Review of Manuscript

Title: **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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Overview: This paper investigates the errors associated with the extinction, scattering, and absorption measurements of the recently commercialized CAPS PMssa 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 PMssa absorption measurement. The authors build a truncation model for the CAPS PMssa 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 PMssa 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.

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."
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 PMssa 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 PMssa 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.

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).
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.
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.
6.) Page 9 line 260 (and elsewhere), the authors note that the CAPS PMssa 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 PMssa 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.
7.) Page 9 line 273, "N2" should read "NO2"
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.
9.) Page 15 line 468, remove "anyway" from "anyway small relative"
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.
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.
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 PMssa instruments measure of particulate absorption, as purge failure is a potential issue for any CAPS PMssa or PMex system. At worse, this section/figure could cause readers to think that the CAPS PMssa 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 PMssa, 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.

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

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.

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 PMssa 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 m2/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 ~1.5x assuming a 1/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.

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 PMssa 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.

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.

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.

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.