e., of finite width but without the presence of additional size distribution modes due to multiply-charged particles). This charge-independent 525 classification approach also enables higher aerosol transmission efficiencies than is possible with the DMA, which improves the signal-to-noise-ratio of any downstream optical measurements. An additional advantage of the AAC relative to the DMA is that it can classify particles over a wider diameter range, including particles with diameters of up to ~5 micrometers. The AAC was operated in the present study with the sheath-to-aerosol flow ratio of around 10:1, which results in geometric standard deviations for the classified aerosols of around 1.14. The set point aerodynamic diameters were converted to volume equivalent 530 diameters using literature values of particle density and assuming the classified particles were spherical.

3.1.3 Particle size distributions of ambient aerosol

Measurements of ambient particle size distributions were required during the Cabauw field campaign (Sect. 3.4.1) as inputs for the truncation correction calculations. These measurements were obtained by a scanning mobility particle sizer (modified version of the TSI SMPS 3034; TSI Inc. Shoreview, MN, USA) covering the mobility diameter range from 10 to 470 nm and

535 an aerodynamic particle sizer (TSI APS 3321; TSI Inc.) nominally covering the aerodynamic diameter range from 0.54 to
20 μm .

The hourly-averaged SMPS and APS size distributions were merged to create total aerosol size distributions covering the
diameter range from 0.0104 to 10 μm for use in the truncation calculations. This was achieved by first converting the measured
540 diameters of the respective instruments to volume equivalent diameters. The SMPS electrical mobility diameters were simply
taken to represent volume equivalent diameter (i.e. shape effects were neglected). The APS aerodynamic diameters were
divided by the square root of particle effective density to translate them into volume equivalent diameters. A constant effective
density of 2 g cm^{-3} was assumed. The joined size distributions were then created by linearly interpolating the SMPS and shifted
APS measurements onto a common diameter scale between 0.0104 to 10 μm . The APS measurements of particles with physical
545 diameters less than 0.6 μm were not used in this joining calculation since they are known to display counting efficiency
problems (Pfeifer et al., 2016).

3.2 Measurements of scattering cross calibration constants

Scattering cross calibration constants (Sect. 2.2.2) were measured with the experimental arrangement shown in Fig. S2.
Ammonium sulphate particles or PSL spheres were generated in a Collison type nebulizer and passed through a diffusion drier
550 filled with silica gel for drying. A filtered bypass line was used after the nebulizer, and the ratio of the flows in this bypass line
and the normal sampling line were adjusted to provide control on the concentration of the nebulized aerosol.

In the default laboratory setup, after drying the particles were passed through a size classifier to produce monodisperse
distributions of particles with modal diameters less than 200 nm (i.e., to produce particles with size parameters less than
555 approximately 1 that fall within or at least near the Rayleigh light scattering regime, see Sect. 2.2.2). Additionally, to investigate
a simplified procedure for potential application in field campaigns, selected calibrations were also performed by bypassing the
size classifier. In this case only PSL particles with diameters less than 200 nm were produced with the nebulizer to keep the
generated aerosol within or near the Rayleigh light scattering regime. Nevertheless, it is possible that larger PSL aggregates
(doublets or triplets) were also generated by the nebulizer. Such aggregates would be large enough to cause non-Rayleigh light
560 scattering. In addition, large numbers of non-PSL, smaller particles (most with diameters $< \sim 30$ nm with tail extending to 100
nm or larger) are also produced when nebulizing PSL due to the presence of surfactants and other impurities in the PSL and
Milli-Q water solutions. The composition of these particles is generally unknown. The possibility that they contained
substantial absorbing components is unlikely but cannot be ruled out, which would violate the required cross calibration
condition that the calibration aerosol has $\text{SSA} = 1$.

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In some of the calibrations a storage volume was placed upstream of the CAPS PM_{ss}a unit being calibrated. In these
experiments the volume was first filled with calibration aerosol and the CAPS PM_{ss}a was then used to draw the concentration

in the volume down to near zero. This enabled measurement of $\beta_{Rayleigh}$ over a broad range of aerosol loads. In other cases the calibration aerosol was simply fed directly to the CAPS PMssa unit being calibrated. In all cases we limited the calibration measurements either during the experiment or later during data processing to b_{ext} values less than 1000 Mm^{-1} to avoid non-linearity issues between the scattering and extinction measurements (Sect. 2.2.2).

Two examples of scattering cross calibration measurements are shown in Figs. S3 and S4. Fig. S3 is an example of a calibration performed with 240 nm PSL particles with the storage volume present to enable measurement across a broad range of aerosol loadings, while Fig. S4 shows an example where the storage volume was not used such that the measurements only cover a narrow range of aerosol loading. The 240 nm PSL particles are slightly larger than the particles we typically use for cross calibration but these two examples are shown here to demonstrate the effect of the storage volume. The top panel of these figures show time series of the b_{ext} and $b_{sca,uncalib.}$ and the bottom left panel displays these variables in a scatter plot on a log-log axis. Onasch et al. (2015) determined $\beta_{Rayleigh}$ as the gradient of a line fit to the scatterplot data. To avoid any potential linear fitting artefacts caused by outlying measurements, we elected to determine $\beta_{Rayleigh}$ as the mean value of the ratio of $b_{sca,uncalib.}/b_{ext}$ for b_{ext} values greater than 50 Mm^{-1} . This lower limit was chosen to avoid low signal-to-noise ratio measurements affecting the determined $\beta_{Rayleigh}$. The bottom right panel of Figs. S3 and S4 displays histograms of the $b_{sca,uncalib.}/b_{ext}$ ratio (with the condition $b_{ext} > 50 \text{ Mm}^{-1}$). It is seen that the values of the ratio are typically normally distributed, regardless of whether the measurements covered a broad range of extinction values or not (Fig. S3 vs S4). We take the standard deviation of the measured ratios to represent the precision with which $\beta_{Rayleigh}$ can be determined.

3.3 Measurements of scattered light truncation as a function of particle diameter

The general experimental setup that is shown in Fig. S2 was also used to measure scattered light truncation as a function of particle diameter, in order to validate our new truncation calculations (Sect. 2.2.3). Size-resolved truncation measurements can be performed directly with the CAPS PMssa using size-classified, non-absorbing test aerosols and taking b_{ext} as $b_{sca,true}$, and $b_{sca,Rayleigh}$ as $b_{sca,meas}$ in Eq. (4). Similarly as for the cross calibration constant $\beta_{Rayleigh}$, we applied a threshold condition of $b_{ext} > 50 \text{ Mm}^{-1}$ when calculating mean ratios of b_{ext} to $b_{sca,Rayleigh}$. For this application, the AAC was always used as the size classifier, since the AAC is able to generate truly monodisperse distributions of particles (i.e., finite width but without additional size modes due to multiply charged particles) and provides a larger upper size limit.

We measured truncation values for three different types of non-absorbing aerosols: PSL, DEHS (Di-Ethyl-Hexyl-Sebacat) and ammonium sulphate. The relevant properties of these aerosols are listed in Table 3. Three aerosol types were used in order to check consistency across different aerosols to provide more robust results. Nebulized and dried PSL and DEHS particles are spherical, while dried ammonium sulphate particles are at least near spherical (e.g. Biskos et al., 2006). Spherical or near spherical particles were used so that the aerosol phase functions could be calculated precisely with Mie theory. The geometric standard deviations of the monodisperse DEHS and ammonium sulphate aerosols were nominally around 1.14 as determined

by the operating conditions of the AAC (Sect. 3.1.2). We used geometric standard deviations of 1.1 in our model calculations for these two aerosol types. The width of PSL size distributions are size-dependent and generally narrower than the transfer function of the AAC as used in these experiments. Therefore, we considered two geometric standard deviations of 1.05 and 1.1 in our model calculations for PSL. Rayleigh normalization factors (i.e., $b_{sca,true}^{Rayleigh} / b_{sca,meas}^{Rayleigh}$, see Eq. 4) were measured at the beginning of each experimental run using particles of the given aerosol type with size parameters less than or close to 1. A number of repeat experiments were performed for some of the aerosol types, as indicated in Table 3.

3.4 Field measurements

3.4.1 Cabauw campaign

The Cabauw field campaign was conducted from 11 September to 20 October 2016 at the KNMI (Koninklijk Nederlands Meteorologisch Instituut) Cabauw Experimental Site for Atmospheric Research (the Netherlands; 51° 58' N, 4° 55' E, -0.7 m a.s.l.). The campaign was conducted in the framework of the ACTRIS project (WP11) and occurred simultaneously with the CINDI-2 MAX-DOAS intercomparison campaign (Kreher et al., 2020). The CAPS630b unit was the CAPS PM_{ss} instrument deployed during this campaign to measure absorption coefficients at 630 nm. Absorption coefficients were also measured at 637 nm with a MAAP (Sect. 3.1.1). Particle size distributions were measured with an SMPS and APS (Sect. 3.1.3). All instruments were housed in a laboratory at the base of the KNMI-mast Cabauw behind identical inlets consisting of PM₁₀ sampling hats protruding 4.5 m from the laboratory roof. The inlets contained large diameter nafion driers, which kept RH in the sampling lines below 50%. All the data used in the present study were averaged over one hour periods. This includes the joined SMPS and APS size distributions (Sect. 3.1.3), which used to calculate hourly-resolved truncation correction factors using the model presented in Appendix A1.

3.4.2 Bologna campaign

The Bologna field campaign was conducted from July 5 to 31, 2017. This campaign was also conducted within the framework of the ACTRIS project. The full campaign consisted of multiple stationary measurement sites that were centered around the city of Bologna in Italy's Po Valley. Additionally, a heavily instrumented mobile measurement van (the MOSQUITA; Bukowiecki et al., 2002; Weimer et al., 2009) travelled between the stationary sites to perform spatially-resolved measurements of black carbon concentrations and properties. The results of these mobile measurements are presented by Pileci et al. (2020a). In the present study we use only one hour of mobile measurements that were performed from the MOSQUITA while it was travelling on the heavily-trafficked A1 highway between Bologna and Lodi on the morning of July 25, 2017. During this time period absorption coefficients were being measured with the CAPS780, and black carbon concentrations with an SP2 (Sect. 3.1.1).

630 3.4.3 Payerne campaign

The Payerne field campaign was conducted from August 26, 2019 to January 14, 2020 in Payerne, Switzerland and involved the PMssa units CAPS450 and CAPS780. The goal of this campaign was to compare the hygroscopic properties of aerosols measured using remote sensing and in situ techniques. In the present study we only present the results of the CAPS PMssa cross calibrations that were performed for the campaign: no ambient measurements are shown. In addition to the cross calibrations that were performed at the Payerne field site with the CAPS450 and CAPS780 units, we also present the results of calibrations performed immediately before and after the campaign in the Aerosol Physics Laboratory of the Paul Scherrer Institute. In addition to the two other PMssa units, the CAPS630a was also included in these laboratory calibrations.

4. Precision of determination of the cross calibration factor and its stability over time

The cross calibration constants that were measured for the CAPS450, CAPS630a and CAPS780 PMssa units during and around the Payerne field campaign are presented in Fig. 5. These measurements are used to assess the stability of the cross calibration constant (variability over the time series) and the precision with which it can be determined (error bars represent ± 1 standard deviation of the ratios of $b_{sca,uncalib.}$ to b_{ext} for each calibration, as visualized in the lower right panels of Figs. S3 and S4). Some of the measurements were performed on size-classified aerosols (solid plot markers), and some were performed without classification (open plot markers), as discussed in Sect. 3.2

To investigate the effect of size classifying the aerosol, four calibrations were purposely performed back-to-back, with and without an AAC size classifier. The results of these back-to-back calibrations are plotted on their own in Fig. S5. In two cases, the cross-calibration constants determined with and without size classification were similar (CAPS630a with a difference of -1.9% between calibrations, and CAPS450 run2, with a difference of 2.8%). However, in the other two cases the $\beta_{Rayleigh}$ value determined without size classification was substantially less than the value determined with classification: -9.7% difference for CAPS450 run1, and -6.6% difference for CAPS780. This is likely because of the presence of PSL doublets or triplets, or because the non-PSL particles that are unavoidably generated during the PSL nebulization process either contained absorbing components or were big and abundant enough to cause substantial non-Rayleigh light scattering. In any case, we assume that the cross calibrations performed with size classification provides the most trustworthy measurement, since it is more certain that all the required conditions for the cross calibration are met.

Despite the potential differences between size classified and non-size classified measurements, some important results are still clearly seen in Fig. 5. Firstly, it is apparent that the variability in $\beta_{Rayleigh}$ over time is instrument-dependent. The different behaviors observed for these three PMssa units represent the range of performances we have observed for CAPS PMssa monitors in the field. The least stable unit in this context was the CAPS450. In the ten days prior to the beginning of the campaign $\beta_{Rayleigh}$ for CAPS450 was observed to decrease from 0.63 to 0.53. During the campaign itself, $\beta_{Rayleigh}$ ranged from

0.43 to 0.28, showing a general decreasing trend as the campaign progressed. This observed drift corresponds to tens to hundreds of % of uncertainty in b_{abs} (Fig. 4). The CAPS450 instrument diagnostics provided no evidence of instrument malfunction, change, or contamination during this period. Therefore, this example demonstrates that regular cross calibration validation measurements are necessary to exclude significant drifts. Given that the precise reason for instability in $\beta_{Rayleigh}$ is unknown, we refrain from providing a stability-based uncertainty estimate for this unit in Table 1. However, the four individual AAC-based calibrations performed in the laboratory prior to the beginning of the campaign still provided a chance to investigate the precision-based uncertainty in $\beta_{Rayleigh}$ for CAPS450. The average standard deviation of the ratios of $b_{sca,uncalib}$ to b_{ext} measured during these four calibrations was 0.009, which is 1.5% of the average $\beta_{Rayleigh}$ of 0.60. Therefore, we conservatively estimate that the $\beta_{Rayleigh}$ can be determined with a precision of 2% for this unit (Table 1).

The averaged value of $\beta_{Rayleigh}$ for CAPS780 was 0.27 over the 14 measurements taken during the 140 days of the campaign (from day 10 to 151 on the cumulative days x-axis). The minimum and maximum values measured during this period were 0.26 and 0.28, respectively. Thus, we conservatively estimate a stability-derived uncertainty value of 8% ($\delta\beta_{Rayleigh}/\beta_{Rayleigh}$) for this unit in Table 1, while noting that this estimate is derived from calibration measurements without a size classifier, which may have contributed to the observed variability. The 8% uncertainty range is represented by the green-shaded uncertainty band in Fig. 5. The precision-based uncertainty estimate in $\beta_{Rayleigh}$ for this unit is determined from the five AAC-based cross calibrations performed in the laboratory before and after the campaign (i.e., the average size of the green error bars). From these measurements we calculate a precision-derived uncertainty of 6%. The CAPS630a was not operated during the Payerne field campaign period. However, laboratory measurements before and after the campaign indicated that $\beta_{Rayleigh}$ for this unit can be determined with a very high precision of 2% and is stable to within 2% over time. We believe that this unit represents an example of the best case performance for cross calibration precision and stability that is possible with the CAPS PMssa.

In addition to continual drifts in $\beta_{Rayleigh}$ over time, it is also interesting to note how $\beta_{Rayleigh}$ can change following known instrument-malfunction events such as contamination of the PMssa optical cavity. Fig. S6 displays CAPS PMssa measured b_{ext} (left panel) and b_{sca} (right panel) at 450 nm against independent measurements of these quantities (CAPS PMex for b_{ext} , nephelometer for b_{sca}) during a field campaign at the rural background site of Melpitz, Germany. Throughout the duration of these measurements the optical cavity of the 450nm PMssa unit became contaminated, moving the instrument outside of its intended range of operation, with average baseline optical loss varying from 758 to 1248 Mm⁻¹. Such contamination events can occur due to large pressure fluctuations in the aerosol sampling line or failure of the instrument's purge flow system. They do not occur just by measuring high aerosol loads. In this case, the instrument-malfunction contamination event This caused an increase in the bias of the b_{ext} measurement relative to the corresponding PMex measurement from 5 to 17%. However, Over the same period, the bias of the b_{sca} measurement with respect to the corresponding nephelometer measurement was unchanged, which implies that $\beta_{Rayleigh}$ did change. This suggests that contamination of the optical cavity of the PMssa unit

~~created a bias in the instrument's b_{ext} measurement by CAPS. We hypothesized that the contamination caused an increase in the total optical loss of the CAPS, resulting in the CAPS measurement entering the nonlinear regime normally described by $b_{\text{ext}} > 1000 \text{ Mm}^{-1}$. However, inspection of the data did not support this hypothesis. We therefore, as specified by the manufacturer, we~~ recommend that the CAPS PMssa baseline should be monitored continuously throughout measurement campaigns for signs of mirror contamination, and that contaminated mirrors are cleaned promptly.

5. Laboratory truncation measurements and comparison against model calculations

The results of the laboratory truncation measurements as a function of AAC-selected particle diameter are shown in Fig. 6 for both PSL and DEHS test aerosols. We refer to these curves as 'truncation curves'. Truncation curves are a useful way to validate truncation calculations since the particle size is a key determinant of the aerosol scattering phase function and consequently γ for spherical particles of known constituent material. Following earlier studies (Liu et al., 2018; Onasch et al., 2015), we display measured truncation values as the inverse of γ as defined by Eq. (4). Measurements are presented at three different wavelengths (corresponding to the three figure columns) as measured by the CAPS450, CAPS630a and CAPS780 PMssa units. The equivalent measurements for ammonium sulphate are shown in Fig. S7. These results are not included in Fig. 6 since we suspect that the ammonium sulphate particles were slightly non-spherical after nebulization and drying, which makes them less useful for comparison with Mie-theory based model curves, as is done below. Nevertheless, it is seen that the ammonium sulphate measurements are qualitatively consistent with the PSL and DEHS results over many repeated experiments. The PSL measurements at 450 and 630 nm presented by Onasch et al. (2015) are also included in Fig. 6. They indicate less truncation than the corresponding measurements from the present study. The reasons for these discrepancies are not entirely clear but may be related to the fact that the Onasch et al. (2015) measurements were obtained after size classification by DMA, although the authors found no substantial evidence of additional size distributions peaks due to multiply charged particles (and such particles would anyway cause greater truncation, not less).

A variety of modelled truncation curves are also displayed in each panel of Fig. 6 for comparison with the measurements. Broadly, these can be classified into calculations that include the process of scattered light reflection from the inner surface of the glass sampling tube, and those that do not. One uncertain parameter is the extra path length outside the integrating sphere that contributes to scattered light collection (the l parameter), which was set to 1 cm in the original model calculations by Onasch et al. (2015) without considering glass tube reflection (dashed grey lines in Panels a and b). The corresponding truncation curves calculated with the new model presented in Appendix A1 and with the process of glass tube reflection switched off (solid light grey lines) agree well with the original model. Calculations made with the new model with the process of glass tube reflection turned on and l set to 1 cm are shown as the solid colored lines. The shaded envelopes around these curves demonstrate the sensitivity of modelled truncation to variation of l between its lower and upper geometrical boundaries

($0 \text{ cm} \leq l \leq 4.7 \text{ cm}$). Finally, the dashed colored curves in each panels are truncation curves calculated with the RTE-based model presented by Liu et al. (2018). These curves include the process of glass tube reflection and assume $l = 1 \text{ cm}$.

730 Measured and modelled truncation curves all display the same general features. Truncation values are relatively flat up to a
volume equivalent diameter of around 200 nm (or $\sim 150 \text{ nm}$ at the 450 nm wavelength). This corresponds to the approximate
limit of the Rayleigh light scattering regime. At larger diameters, particles begin scattering relatively more light in near-forward
directions, where it escapes from the CAPS PMssa integrating sphere. As a result of this loss of scattered light, the truncation
curve begins decreasing with increasing particle diameters in a complicated but well-known manner due to the variation of the
735 scattering phase function with particle diameter. Specifically, at diameters $> \sim 1 \mu\text{m}$, peaks in the truncation curves occur due
to Mie resonances. The peaks are slightly broadened by the small polydispersity of the AAC-selected size distributions. This
can be seen when comparing the Onasch et al. (2015) modelled curves, which were calculated assuming a perfectly
monodisperse distribution of particles, and the modelled curves from the present study, which consider the finite width of the
experimental size distributions (in the case of PSL two geometric standard deviations of 1.05 and 1.1 are modeled). The
740 remarkable fact that the Mie resonances are discernable in the measured PSL and DEHS truncation curves, even if perfect
quantitative agreement isn't obtained with the models, provides high confidence in the AAC-CAPS PMssa setup for measuring
scattered light truncation. The Mie resonances are not as apparent in the ammonium sulphate measurements, which we suspect
is likely due to particle non-sphericity as mentioned above.

745 In general, good agreement is obtained between the new model calculations including the process of glass tube reflection and
the RTE model calculations. This is encouraging given these models are based on fundamentally different approaches for
calculating truncation. Both of these modelled curves predict generally greater truncation than the calculations that neglect the
process of glass tube reflection, as expected from theoretical considerations (Sect. 2.2.3). The measurements are in better
agreement with the modeled curves that include glass tube reflection, demonstrating that this process must be considered in
750 the calculations. This is a robust result that is consistent across all three aerosol types for particle diameters up to $5 \mu\text{m}$ and all
three optical wavelengths that were investigated.

Considering the calculations made with the new model presented in Appendix 1, the best agreement with the measured data
appears to be obtained for an l value of 1 cm. However, there is enough scatter in the measurements to argue that any l value
755 is plausible within the geometric limits of this parameter (from 0 to 4.7 cm). Although varying l over this range captures the
measurements well, we stress that this does not imply that variable l is the physical reason for the measurement imprecision.
For one, it can be argued that the lower limit l value of 0 cm is unrealistic since it is expected that particles at the boundary of
the sphere will certainly scatter light into the sphere. Similarly, our model uses an idealized geometry of the interior of the
CAPS PMssa cell. Nevertheless, varying l from 0 to 4.7 cm produces differences in calculated truncation that are similar to
760 the differences observed between repeat measurements, as well as to the differences between calculations made with the two

models that include the process of glass tube reflection (i.e., the model presented in Appendix 1 and the RTE model). Therefore, similarly to Onasch et al. (2015), we use l as a convenient tuning parameter to produce an uncertainty envelope that captures the range of measured truncation curves reasonably well.

6. Examples from the field: measurements of atmospheric aerosol absorption coefficients with the CAPS PMssa

765 6.1 Bologna example: instrument sensitivity and rapid response time

Two key features of the CAPS PMssa as a flow-through, continuously-measuring optical instrument are its sensitivity and rapid response time. Onasch et al. (2015) demonstrate the instrument is able to respond to changes in b_{ext} and b_{sca} of less than 1 Mm^{-1} on timescales of only ~ 1 second. These specifications suggest that EMS-derived b_{abs} values measured by CAPS PMssa will display similar responsiveness and sensitivity. If so, these specifications would represent a major improvement over the
770 equivalent specifications for b_{abs} values measured by filter-based absorption photometers, which are based on the slower process of accumulation and detection of aerosol samples on a filter.

To investigate these features under real-world conditions, Fig. 7 presents co-located measurements of aerosol absorption coefficients obtained with the CAPS780 instrument and rBC mass concentration measurements obtained with an SP2. The
775 measurements were obtained while travelling along a busy highway road in a mobile laboratory near Bologna, Italy, where black carbon was shown to be the dominant source of absorbing aerosol. Indeed, the observed correlation between the independent measurements of black carbon and aerosol absorption is remarkably good, especially given the short averaging time of 5 seconds. In particular, the sharp peaks that are observed in both time series are found to align with each other extremely well. These peaks correspond to black carbon emissions from passing vehicles on the highway. Although this is
780 only a one-hour sample of data, this example demonstrates the EMS-derived b_{abs} values can be measured at very high time resolution with the CAPS PMssa, comparable to what can be achieved with the single-particle level measurements of rBC mass from an SP2.

Although Fig. 7 demonstrates the responsiveness of the CAPS PMssa, it must be stressed that the absolute values of the plotted
785 b_{abs} measurements are still uncertain and should not be used quantitatively (the SP2 measurements of rBC mass are also underestimated and should not be used quantitatively, since the instrument was unable to detect an unknown fraction of the total rBC mass due to the small size of the freshly-emitted BC cores; (Pileci et al., 2020b)). Specifically for the CAPS PMssa measurements, the plotted quantity is $b_{abs, Rayleigh}$ to indicate that the underlying b_{sca} measurements have not been corrected for the truncation of non-Rayleigh scattered light. To do so accurately would require co-located, equally high time resolution
790 measurements of either scattering phase functions or information that could be used to calculate phase functions (e.g. size distributions, fractal BC properties) for the freshly emitted BC-containing emissions plumes. Nevertheless, the ability to

measure relative b_{abs} values at high time resolution with the CAPS PM_{ss}a creates possibilities for new types of experiments that were not previously feasible with traditional absorption photometers.

795 6.2 Cabauw example: comparison of CAPS PM_{ss}a absorption measurements against independent measurements with a MAAP

In this section we use an example dataset from the Cabauw campaign to ~~perform a full quantitative assessment of~~ make a direct absorption instrument-to-instrument comparison between the CAPS PM_{ss}a and the MAAP. This comparison is performed in order to gain insight into the ability of the CAPS PM_{ss}a to measure absolute aerosol absorption coefficients, ~~including a detailed characterization of the scattered light truncation effects, as well as the uncertainties in the other underlying measurements.~~ As discussed in Section 3.1.1, the MAAP was used for this comparison because it provides stable and reproducible absorption measurements. However, it should be reiterated that the MAAP is not a true absorption reference standard: this instrument is associated with its own measurement uncertainties, which are not well characterized for aerosols with large contributions of super-micrometer particles. Despite these issues, our focus here is only on the uncertainties related to the CAPS PM_{ss}a absorption measurements, including detailed characterization of the scattered light truncation effect. The CAPS630b was the PM_{ss}a unit operated during ~~this~~ Cabauw campaign. ~~This~~ The unit ran autonomously, continuously, and stably over the one month of operation, which we have found to be typical for CAPS PM_{ss}a units operated at stationary field sites.

6.2.1 CAPS PM_{ss}a baseline characteristics

810 The scattering and extinction baseline measurements over the campaign are displayed in Fig. S8. These measurements were performed for 1 min every 10 mins using the auto-baselining feature of the instrument. The average standard deviations of the extinction and scattering baseline measurements over all 1 min baseline periods were 0.35 and 0.66 Mm⁻¹, respectively. We take these values to represent the precision-based uncertainty estimates in $b_{ext,baseline}$ and $b_{sca,baseline}$ in Table 1. Both baseline time series indicate that the optical cavity was generally clean and suffered no major contamination events during the campaign.

815 Fig. S8 also contains time series of the baseline drift in each channel, which were calculated from the differences between two successive baseline measurements (i.e., over a period of 10 mins, which means the calculated metric does not include possible variations over shorter time scales). On average, the extinction baseline drifted by 0.026 Mm⁻¹ min⁻¹, and the scattering baseline by 0.013 Mm⁻¹ min⁻¹. Individual values of up to 0.3 and 0.09 Mm⁻¹ min⁻¹ were observed in the extinction and scattering channels, respectively. To minimize the impacts of these drifts, both the extinction and scattering measurements were

820 reprocessed using the method of linear interpolation between successive baseline measurements (Pfeifer et al., 2020). Fig. S9 indicates that this reprocessing had a noticeable effect at a 1-second time resolution for b_{ext} values less than ~20 Mm⁻¹ and b_{sca} values less than ~5 Mm⁻¹. However, these effects are averaged out when considering hourly-averaged data.

6.2.2 Uncertainties in the CAPS PM_{ss} b_{sca} correction factors β and γ

825 The cross calibration constant for the CAPS630b could be measured with high precision (~2%) and appeared to be very stable based on measurements at the beginning (0.82; Fig. S10) and at the end of the campaign (0.80; Fig. S11), which differed by only 2.5%. This is comparable with the optimum performance we have observed for CAPS PM_{ss} units with regards to cross calibration (see Sect. 4).

830 In the absence of direct scattering phase function measurements, hourly time resolved truncation correction factors were calculated with the Mie-theory based model presented in Appendix A1. Joined size distributions measured by SMPS and APS were input into the model. To estimate the uncertainty in γ a sensitivity analysis was performed with respect to the model's input parameters. This analysis is presented in the Supplementary Information as Sect. S1 and visualized in Fig. S12. Specifically, we investigated the sensitivity of γ to the ambient aerosol refractive index (real parts between 1.50 to 1.59, and imaginary parts between 0.00 to 0.01, based on the summary of measurements presented by Espinosa et al. (2019), the l parameter (varied from 0 to 4.7 cm based on the results presented in Sect. 5), and the accuracy of the coarse mode size distribution measurements between diameters of 2.5 and 10 μm . The truncation correction was found to be most sensitive to the l parameter (~4% across the range of tested parameters), weakly sensitive to the real part of the refractive index and size distribution information between 2.5 and 10 μm , and barely sensitive at all to complex part of the refractive index (not shown). Overall, we estimate an uncertainty in γ of 6% for the Cabauw campaign based on this analysis (difference between the 840 minimum and maximum average values of all the simulated distributions shown in Fig. S12). However, it must be stressed that this estimate is limited by our Mie-theory based calculations, which do not include potential effects due to morphologically-complex particles such as freshly-emitted fractal BC aggregates. That is, the estimate only covers the parametric uncertainty in our Mie-theory based calculations.

6.2.3 Truncation effects for fine-mode dominated and coarse-mode containing samples

845 The distributions of the time-resolved truncation correction factors calculated over the Cabauw campaign are clearly bi-modal (Fig. S12). This indicates that there were two limiting types of aerosols measured during the campaign: i) fine-mode dominated aerosol that only required a minor truncation correction; and ii) aerosol with substantial coarse mode fraction that required a larger truncation correction. For further investigation, we extracted two subsets of data representing the fine-mode dominated and coarse-mode containing samples. This was done by selecting those aerosols whose coarse-mode number fractions (defined 850 as $N_{D_p > 1\mu\text{m}}/N_{total}$ calculated from size distributions) are in the lower and upper quartiles of all the data, respectively. The median normalized size distributions for these two categories are plotted in Fig. S13. The figure clearly shows a coarse mode of particles with diameters between ~0.6 and 5 μm that was present in the coarse-mode containing samples but not the fine-mode-dominated aerosols. Despite the small number fractions, these coarse mode particles can make substantial contributions

to the scattering coefficients and they produce a greater fraction of forward and backward scattered light, thereby affecting
855 truncation disproportionately.

The distributions of the time-resolved γ values for the fine-mode dominated and coarse-mode containing groups of samples are displayed in Fig. S14 (for the same ranges of model inputs that were examined for the full Cabauw dataset in Fig. S12). As expected, the required truncation correction values are substantially smaller for the fine-mode dominated group than the
860 coarse-mode containing samples. This follows from the normalized ensemble scattering phase functions for each group, which are displayed in the right panel of Fig. S13. These functions were calculated from the median size distributions with Mie theory and an assumed refractive index of $1.59 + 0.01i$ (functions like these are required as inputs for the γ calculation, as shown in Fig. 3). It is seen that the phase function for the fine-mode dominated category is less focused in the near-forward (scattering angles $< 25^\circ$) and backward (scattering angles $> 125^\circ$) scattering directions than that of the coarse-mode containing category,
865 which is why these samples are associated with lower γ values.

Fig. S13 also compares the Mie calculated scattering phase functions with measurements obtained by Espinosa et al. (2018) from a broad range of aircraft flights conducted throughout the USA. Three categories of measurements are shown: two categories of coarse-mode containing aerosols (coarse categories 1 and 2) and one category of ‘fine’ aerosols (which is
870 comprised of measurements for aerosols classified as ‘urban’, ‘biomass burning’ and ‘biogenic’, all of which were observed to have very similar scattering phase functions). The calculated scattering phase functions agree reasonably well with the corresponding measurements at near-forward scattering angles (comparing the fine-mode dominated and ‘fine’ categories; and the coarse-mode containing and the two coarse categories). However, at near-backward scattering angles the Mie calculations predict proportionally more light scattering than is observed in the measurements. If it is assumed that the measurements by
875 Espinosa et al. (2018) are reasonably representative of the average scattering phase functions of these aerosol types also in European continental air masses, this comparison suggests that the Mie calculations would tend to slightly overestimate the truncation correction factors displayed in Figs. S12 and S14, and that the degree of overestimation would be greater for the coarse-mode containing group than the fine-mode dominated samples. Considering only the parametric uncertainty in the calculated γ values (i.e., that uncertainty related to the Mie truncation model inputs), we estimate values of 4% and 9% for the
880 fine-mode dominated and coarse-mode containing groups, respectively, based on the minimum and maximum mean values for each of the groups shown in Fig. S14.

6.2.4 Comparison of CAPS PM_{ss} and MAAP b_{abs} measurements

Considering all of the underlying CAPS PM_{ss} uncertainties for the Cabauw dataset that are summarized in Table 1, it is clear that the largest individual source of uncertainty is related to the truncation correction that must be applied to b_{sca} . To investigate
885 the effect of this uncertainty on the ultimate derived b_{abs} , Fig. 8 compares these measurements against independent b_{abs} measurements obtained with a MAAP under three different truncation correction scenarios (corresponding to the three rows

of the figure). It is worth recalling that the coefficients from both instruments were adjusted to standard temperature (273.15 K) and pressure (1 atm) for this quantitative comparison. The plot is further split into three columns, following the data grouping done in the previous subsection, so that the results for the fine-mode dominated samples, the coarse-mode containing samples, and the full dataset taken as a whole can be inspected separately. Each subplot contains an uncertainty envelope that represents the 95th percentile of hourly-resolved theoretical uncertainty values calculated with the error model and inputs presented in Sect. 2.3 and Table 1. Uncertainties in γ of 4, 9, and 6% were used for the fine-mode dominated samples, coarse-mode containing sample and full dataset, respectively. The uncertainty envelopes are plotted around straight lines (shown as solid orange lines) representing the mean ratios of the CAPS to MAAP b_{abs} values displayed in each subplot. Mean ratios were chosen rather than standard linear fits to avoid biasing the plotted lines towards the highest measured values, and because averaging to 1 hour effectively removed the random noise at the lowest measured values (e.g. as can be seen in Fig. S9).

The first row of this figure (Figs. 8a, b, and c) displays the CAPS PM_{ss} b_{abs} measurements that were processed with time-resolved truncation correction factors calculated with the Mie-theory based model (Appendix A1) with m set to $1.59 + 0.01i$ and l to 4.7 cm. This processing results in generally good agreement between the CAPS PM_{ss} and MAAP b_{abs} measurements. Strong correlation is seen between the two independent measurements for all three subsets of the data (Figs. 8a, b, and c). However, on average, the absolute values of the two measurements are systematically offset by about 20%. In particular, the mean ratios of CAPS b_{abs} to MAAP b_{abs} (i.e., as indicated by the solid orange line in each sub-figure) varies from 0.78~~81~~ to 0.814. The precise reasons for these systematic offsets are not clear. One possibility is the geometry correction factor applied to the CAPS630b PM_{ss} data (0.73) was inaccurate. However, we consider it unlikely that this factor alone could fully explain the observed discrepancy (it would need to be lower by ~20% – since b_{abs} is directly proportional to $1/\alpha$, Eq. (7) – and this is beyond the range of α values that have so far been measured for PM_{ss} units, see Sect. 2.2.1). Another possibility is that the MAAP absorption coefficients were systematically overestimated (uncertainties of up to 7% can be expected based on previous studies, see Section 3.1.1). Regardless of the precise reasons for the systematic offset between the measurements, importantly it is important to note that the bias between the CAPS PM_{ss} and MAAP measurements displays no, or only minor, dependence on SSA (which would be detectable as colour trends across the scatter plots). This indicates that it is likely that the CAPS PM_{ss} b_{sca} measurements have not been over-corrected for truncation, since such over-correction would affect the high SSA samples more than the low SSA samples. For these reasons, we believe that Figs. 8a, b, and c represent an example of reasonably well estimated truncation correction. We note that this result was achieved with an l value of 4.7 cm, which is within the plausible range for this model input parameter based on the results presented in Sect. 5.

The second row of the figure (Figs. 8d, e, and f) displays measurements corrected with time-resolved γ values calculated with the Mie model with $l = 1$ cm, which represents the best guess for this parameter based on the laboratory truncation curve measurements presented in Sect. 5. Under this truncation correction scenario, a sizeable number (17.3%) of the hourly-averaged coarse-mode containing CAPS b_{abs} measurements shown in Fig. 8e are negative because $b_{sca} > b_{ext}$, which is

physically not possible. In addition, the bias between the CAPS PM_{ssa} and MAAP measurements displays an SSA dependence, most clearly seen in Figs. 8e and f. Together, these pieces of evidence indicate that the CAPS b_{sca} measurements have been slightly overestimated, leading to absolute b_{abs} values that are biased low. Despite the poor agreement between the absolute CAPS PM_{ssa} and MAAP values for this truncation scenario, it is noteworthy that the two measurements still correlate well with each other.

The third and final row of this figure (Figs. 8g, h, and i) displays measurements that have been corrected for Rayleigh light scattering only. This means a constant γ value of 1 was applied to the b_{sca} measurements. As shown in Fig. 2, this is the same as simply taking the b_{sca} values output by the instrument firmware (i.e., $b_{sca, Rayleigh}$), assuming that the firmware contained the correct α and β values. Here it is clear that the truncation correction has now been underestimated, leading to b_{abs} values that are greater than the corresponding y-axis values in the first row of the figure. Again, this is most clearly seen for the coarse-mode containing group of samples (Fig. 8h). On average, the b_{abs} values for these samples are 1.8694 times higher than the corresponding MAAP measurements.

To further examine the sensitivity of CAPS PM_{ssa} b_{abs} measurements to the scattering truncation effect, five additional truncation correction scenarios are displayed in the five rows of Fig. S15. These results generally support the results displayed in Fig. 8. Additionally, the following conclusions can be drawn: (i) for a given l value (4.7 cm in this case), variation of the ambient aerosol refractive index between $1.5 + 0.01i$ and $1.59 + 0.01i$ has only a minor effect on CAPS PM_{ssa} measured b_{abs} , which can be seen by comparing Figs. S15a-f and Figs. 8a-c, (ii) setting l to 0 cm in the model (the lower limit for l that we derived from Fig. 6) results in substantially overestimated truncation and b_{sca} , which in turn leads to a substantial fraction (35.3%) of negative b_{abs} values for the coarse-mode containing group (Fig. S15h), and (iii) using constant γ values calculated from campaign-averaged size distributions (the averaged joined SMPS and APS particle size distribution, Figs. S15j-l; and the averaged SMPS distribution only Figs. S15m-o) generally leads to poorer results for the coarse-mode containing samples compared to the corresponding results obtained with time-resolved truncation correction, but the results for the fine-mode dominated samples are similar. This latter result is pertinent to field studies where time-resolved scattering phase function information is not readily available (either directly or indirectly, e.g. in the form of measured size distributions), such that a user might be forced to use a constant truncation correction factor.

6.2.5 Summary of the Cabauw results

The Cabauw example demonstrates that the biggest hurdle that must be overcome when measuring atmospheric aerosol absorption coefficients with the CAPS PM_{ssa} is accurate accounting of the scattered light truncation effect. Unfortunately, there are many potential sources of errors in γ , as discussed in a general sense in Sect. 2.3. Even if the scattering phase functions for the atmospheric aerosols being measured were known with high accuracy, γ would still carry substantial uncertainty related to the instrument geometry that must be considered in the truncation calculation, which we choose to represent through the

glass sample tube extension length l . This uncertainty can lead to a broad range of different final b_{abs} outcomes, as we have
955 shown by examining the sensitivity of b_{abs} to variation in the l parameter.

Although the uncertainty in γ cannot be totally avoided, its effect on b_{abs} can be substantially mitigated by restricting datasets to only those aerosol samples that do not display strongly forward-focused light scattering. For the Cabauw example, we successfully achieved this by separately analysing the fine-mode dominated samples. The results for this group of samples
960 were generally very consistent over the range of truncation correction scenarios we investigated. Considering the cases shown in Figs. 8 and S15, the correlation coefficients between CAPS PM_{ss} and MAAP b_{abs} varied between 0.91 and 0.96, while the average ratios of the two measurements varied from 0.614 to 0.959 (for a sample size of 150). Although the precise reasons for the systematic offset between the CAPS PM_{ss} and MAAP measurements are still unclear, the consistency of the results against the changes in γ is highly encouraging. This suggests that despite the remaining truncation uncertainties, the CAPS
965 PM_{ss} can still provide a reliable b_{abs} measurement for fine-mode dominated atmospheric aerosols.

In contrast, the equivalent results for the coarse-mode containing samples were more problematic. For this group and for the truncation correction scenarios shown in Figs. 8 and S15, the correlation coefficients between CAPS PM_{ss} and MAAP b_{abs} varied between 0.88 and 0.95, while the average ratios of the two measurements varied widely from 0.03 and 1.8694 (again
970 for a sample size of 150). The truncation problems for coarse-mode containing samples are two-fold. Firstly, the scattering phase functions for such samples are highly asymmetric, with enhanced forward- and backward-scattering, which means the corresponding γ values are large and more sensitive to small changes in particle size, shape, and/or composition (as demonstrated with respect to particle size, for example, by the steepness of the truncation curves displayed in Fig. 6 at particle diameters greater than $\sim 1 \mu\text{m}$). This sensitivity is likely responsible for the large variability in the mean ratios of CAPS PM_{ss} to MAAP b_{abs} that were obtained for the coarse-mode containing samples across the tested truncation scenarios.
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Adding to this is a second problem, which is that the Mie-theory-predicted phase functions are likely to be inaccurate for supermicrometer particles of complex morphology (e.g. mineral dust aerosols; Curtis et al., 2008). This problem could potentially be overcome by performing truncation calculations with scattering phase functions that have been measured directly, or
980 calculated with consideration of complex particle morphologies (e.g. using more sophisticated optical models, or potentially with scattering phase functions parameterized according to the asymmetry parameter, as inspired by the truncation relationships presented by Liu et al., 2018). However, further work is required to determine how much such truncation correction methods could improve the reliability of CAPS PM_{ss} measurements of aerosols with substantial coarse-mode number fractions.

985 7. Conclusions and recommendations for future studies that use the CAPS PMssa to measure absorption coefficients

We have developed a detailed error model for the CAPS PMssa (Sect. 2.3) and used this as a framework for assessing the ability of the instrument to measure aerosol absorption coefficients via the EMS method. In combination with empirical data, this error analysis underlines the importance of minimizing errors in b_{ext} and b_{sca} . Two key sources of error were identified as requiring further investigation: uncertainties in the instrument cross calibration constant and those related to the truncation
990 correction. Properly accounting for scattered light truncation is the more difficult problem. Our laboratory measurements demonstrate that the process of glass tube reflection (Liu et al., 2018) must be considered in the truncation calculation (Sect. 5). This process was neglected in earlier truncation models (Onasch et al., 2015). However, uncertainty still remains regarding the length of the optical cavity that should be considered in the calculation. Furthermore, if co-located scattering phase functions cannot be measured directly for input to the calculation, one must carefully consider the large range of potential
995 errors that can arise in calculated scattering phase functions. The uncertainties in the cross calibration constant are less problematic. The cross calibration constants can be measured with high precision but regular measurements are required to identify potential drifts. The required frequency of regular cross calibrations varies between instruments (Sect. 4).

We presented two example field datasets to illustrate the potential and limitations of using the CAPS PMssa to measure
1000 atmospheric aerosol absorption. The first example from Bologna demonstrates that the CAPS PMssa can be used to provide much higher time resolution measurements of relative absorption coefficients than is possible with filter-based absorption photometers. The second example from Cabauw confirms that a proper truncation correction is the biggest hurdle to overcome to accurately measure b_{abs} for atmospheric aerosols with the CAPS PMssa. Nevertheless, we demonstrated that under certain conditions – in this case when fine, submicrometer aerosols dominated the particle size distributions – the CAPS PMssa
1005 provides consistent EMS measurements over a range of different truncation scenarios, even for SSA values greater than 0.95.

Based on the lessons learned in the present study, we recommend that the following steps be taken in future studies that use the CAPS PMssa to measure aerosol absorption with the EMS method. Although our focus has been on atmospheric measurements, these recommendations are also applicable to other types of experiments, such as emissions testing and other
1010 laboratory experiments. Furthermore, although our focus has been heavily focused on b_{abs} where the consequences of errors are the most severe, these recommendations also apply to CAPS PMssa measurements of b_{ext} , b_{sca} , and SSA.

- Accurate knowledge of the geometry correction factor, α for a given unit is essential for performing accurate absolute measurements. Due to the manner in which the CAPS PMssa is cross calibrated, both b_{sca} and b_{abs} are directly
1015 proportional to α , and relative errors in α propagate linearly to errors in b_{sca} and b_{abs} . Although it seems that the α for a given unit is relatively stable over time, an obvious future improvement to the instrument would be to include a diagnostic for monitoring α during instrument operation (e.g. by measuring and recording the instrument purge flows).

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- The periodic scattering and extinction baseline measurements that are performed by the CAPS PM_{ssa} over the course of a measurement campaign should always be inspected (e.g. Fig. S8), to ensure reliable instrument operation and to check for the occurrence of potential contamination events. Contamination can drastically reduce the instrument sensitivity and also alter a unit's cross calibration constant (Fig. S6).
 - Following Pfeifer et al. (2020), we recommend that both the scattering and extinction coefficients should be reprocessed with linear (or cubic spline) interpolation between successive baseline periods, rather than relying on the default firmware method of step-function interpolation. The Cabauw results at an optical wavelength of 630 nm indicated that such reprocessing was not important at the hourly-time resolution level, since the impact of baseline drifts on shorter time scales cancelled each other out. Nevertheless, the reprocessing should always be done as a precaution against rapidly changing carrier gas compositions, particularly for lower wavelength units (e.g. 450 and 530 nm) operating in urban settings or other situations where high NO_x concentrations might be encountered (Pfeifer et al., 2020).
 - The scattering cross calibration constants for some PM_{ssa} units can be measured with sufficiently high precision (~2%) and are stable over time (~2%) for accurate EMS measurements. However, for other units the performance can be poorer, especially with respect to stability. Regular cross calibrations should be performed for each individual unit to determine its behavior in this regard. This information should be used to inform experimental designs (e.g. required frequency of cross calibrations for individual PM_{ssa} units).
 - When calculating scattered light truncation, a model should be used that includes the process of reflection from the inner surface of the glass sampling tube. Uncertainty still remains regarding the choice of the l parameter – the extra path length outside the integrating sphere to be considered in the truncation calculation. In the absence of a suitable independent reference, we recommend setting l to 1 cm, which was the value that resulted in the best agreement between the measured and modeled truncation curves displayed in Fig. 6. However, an uncertainty band formed by varying l between of 0 and 4.7 cm should be considered. If an independent reference point is available for a particular experiment, these measurements can be used to assess the most appropriate l value. The Cabauw dataset provided an example of how this can be done using co-located MAAP absorption measurements (Sect. 6.2.4).
 - Uncertainty in the truncation correction also results from the treatment of the scattering phase function.
 - o In the ideal case, the ensemble scattering phase functions should be obtained directly with co-located polar nephelometer measurements. Future studies should be performed to determine if such co-located measurements will enable the CAPS PM_{ssa} to reliably measure absorption coefficients even for aerosols containing high fractions of particles with highly asymmetric scattering phase functions (e.g. super-micrometer particles generally, fractal BC, dust).
 - o If phase functions have to be calculated or assumed, then the aerosol sample to be measured should be conditioned to ensure that its scattering phase function is not too highly forward focused. For the Cabauw example, this was achieved in the post-processing stage by separately analyzing the fine-mode dominated
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1055 samples. It could also be achieved at the measurement stage. For example, if the light-absorbing particles of interest reside primarily in the fine mode, a PM1 selective inlet could be placed upstream of a CAPS PMssa unit to ensure that it only measures sub-micrometer particles. These approaches will not eliminate the uncertainties in the truncation calculation, but they can mitigate the influence of those uncertainties on the precision of the derived b_{abs} values.

1060 Regarding the final point, the influence of fractal BC aggregate particles deserves special mention. These particles constitute one of the key types of absorbing aerosols, especially in field or test-bench measurements of fresh emissions from combustion sources. Fractal aggregates scatter more light into near-forward directions relative to equivalently sized spherical particles, in a manner that cannot be predicted by Mie theory (Liu and Mishchenko, 2007). Even when more advanced scattering calculations are performed, the morphology (e.g. primary particle sphere size and fractal dimension) of fractal aggregates is often difficult to constrain. These effects create the potential for large and systematic errors in calculated γ values, which sets up a trade-off when measuring aerosols with high proportions of fractal BC aggregates with the CAPS PMssa. As the fraction of absorbing fractal BC increases, SSA decreases, which decreases subtractive error amplification (Fig. 4). However, at the same time, errors in γ likely increase due to the shift in the scattering phase function towards forward directions, which would at least partially offset the reduction in error due to the lower SSA. Therefore, it should not be assumed that errors in EMS-derived b_{abs} will always be necessarily lower for aerosols of low SSA. Knowledge of the magnitude and even signs of the potential errors in γ due to the presence of absorbing fractal aggregates is currently very limited and further studies are required to investigate this issue in more detail.

8. Appendix A1: New theoretical model for calculating scattered light truncation in the CAPS PMssa monitor including the process of glass tube reflection

8.1 General formulation for describing aerosol scattering coefficients measured by integrating nephelometers

1075 An integrating nephelometer measures the integrated particulate scattering coefficient of an aerosol sample b_{sca} (also typically denoted as σ_{sp}) at a given wavelength λ . An ideal integrating nephelometer would be sensitive to light scattered in all possible directions. Formally, if $S_p(\lambda, \Omega)$ ($\text{m}^{-1} \text{sr}^{-1}$) was a function describing the distribution of scattered light of wavelength λ as a function of solid angle Ω for some aerosol, an ideal integrating nephelometer would collect light scattered over all 4π steradians with equal sensitivity,

$$b_{sca}(\lambda) = \int_0^{4\pi} S_p(\lambda, \Omega) d\Omega = \int_0^{2\pi} \int_0^{\pi} S_p(\lambda, \theta, \phi) \sin(\theta) d\theta d\phi \quad , \quad (\text{A1})$$

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where θ and φ represent the polar and azimuthal scattering angles, respectively, in a polar coordinate system. If the scattering process is rotationally symmetric with respect to the azimuthal coordinate – which is true for the case considered here of spherically homogeneous particles illuminated by unpolarized light (Mishchenko et al., 2002) – the integral over φ equals 2π and

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$$b_{sca}(\lambda) = 2\pi \int_0^\pi S_p(\lambda, \theta) \sin(\theta) d\theta . \quad (A2)$$

In practice, real integrating nephelometers are unable to collect some fraction of near-forward and near-backward scattered light due to physical design limitations. This issue is known as scattered light truncation. If truncation is not accounted for then particulate scattering coefficients measured with an integrating nephelometer will be systematically underestimated. In the Rayleigh regime (particle diameter $D_p \ll$ the wavelength of light λ), truncation is independent of D_p and particle shape. In the Mie regime ($D_p \sim \lambda$), truncation is a complicated function of D_p and particle shape, with larger particles tending to produce larger truncation as the fraction of light scattered in forward directions with small θ increases.

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8.2 Calculating scattered light truncation in the CAPS PMssa

The CAPS PMssa measures particulate scattering coefficients at a single wavelength λ with an integrating nephelometer of the reciprocal, integrating sphere design (Heintzenberg and Charlson, 1996). The integrating sphere has a nominal diameter $L = 10$ cm. Aerosol particles with number size distribution $dN/d\log D_p$ travel through the sphere along a central axis in a horizontal, cylindrical glass tube of nominal diameter $d = 1$ cm. Light scattered from the aerosol ensemble is detected with a photomultiplier tube (PMT) placed at one point on the integrating sphere.

To correct for the scattered light truncation effect in the CAPS PMssa we apply a truncation correction factor γ to the measured scattering coefficients ($b_{sca, Rayleigh}$), as discussed in Sect. 2.2.3 of the main text. We define γ as the normalized ratio of the true integrated scattering coefficient, $b_{sca, true}$ (i.e., what would be measured with an ideal integrating nephelometer) to the truncation-affected scattering coefficient that is actually accessible to measurement by the instrument, $b_{sca, meas}$. Repeating Eq. (4) from the main text for convenience,

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$$\gamma = \frac{b_{sca, true}}{b_{sca, meas}} \cdot k_{Rayleigh} = \frac{b_{sca, true}}{b_{sca, meas}} \cdot \left(\frac{b_{sca, meas}^{Rayleigh}}{b_{sca, true}^{Rayleigh}} \right) . \quad (A3)$$

The normalization factor $k_{Rayleigh}$ is required to represent the fact that some scattered light truncation is already implicitly accounted for in the CAPS PMSSA cross calibration constant. For the recommended case of cross calibration with Rayleigh scatterers (i.e., Eq. 3), $k_{Rayleigh}$ represents the truncation of the Rayleigh scattered light from the calibration aerosol.

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The coefficients $b_{sca,true}$ and $b_{sca,meas}$ in Eq. (A3) can be calculated using specific versions of the general Eq. (A1) for calculating aerosol scattering coefficients $b_{sca}(\lambda)$. Following on from earlier integrating nephelometry studies (Anderson et al., 1996; Heintzenberg and Charlson, 1996; Moosmüller and Arnott, 2003; Müller et al., 2011b; Peñaloza M, 1999), we express this equation as a function of the scattering function of the particle population $S_p(\theta, \lambda)$, the light collection efficiency of the integrating sphere $\eta(\theta, \lambda)$, and the angular sensitivity function $Z(\theta)$ of the combined optical system:

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$$b_{sca}(\lambda) = 2\pi \int_0^\pi S_p(\theta, \lambda) \eta(\theta, \lambda) Z(\theta) d\theta \quad . \quad (A4)$$

In this equation aerosol properties are represented by $S_p(\theta, \lambda)$ and instrument properties by $\eta(\theta, \lambda)$ and $Z(\theta)$ (which are assumed to be independent of the azimuthal scattering angle ϕ). Examples of each of these three functions are shown in Fig. 3 of the main text for the specific cases of $b_{sca,true}$ and $b_{sca,meas}$.

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To calculate $S_p(\theta, \lambda)$, we assume that we are measuring an ensemble of spherical, homogeneous particles. In this case, $S_p(\theta, \lambda)$ is given by

$$S_p(\theta, \lambda) = \int_{-\infty}^{\infty} \frac{S_{11}(\theta, X, m, \lambda)}{X(D_p, \lambda)^2} \frac{\pi D_p^2}{4} \frac{dN}{d \log D_p} d \log D_p \quad , \quad (A5)$$

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where X is the particle size parameter ($= \pi D_p / \lambda$) and $S_{11}(\theta, X, m, \lambda)$ is the intensity-related scattering matrix element of a single particle with size parameter X and complex refractive index $m(\lambda)$ (Eq. 3.16 in Bohren and Huffman, 1998). For unpolarized incident light, like that used in the CAPS PMSSA, S_{11} describes the angular distribution of scattered light intensity. In this work we calculated S_{11} with Mie theory (i.e., assuming homogenous spherical particles). Specifically, we calculated S_{11} with modified version of Bohren and Huffman's Fortran routine (bhmic.f; Bohren and Huffman, 1998) by B.T. Draine (available at <https://www.astro.princeton.edu/~draine/scattering.html>, last access 14.05.2020). We further modified this routine to accept programmatic inputs and outputs. Alternative forms of $S_p(\theta, \lambda)$ could be calculated with optical models that are not restricted to the assumption of spherical, homogenous particles, or $S_p(\theta, \lambda)$ could be measured directly with a polar nephelometer.

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1135 For an ideal integrating nephelometer, $Z(\theta) = \sin \theta$ (i.e., Eq. A2). Within the CAPS PM_{ssa} integrating sphere, no baffle is
employed in front of the PMT to prevent the detection of directly scattered light (i.e. light that has not undergone any reflections
from the interior surface of the sphere). This could potentially lead to deviations from the ideal angular sensitivity condition.
However, the fraction of the interior surface area of the sphere taken up by the PMT is only 0.6%, and calculations of the
sphere properties indicate that the PMT light detection efficiency is independent of scattering angle to within 1% (Onasch et
1140 al., 2015). As a result, Onasch et al. (2015) assumed that the ideal condition of $Z(\theta) = \sin \theta$ is applicable for the CAPS PM_{ssa}.
We make the same assumption here.

In other formulations of Eq. (A4), $Z(\theta)$ is typically expressed together with $\eta(\theta, \lambda)$ in a single function (e.g. Anderson et al.
(1996) refer to this single function as ‘ $f(\theta)$ ’, while Müller et al. (2011) use ‘ $Z_{is}(\theta)$ ’). We reformulate this function as two
1145 separate components to highlight the importance of $\eta(\theta, \lambda)$, which describes the efficiency with which a given instrument can
collect light. $\eta(\theta, \lambda)$ can vary between 0 (no light collected) and 1 (all light collected). Considering $\eta(\theta, \lambda)$ explicitly allows for
the simple and transparent introduction of additional physical processes into light scattering calculations (e.g. reflection from
the glass tube containing the aerosol sample, as discussed below). Additionally, comparison of the $\eta(\theta, \lambda)$ curves of different
instruments provides a clear and intuitive comparison of the abilities of the instruments to collect scattered light.

1150 Within this formulation the notion that an ideal integrating nephelometer collects scattered light over all possible directions is
expressed by the condition $\eta_{ideal}(\theta, \lambda) = 1$ for $0 \leq \theta \leq \pi$ rad (as shown in Fig. 3). Scattered light truncation in real integrating
nephelometers can be expressed by setting $\eta(\theta, \lambda)$ to 0 at angles where light is undetected. For example, for the specific case
of the well-characterized TSI 3563 cell-direct integrating nephelometer (TSI Inc., St. Paul, MN, USA),

$$1155 \quad \eta_{TSI\ 3563}(\theta, \lambda) = \begin{cases} \mathbf{1} & \text{if } \theta_1 < \theta < \theta_2 \\ \mathbf{0} & \text{if } 0 \leq \theta \leq \theta_1 \text{ or } \theta_2 \leq \theta \leq \pi \end{cases} \quad (\text{A6})$$

where $\theta_1 = 7^\circ$ and $\theta_2 = 170^\circ$ are referred to as the truncation angles of the instrument (Anderson and Ogren, 1998). For
integrating sphere type nephelometers like the one used in the CAPS PM_{ssa}, truncation occurs because scattered light escapes
through the aerosol sample entry and exit apertures in the sphere, as shown by the example truncation angles θ_1 and θ_2 in Fig.
1. In this case, the truncation angles depend on particle position along the longitudinal axis of the glass aerosol sample tube
(Onasch et al., 2015), which is indicated as the z -dimension in Fig. 1. It is also possible for particles beyond the boundaries of
the integrating sphere to contribute to the measured scattering signal by scattering light into the sphere (Varma et al., 2003).
In the CAPS PM_{ssa}, extra path lengths in the range from $l = 0$ to 4.7 cm outside the sphere boundaries must be considered.
The upper limit of this range is determined by the fixed positions of the aerosol flow tubing and ports in the optical cavity.
1165 Considering the instrument geometry shown in Fig. 1 the z -dependent truncation angles in the CAPS PM_{ssa} can be expressed
as:

$$\begin{aligned}
\theta_1(z) &= \begin{cases} \tan^{-1}\left(\frac{d/2}{L/2-z}\right) & \text{for } z \in [-(L/2+l), L/2] \\ \pi + \tan^{-1}\left(\frac{d/2}{L/2-z}\right) & \text{for } z \in [L/2, (L/2+l)] \end{cases} \\
\theta_2(z) &= \begin{cases} \pi + \tan^{-1}\left(\frac{d/2}{-L/2-z}\right) & \text{for } z \in [-L/2, (L/2+l)] \\ \tan^{-1}\left(\frac{d/2}{-L/2-z}\right) & \text{for } z \in [-(L/2+l), -L/2] \end{cases}
\end{aligned} \tag{A7}$$

Here it is assumed that the collimated light beam circulating in the optical cavity is confined along the central z -axis of the instrument with negligible width relative to the diameter of the glass sampling tube, and that multiple scattering effects from particles outside the collimated beam can be neglected.

Given these z -dependent truncations angles, and assuming that there is no scattered light reflection from the glass sampling tube ('no-refl'), the light collection efficiency function at a particular z -position (i.e., 'spot') in the CAPS PMssa optical cavity can be expressed as:

$$\eta_{spot_{meas}}^{no-refl}(\theta, \lambda, z) = \begin{cases} \mathbf{1} & \text{if } \theta_1(z) < \theta < \theta_2(z) \\ \mathbf{0} & \text{if } \mathbf{0} \leq \theta \leq \theta_1(z) \text{ or } \theta_2(z) \leq \theta \leq \pi \end{cases} \tag{A8}$$

The subscript 'meas' indicates that this is a function pertaining specifically to the CAPS PMssa (as opposed an ideal integrating nephelometer), following the notation in Eq. (A3).

In addition to highlighting the truncation angles, the η formulation also allows explicit introduction of additional physical processes that can reduce the probability of scattered light detection below 1 (which can be a function of both z and θ). In particular, we consider the process of reflection from the CAPS PMssa glass sampling tube (Fig. 1). This process is included in the radiative transfer theory model of Liu et al. (2018), but not the original scattered light truncation model presented by Onasch et al. (2015). To express this process in an η function we first calculate the probability R that light at an angle of incidence of $\theta_i (= \pi - \theta)$ is reflected from the interface between the glass sampling tube and air (with refractive indices m_{glass} and m_{air} , respectively) using the Fresnel equations for unpolarized incident light:

$$R(\theta_i, m_{glass}, m_{air}) = \frac{1}{2} \left(\left| \frac{m_{air} \cos \theta_i - m_{glass} \sqrt{1 - \left(\frac{m_{air}}{m_{glass}} \sin \theta_i\right)^2}}{m_{air} \cos \theta_i + m_{glass} \sqrt{1 - \left(\frac{m_{air}}{m_{glass}} \sin \theta_i\right)^2}} \right|^2 + \left| \frac{m_{glass} \cos \theta_i - m_{air} \sqrt{1 - \left(\frac{m_{air}}{m_{glass}} \sin \theta_i\right)^2}}{m_{glass} \cos \theta_i + m_{air} \sqrt{1 - \left(\frac{m_{air}}{m_{glass}} \sin \theta_i\right)^2}} \right|^2 \right). \quad (A9)$$

1190 In the present study we assume $m_{glass} = 1.5 + 0i$ and $m_{air} = 1 + 0i$.

We calculate the number of reflections N that light would need to undergo to exit the integrating sphere when scattered at an angle θ from a particle at position z as the floor of the ratio of the distance of the particle from a sphere exit (along the z -dimension), to the z -component of the distance that the light would travel between each reflection event from the glass tube.

1195 An extra term is added to the numerator of this ratio to reflect the assumption that the particle lies along the center-line of the sampling tube:

$$N(z, \theta) = \begin{cases} \left\lfloor \frac{\left| \frac{L}{2} - z + \left(\frac{d/2}{\tan \theta} \right) \right|}{d/\tan \theta} \right\rfloor & \text{if } z < L/2 & \text{if } \theta < \frac{\pi}{2} \\ 0 & \text{if } z \geq L/2 \\ \left\lfloor \frac{\left| \frac{L}{2} + z + \left(\frac{d/2}{\tan(\pi - \theta)} \right) \right|}{d/\tan(\pi - \theta)} \right\rfloor & \text{if } z > -L/2 & \text{if } \theta > \frac{\pi}{2} \\ 0 & \text{if } z \leq -L/2 \end{cases}. \quad (A10)$$

Equations (A9) and (A10) are calculated over the scattering angle range $0 \leq \theta \leq \pi$, and then combined to calculate a z - and
 1200 θ -dependent probability function that represents the total fraction of light reflected out of the integrating sphere. We term this function R_{total} :

$$R_{total}(z, \theta, \mathbf{m}_{glass}, \mathbf{m}_{air}) = R(\theta, \mathbf{m}_{glass}, \mathbf{m}_{air})^{N(z, \theta)} . \quad (A11)$$

For a particular z -position in the optical cavity, R_{total} can be combined with $\eta_{spot_{meas}^{no-refl}}(\theta, \lambda, z)$ as defined in Eq. (A8) in order to calculate a light collection efficiency function that takes account of the process of glass tube reflection:

$$\eta_{spot_{meas}^{refl}}(\theta, \lambda, z) = \eta_{spot_{meas}^{no-refl}}(\theta, \lambda, z) \cdot (1 - R_{total}) . \quad (A12)$$

Supplementary Fig. S16 displays example R , N , and R_{total} curves as a function of scattering angle θ for the case of a particle in the center of the optical cavity ($z = 0$). Figure 3 displays example $\eta_{spot_{meas}^{refl}}$ and $\eta_{spot_{meas}^{no-refl}}$ curves (black and blue solid lines, respectively) for five different positions in the cavity spanning the range from $z = -6$ to 6 cm.

A single, integrated light collection efficiency curve $\eta(\theta, \lambda)$ can be generated for use in Eq. (A4) by integrating an η_{spot} function over all possible z -positions. That is,

$$\eta(\theta, \lambda) = \frac{1}{L + 2l} \int_{-(L/2+l)}^{L/2+l} \eta_{spot}(\theta, \lambda, z) dz . \quad (A13)$$

This operation assumes that the aerosol particles in the instrument are homogeneously distributed along the central z -axis of the glass sampling tube, which is reasonable assumption to make for the aerosol number concentrations typically observed in the atmosphere (Qian et al., 2012). For concentrations much lower than this longer averaging times could be used to avoid any noise issues related to inhomogeneity. Example integrated light collection efficiency curves are displayed Fig. 3 for the cases where glass tube reflection is considered (black curve; ‘with glass reflection’) and is not considered (blue curve; ‘no glass reflection’). In the terminology presented in this Appendix, these are the curves $\eta_{meas}^{refl}(\theta, \mathbf{m}_{glass}, \mathbf{m}_{air})$ and $\eta_{meas}^{no-refl}(\theta)$, respectively, obtained by integrating the corresponding η_{spot} curves in Eq. (A13). The extra dependencies of η_{meas}^{refl} on \mathbf{m}_{glass} and \mathbf{m}_{air} come from the dependence of this function on R_{total} (Eq. A12).

All of the elements are now in place to use Eq. (A3) to calculate the truncation correction factor γ for some aerosol with scattering phase function $S_p(\theta, \lambda)$. $b_{sca, true}$ is calculated by substituting $S_p(\theta, \lambda)$, $Z(\theta) = \sin \theta$, and $\eta(\theta, \lambda) = \eta_{ideal}(\theta, \lambda) = 1$ into Eq. (A4). $b_{sca, meas}$ is calculated in the same manner, except $\eta(\theta, \lambda)$ is set to $\eta_{meas}^{refl}(\theta, \mathbf{m}_{glass}, \mathbf{m}_{air})$, if one wishes to account for the process of glass tube reflection, or $\eta_{meas}^{no-refl}(\theta)$ if one wishes to neglect this process. The normalization factor $k_{Rayleigh}$ is

calculated in a similar way using the Rayleigh scattering phase function (Eq. 5.6 in Bohren and Huffman, 1998) and again
 1230 assuming $Z(\theta) = \sin \theta$. Specifically,

$$k_{Rayleigh} = \frac{\int_0^\pi (1 + \cos^2 \theta) \sin \theta \eta_{meas}(\theta) d\theta}{\int_0^\pi (1 + \cos^2 \theta) \sin \theta \eta_{ideal}(\theta) d\theta} \quad . \quad (A14)$$

9. Appendix A2: Error model for the CAPS PMssa

To build an error model for the CAPS PMssa we begin with Eq. (2) from the main text, which is repeated here for convenience.
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$$\mathbf{b}_{abs} = \mathbf{b}_{ext} - \mathbf{b}_{sca} = \frac{1}{\alpha} \cdot (\mathbf{b}_{ext,sample} - \mathbf{b}_{ext,baseline}) - \frac{\gamma}{\beta} \cdot (\mathbf{b}_{sca,sample} - \mathbf{b}_{sca,baseline}) \quad . \quad (A15)$$

Individual uncertainty estimates for the seven parameters on the right hand side of this equation are given in Table 1. We
 assume that all of these errors are uncorrelated with each other. This is not generally true for the errors in the geometry
 correction factor α and the cross calibration constant β , since β is directly proportional to α (Eq. 3; note that this is not the case
 1240 for γ , since γ is defined as a normalized ratio). However, in the specific case of a cross-calibrated instrument, we take the errors
 in β to represent uncertainties arising from (i.e., precision) and after (i.e., drift) a cross calibration measurement. This part of
 the overall uncertainty in β is uncorrelated with the error in α .

The error model is then constructed by applying the standard rules of error propagation to Eq. (A15), given the assumption of
 1245 uncorrelated errors:

$$\delta \mathbf{b}_{abs} = \sqrt{(\delta \mathbf{b}_{ext})^2 + (\delta \mathbf{b}_{sca})^2} \quad , \quad (A16)$$

where

$$\delta \mathbf{b}_{ext} = \mathbf{b}_{ext} \cdot \sqrt{\left(\frac{\delta \alpha}{\alpha}\right)^2 + \left(\frac{\sqrt{(\delta \mathbf{b}_{ext,sample})^2 + (\delta \mathbf{b}_{ext,baseline})^2}}{\mathbf{b}_{ext,sample} - \mathbf{b}_{ext,baseline}}\right)^2} \quad , \quad (A17)$$

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and

$$\delta b_{sca} = b_{sca} \cdot \sqrt{\left(\frac{\delta\beta}{\beta}\right)^2 + \left(\frac{\delta\gamma}{\gamma}\right)^2 + \left(\frac{\sqrt{(\delta b_{sca,sample})^2 + (\delta b_{sca,baseline})^2}}{b_{sca,sample} - b_{sca,baseline}}\right)^2} . \quad (A18)$$

1255 The same quantities can also be used to calculate errors in SSA ($=b_{sca}/b_{ext}$) measured by the CAPS PMssa:

$$\frac{\delta SSA}{SSA} = \sqrt{\left(\frac{\delta b_{sca}}{b_{sca}}\right)^2 + \left(\frac{\delta b_{ext}}{b_{ext}}\right)^2} . \quad (A19)$$

10. Data availability

Data archiving is currently underway. Data will be available on Zenodo if the manuscript is accepted for publication.

1260 11. Competing interests

The authors declare that they have no conflicts of interest.

12. Author contributions

1265 RLM, JCC, and MGB developed the error model based on the theoretical description of the instrument. RLM developed the new truncation model presented in Appendix A1 together with MGB. RLM, BB, MI, and MGB designed and/or performed the laboratory measurements of cross calibration constants and truncation values. RLM and FL calculated truncation values to compare with the laboratory measurements. RLM and MGB designed the Bologna mobile experiment. MB and REP took the measurements and analyzed the raw data during the Bologna campaign. JCC and TM took the measurements and analyzed the raw data during the Melpitz campaign. JSH, MMM, KE, and MGB designed the Cabauw experiment. JSH coordinated the Cabauw campaign. RLM and PF took the measurements and analyzed the raw data during the Cabauw campaign. RLM
1270 performed the data analysis and interpretation and wrote the manuscript with input from JCC and MGB. All co-authors reviewed and commented on the manuscript.

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Table 1: Summary and description of uncertainties in the individual parameters comprising the error model described in Sect. 2.3. The precision column represents uncertainty due to the limited precision with which a particular parameter can be determined during calibration or measurement, and the drift column represents uncertainty due to possible drift of a parameter between available measurements. Estimated values are taken from previous studies or this study as indicated. The estimated values with units of Mm^{-1} correspond to absolute errors, and those with percentages relative errors.

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Parameter	Symbol	Precision	Drift (stability-based uncertainty)	Description	References
Sample extinction coefficient	$b_{ext,sample}$	1 Mm^{-1}	NA	Conservative estimate of short-term, random noise.	(Onasch et al., 2015)
Baseline extinction coefficient	$b_{ext,baseline}$	0.35 Mm^{-1}	0.3 Mm^{-1} over 10 mins (CAPS630b)	Values estimated from the Cabauw field dataset (Fig. S8).	This study
Sample scattering coefficient	$b_{sca,sample}$	1 Mm^{-1}	NA	Conservative estimate of short-term, random noise.	(Onasch et al., 2015)
Baseline scattering coefficient	$b_{sca,baseline}$	0.66 Mm^{-1}	0.1 Mm^{-1} over 10 mins (CAPS630b)	Values estimated from the Cabauw field dataset (Fig. S8).	This study
Geometry correction factor	α	1%	3% over 1 year	Drift value determined from regular CAPS PMex measurements at the European Center for Aerosol Calibration (ECAC; http://www.actris-ecac.eu/)	(Petzold et al., 2013; Pfeifer et al., 2020)

Scattering cross calibration factor	β	2% (CAPS450)	NA (CAPS450)	Values estimated from the Payerne (Fig. 5) and Cabauw (Sect. 6.2.2) field datasets.	This study
		2% (CAPS630a)	2% (CAPS630a)		
		2% (CAPS630b)	2.5% (CAPS630b)		
		6% (CAPS780)	8% (CAPS780)		
Truncation correction factor	γ	4% for fine-mode dominated aerosol	NA	Values derived from the sensitivity analysis discussed in Sect. 6.2.3.	This study
		9% for coarse-mode containing aerosol			

Table 2: CAPS PM_{ss}a instrument units that were used in the present study

Unit ID	Wavelength (nm)	Institute	Serial number	Geometry correction factor (α)
CAPS450	450	PSI	314003	0.78
CAPS630a	630	PSI	313004	0.71
CAPS630b	630	Demokritos	313003	0.7 – 0.73
CAPS780	780	PSI	314002	0.78

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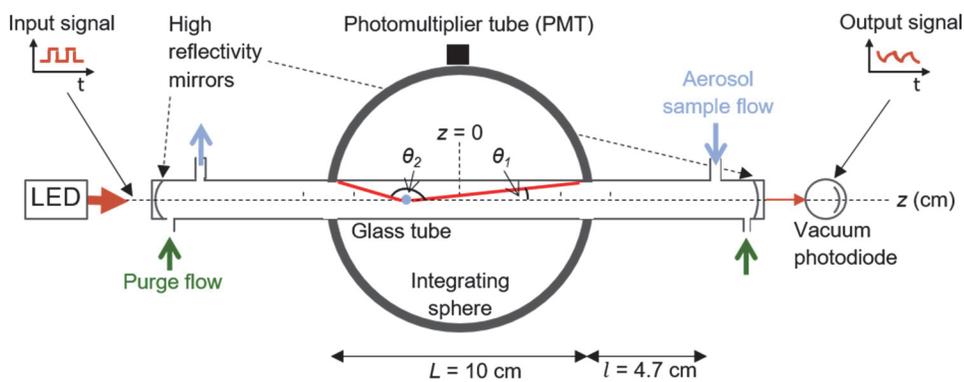
Table 3: Test aerosols used to measure scattered light truncation in the CAPS PM_{ss}a as a function of particle diameter and the values of parameters used in the corresponding model calculations. All particles were size classified by AAC.

Test aerosol	Refractive index (wavelength)	Geometric standard deviation of the size distributions	Dates-Number of experiment-repeat
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experiments at each wavelength (dates)

PSL spheres	$1.59 + 0i$ (at 450, 630 & 780 nm)	1.05 and 1.1	<u>2 at 450 nm, 3 at 630 & 780 nm (16 to 22-08-2019; 16 to 20-01-2020) between Apr. 2018 & Jan. 2020)</u>
DEHS (Di-Ethyl-Hexyl-Sebacat)	$1.46 + 0i$ (at 450 nm); $1.45 + 0i$ (at 630 & 780 nm)	1.1	<u>1 at 450, 630 & 780 nm (14 to 16-01-Jan. 2019) 26-04 to 08-05-2018; 16 to 30-08-2018) at 450 nm, 4 at 630 nm, & 2 at 780 nm (between Apr. & Aug. 2018)</u>
Ammonium sulphate	$1.520 + 0i$ (at 450, 630 & 780 nm)	1.1	

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1515 **Figure 1: Schematic diagram of the CAPS PMssa monitor with relevant components and variables highlighted. A glass tube encapsulates the aerosol sample to be measured. A light-emitting-diode (LED) delivers a square-wave modulated light signal as input to the optical cavity. The phase shift of the output signal from the cavity relative to the input signal is measured by a vacuum photodiode: this is the extinction channel of the instrument. Light scattered from the aerosol sample is collected by the integrating sphere and measured with a photomultiplier tube (PMT): this is the scattering channel of the instrument. θ_1 and θ_2 are the two truncation angles for light scattered from a particle at position z along the instrument axis (without considering reflection from the glass tube).**

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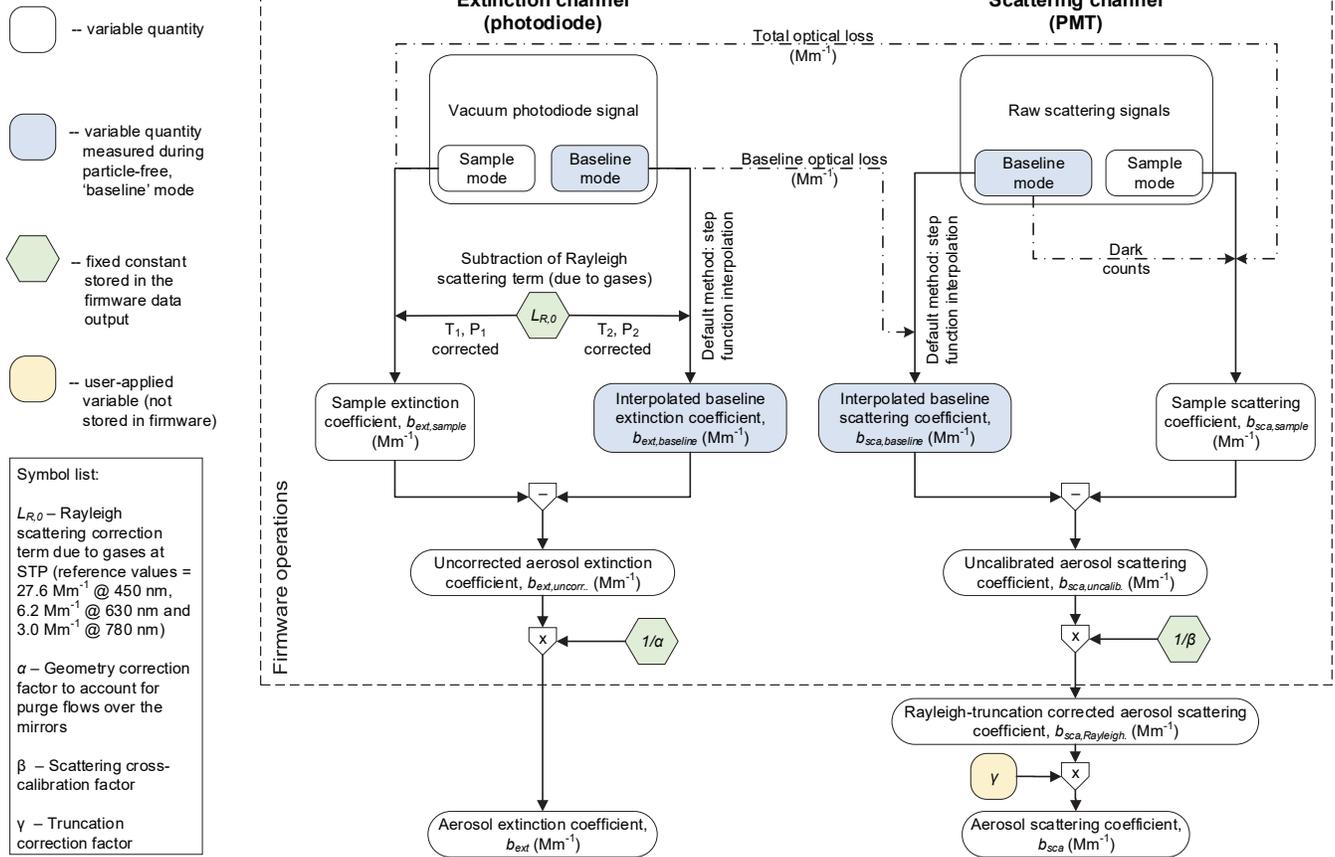


Figure 2: Data processing chain for the extinction and scattering channels of the CAPS PMssa. Blue boxes indicate quantities that are measured during the periodic ‘baseline’ mode of operation of the instrument. Hexagonal containers indicate fixed constants, rounded rectangular containers represent variable quantities.

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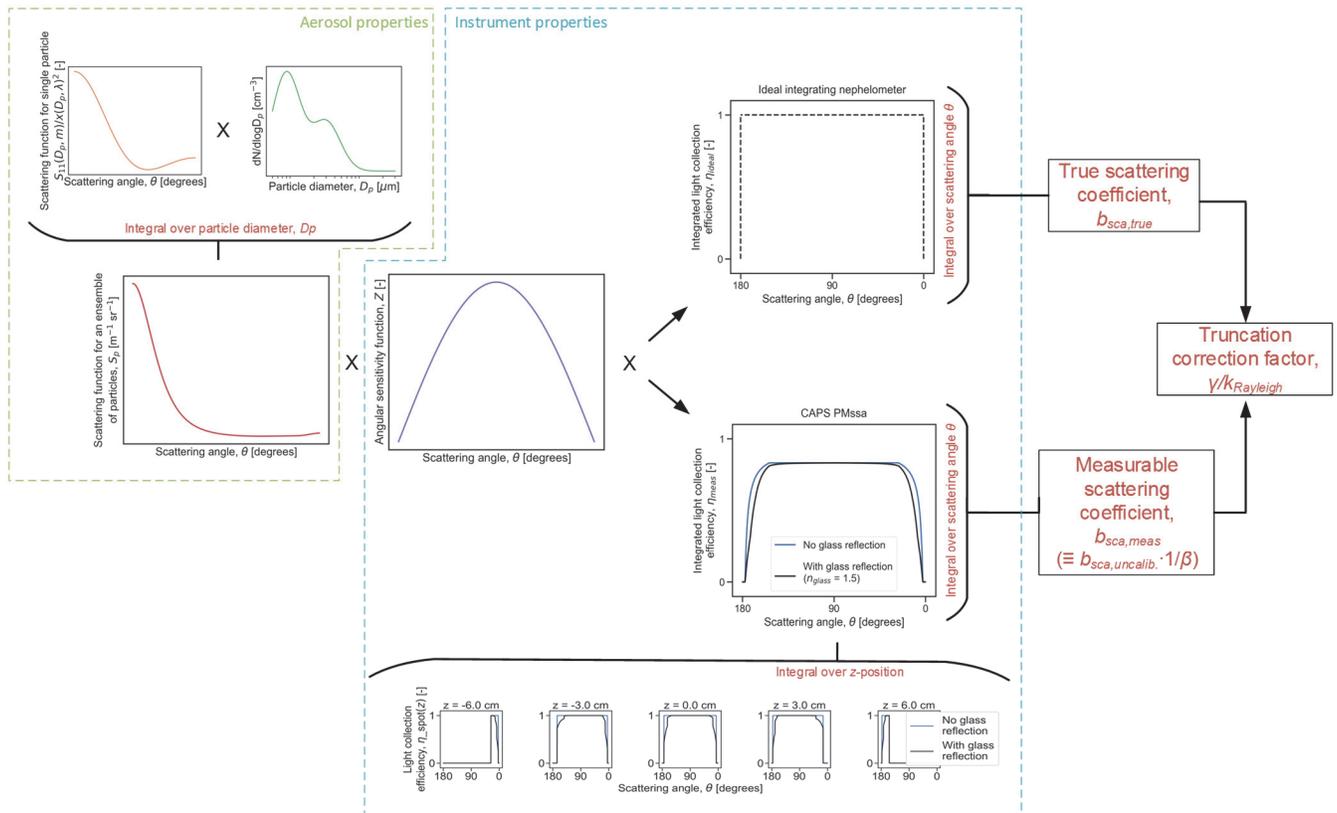
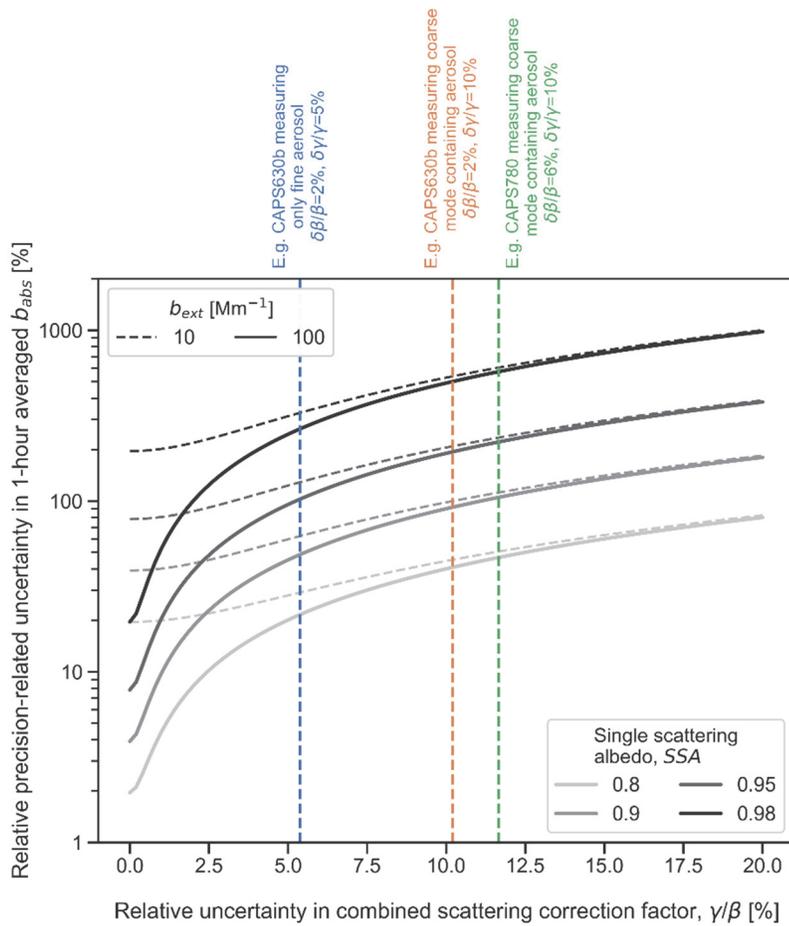


Figure 3: Schematic diagram of the new model for calculating truncation correction factors for the CAPS PMssa. Full details of the calculations are presented in Appendix A1. The model requires as input a light collection efficiency function and an angular sensitivity function, which are determined by the geometry of the CAPS PMssa optical system; and a scattered light intensity function for the ensemble of particles being measured, which is a function of the particle size distribution ($dN/d\log D_p$) and size-dependent aerosol scattering phase function. The main output of the model is the truncation correction factor, γ .

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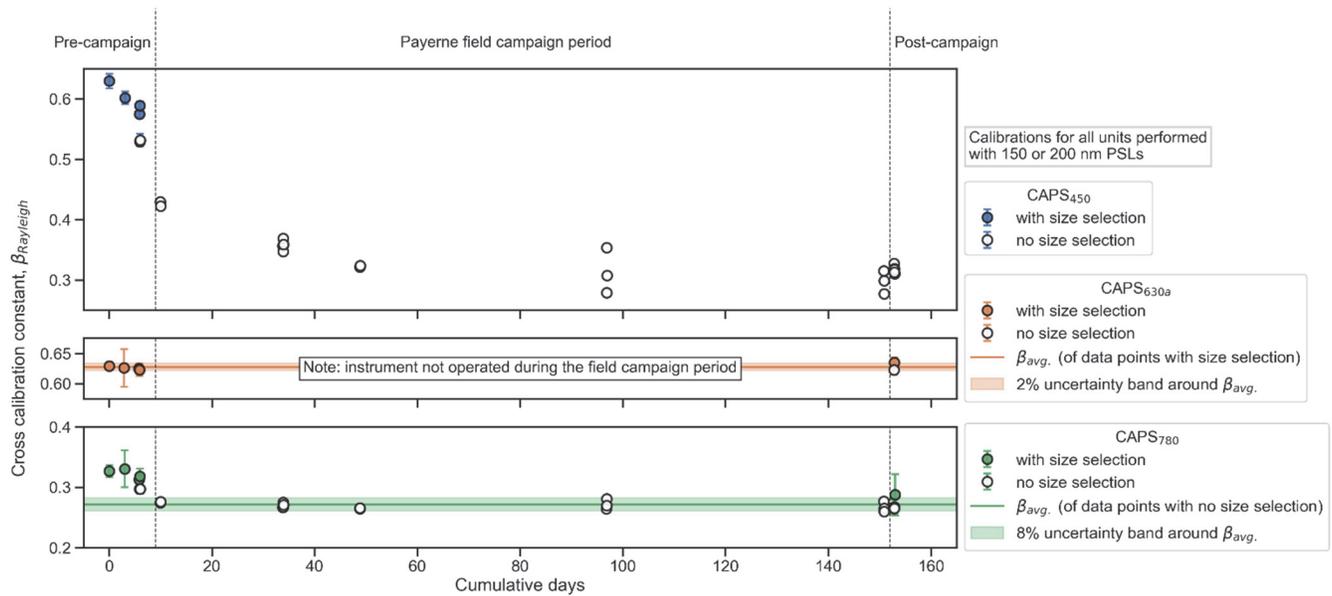


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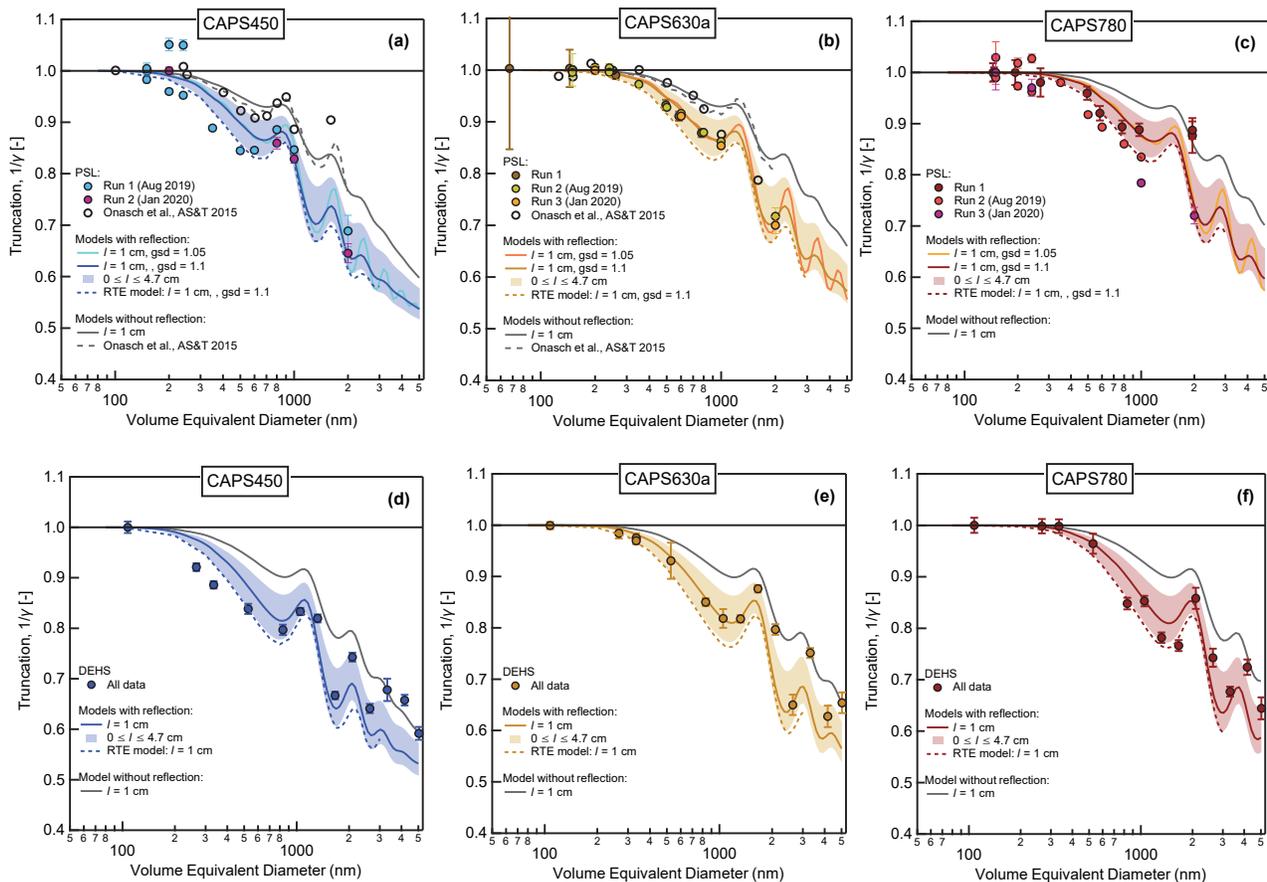
Figure 4: Theoretically-calculated relative uncertainty in 1-hour averaged CAPS PM_{2.5} b_{abs} measurements as a function of the relative uncertainty in the combined scattering correction factor (defined in Eq. 6 using the ratio of the truncation correction factor γ and the instrument cross-calibration factor β). Curves are shown for four different SSA values (grey shading) and two different aerosol loadings (b_{ext} of 10 and 100 Mm^{-1}). The curves were generated using the error model presented in Sect. 2.3 and Appendix A2 with inputs that were chosen to represent instrument characteristics during the Cabauw field campaign, as detailed in the main text.

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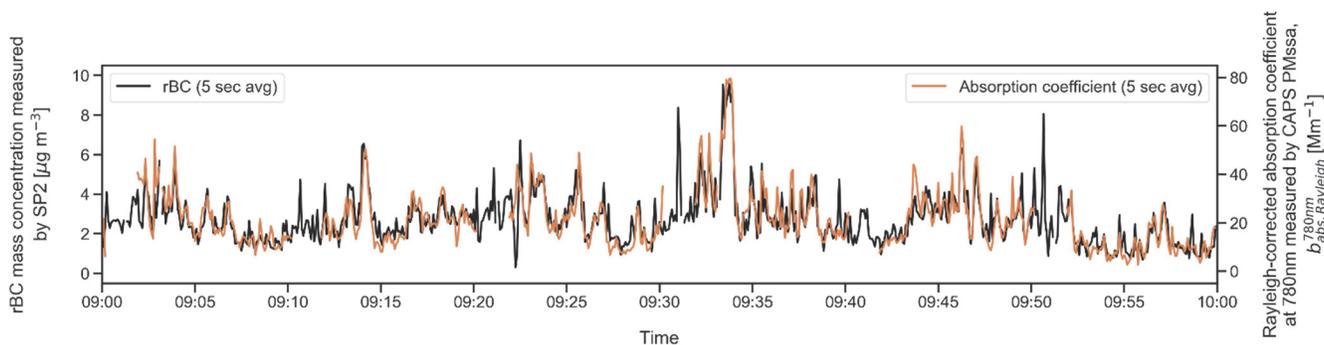
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1550 **Figure 5: Rayleigh-regime cross calibration constants ($\beta_{Rayleigh}$) for three CAPS PM_{ss}a units (CAPS450, CAPS630a, CAPS780) measured before, during and after the Payerne field campaign. Error bars indicate the standard deviation of the measured ratios used to determine each $\beta_{Rayleigh}$ value (see Sect. 3.2).**

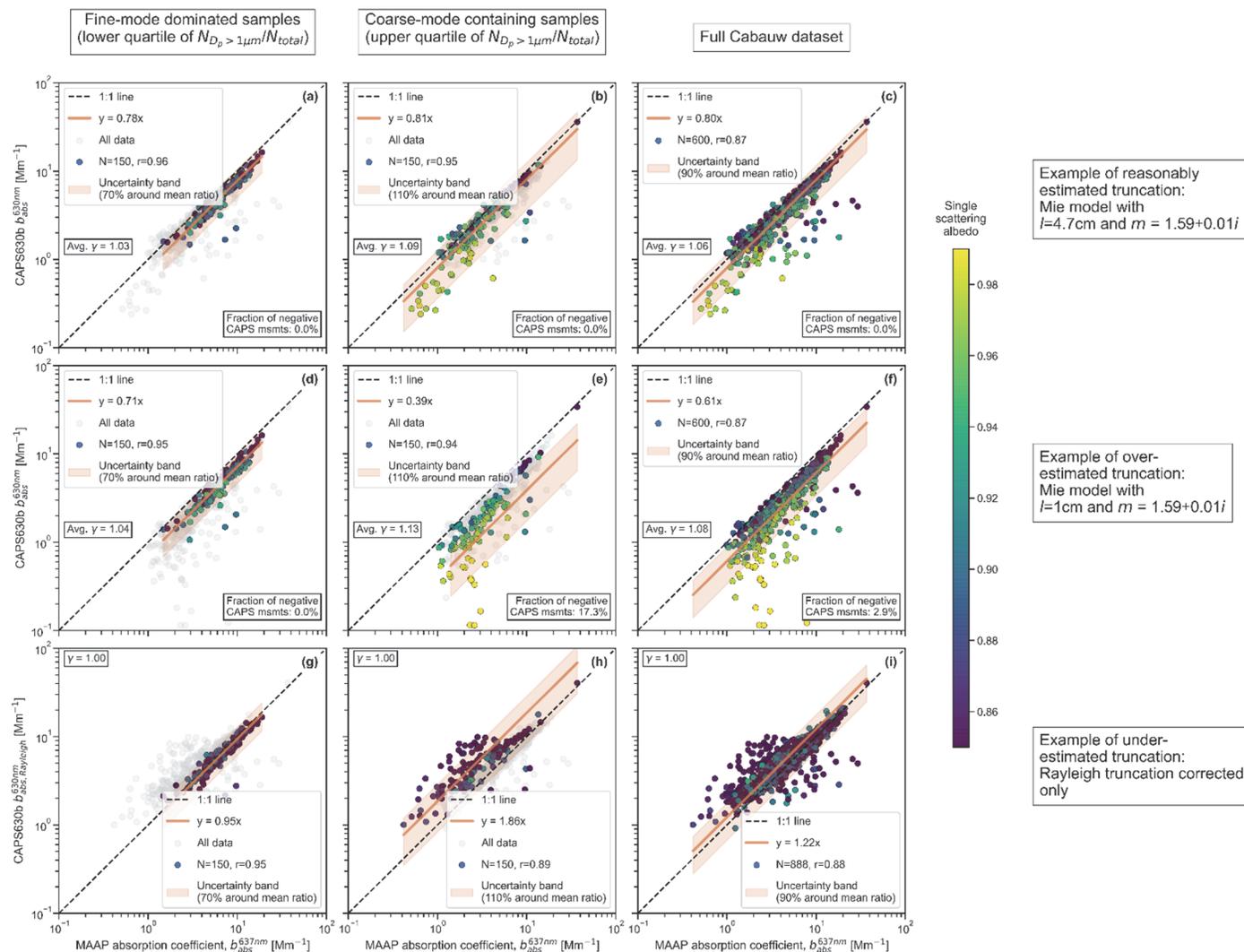


1555 **Figure 6: Measured and modeled truncation values as a function of volume equivalent particle diameter for PSL and DEHS aerosols. The truncation values plotted on the y-axes correspond to the inverse of the truncation correction factor γ defined by Eq. (4). Modeled curves were calculated with the truncation model presented in Appendix 1 as well as the radiative transfer equation (RTE) model presented by Liu et al. (2018). The parameter l represents the extra path length beyond the integrating sphere, and gsd refers to the geometric standard deviation of the modeled test aerosols.**



1560 **Figure 7: Time series of a 1-hour period of 5-sec averaged rBC mass concentration and absorption coefficient measurements made from the mobile laboratory traveling on a highway road during the Bologna campaign. rBC mass concentrations were measured by**

SP2 – a sensitive, single-particle-based instrument. Absorption coefficients were measured at 780 nm with the unit CAPS780. Note that the absorption coefficients have only been corrected for Rayleigh scattering truncation and therefore should not be interpreted quantitatively (e.g. they cannot be used to calculate **MAC**—mass absorption cross sections for the of BC—values). **In addition, the SP2 measurements have not been corrected for missing mass and should also not be interpreted quantitatively (Pileci et al., 2020b).** Spikes in the time series correspond to emissions from passing vehicles on the highway.



1570 **Figure 8:** Comparison of CAPS PMssa absorption coefficients at 630 nm (b_{abs}^{630nm}) and MAAP absorption coefficients at 637 nm (b_{abs}^{637nm}) for three different truncation correction scenarios. The three columns represent different subsets of the dataset, as indicated in the column titles. The three rows represent the different truncation correction scenarios, as indicated in the row labels. The top row (panels a-c) display CAPS PMssa measurements that were corrected using the Mie-based truncation correction scheme presented in Appendix A1 with m set to $1.59+0.01i$ and l set to 4.7 cm. The middle row (panels d-f) displays the same type of data with l set to 1 cm. The bottom row (panels g-i) contains CAPS PMssa measurements corrected for the truncation of Rayleigh scattering only (i.e., $\gamma = 1$). All data points are colored by the SSA values calculated from the same CAPS PMssa data shown in each panel. The mean ratios of CAPS PMssa to MAAP b_{abs} are plotted as solid orange lines in each panel. The uncertainty bands represent the 95th percentiles of theoretical uncertainties calculated with the error model and inputs described in Sect. 2.3, with relative

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1580 | uncertainties in γ of 4, 9, and 6% for the fine-mode dominated, coarse-mode containing, and full dataset, respectively. Uncertainty in MAAP b_{abs} of up to 7% can be expected based on previous studies (Section 3.1.1): this uncertainty is not displayed for visual clarity.