Answers to the Reviewer#1

We thank the anonymous reviewer #1 for the detailed review. Including the suggestions made has significantly enhanced the paper. In the following you will find our response to the reviewer directly marked in red.

Anonymous Referee #1

Received and published: 29 May 2019

This manuscript provides an analysis of a set of laboratory experiments comparing the recently developed CAPS PMssa instrument, which measures aerosol extinction and absorption (from which is derived single-scatter albedo) using cavity phase-shift and integrating sphere techniques, respectively. Because this instrument can determine SSA through from a single sample, and bypasses the need for relatively uncertain ab- sorption measurements using filter media, it has the potential to be extremely valuable. The aerosol direct effect remains a large uncertainty in the Earth's radiative balance, and this instrument, if accurate and widely used, has the potential to help reduce this uncertainty. The topic is entirely within the scope of AMT and there should be many readers interested in the subject. The experiments described in the manuscript appear to be well conducted and have produced high quality data.

Regrettably, there are some significant issues with the manuscript, two especially, that will require a major revision. These two issues are:

1) There is no error analysis of the techniques being compared. Instead, variance in the measurements is used as surrogate for uncertainty. The authors need to directly and independently determine the uncertainty in the phase-shift extinction measurement and the uncertainty in the scattering measurement, and propagate these uncertainties through to the final SSA product. This uncertainty analysis must include consideration of both potential biases (which might include determination of pressure and temperature in the instrument, for example) and random uncertainties (which might include noise in the measurement that requires averaging). As it currently stands with this manuscript, if I were to purchase two of the CAPS PMssa instruments and compare them and find that they disagree by 5% in extinction and/or scattering, I would not know if this is within expectations or would indicate a problem with one of the instruments. A complete uncertainty analysis needs to be applied to all the instrument combinations used. In Table 6, it is not at all clear where these "uncertainties" come from; they appear to be either the scatter in the data (plotted in Fig. 12) or else the difference between the mean values of the data and the "expected" values of the calibration aerosols. Because the SSA of the calibration aerosols is not truly known, there is no absolute standard provided against which to evaluate the different approaches to measuring SSA, so a fundamental uncertainty propagation is needed.

This paper does NOT address in any explicit way, nor was it designed to address, the question of the absolute uncertainties of these techniques. It was designed to address the question of how well they correlate. Thus, the results are given in correlation coefficients (slope and intercept) and their statistical uncertainties

We have added Section 2.3 (new) summarizing the measurement uncertainties reported by the different instrument paper. In particular the uncertainty of the CAPS PMssa was described in detail by Onasch et al. (2013). We have added a passage to the manuscript referencing the detailed error analysis by Onash et al. more pronounced. Temperature and pressure variabilities as potential biases are part of the used measurement error and minimized by regular taking baseline measurements.

2) The linear regressions which provide the bases for the evaluated values appear to have some sort of error. Examining Fig. 4, the fitted line shown on the scatterplot lies below all of the datapoints. Using Table A1, I plotted the data shown in Fig. 4 and performed a linear regression. I got a slope of ~1.08 and the fitted line passed directly through the data. This compares with the "all" fit shown in Table 3 of the manuscript, which gives a slope of 0.97. I tried a two-sided regression, a one-sided regression, and one-sided regression forced through zero intercept. All gave slopes >1.06. Inspecting the other scatterplots in the manuscript (e.g. Figs. 8, Fig. 11 for absorptions <70 Mm-1), the fitted slopes do not seem to go through the data. Unless I've made an error, it appears that the values appearing in all the tables are suspect because of this fitting issue. Thank you for including all the data in the supplemental material tables, which makes finding an apparent problem like this easier.

The line shown in the linear regression was misplaced. The line is actually just a 1-1 line to help readers to evaluate the results in comparison to a perfect correlation (1-1). We have replaced the figures and added the information to the figure caption.

In addition to these two principal issues, there are some smaller items that need addressing.

a) The table captions all need to be more precise. For example, Table 3 might have a caption of, "Linear regression parameters slope (m), intercept (b) and their standard deviations and the linear regression coefficient R2."

We have added your suggestion to the captions.

b) I was trying to understand for quite some time how the column labeled "SSA" in Table 3 was calculated before realizing that it is simply an estimate of the SSA for the aerosol type being generated. It might be clearer to move the SSA column to the second column of the table and label it "estimated SSA".

We have added your suggestion to the table.

c) The figures all need to use a heavier line width and larger, denser font. It is quite hard to read the labels and identify the symbols and lines.

We have improved the figures resolution. But a final version (more readable) of each figure will depend on the layout specified by the AMT. Therefore, we will wait until the final version of the article is given (format wise) to optimize the figures sizes and resolutions.

d) The descriptions in the tables and figures of "PSAP-Neph" is confusing; it suggests that you are subtracting the scattering data from the absorption data. I suggest you use PSAP+Neph for Table 1 and Fig. 4, and "PSAP & Neph" for Table 6.

We have added your suggestion

e) Figs. 5 and 9 are not needed since the data appear in tables already.

It is a visual results representation, which we believe is important to many readers.

f) Fig. 12 should also show the SSA determined for the ammonium sulfate aerosol; this would give a good idea of the scatter about a known, non-absorbing compound.

We have added the AS Data to Figure 14 (new)

g) The title includes "Field Deployment". There is no field deployment of the instrument described in this manuscript, just laboratory tests.

Good point! The field deployment was excluded from this article, thus the title has been modified.

h) Lines 93 to 97, the description of the roles of MFC#1 and #2 in regulating make-up air appear to be switched with MFC#3 and #4 in Fig. 1.

Right. It is corrected (lines 96-97) now.

i) Section 2, please describe the truncation angles for the various instruments and typical magnitudes of the correction factors. The uncertainty in these correction factors need to be part of the total uncertainty analysis and error propagation.

There are many studies about the truncation angles and corrections for the proven technologies (most important and used ones are referred in the article). For the SSA Monitor the information has been added to section 2 (lines 196-204) in the description of the instrument, since it is what is being evaluated in this article. For the other instruments, we included the uncertainty section 2-3 citing the relevant literature in a pronounced part.

j) Please use 2-sided (orthogonal distance) regression when performing the linear regressions. There are uncertainties in both x and y dimensions that should be accounted for. Please weight the regressions by the uncertainty in the measurements.

All uncertainties (standard deviations) are presented for both x and y. Sometimes the values are so small that they are smaller than the data point marker.

k) The figure captions (e.g., Fig. 3: "Time series of the measurements by the extinction channel" do not adequately describe the contents of the figures, which in this case shows results from 3 different instruments/combinations, not just the extinction channel of the CAPS PMssa. The same for the other time plots.

We have updated the captions.

There is a lot of good information from some carefully performed and important lab- oratory studies in the manuscript. I encourage the authors to address the concerns indicated above and submit a revised manuscript that more fundamentally addresses uncertainties and that uses accurately determined regression slopes using properly weighted 2-sided linear regressions.

Answers to the Reviewer#2

We thank the anonymous reviewer#2 for the detailed review. Including the suggestions by the referee has significantly enhanced the paper. In the following you will find our response to the reviewer directly marked in red.

REVIEW NOTES and comments:

Laboratory Validation and Field Deployment of a Compact Single-Scattering (SSA) Albedo Monitor

Journal: AMT
Title: Laboratory Validation and Field Deployment of a Compact Single-Scattering Albedo (SSA) Monitor
Author(s): Julia Perim de Faria et al.
MS No.: amt-2019-146
MS Type: Research article

Matrix Scores: Criteria:

Scientific Significance – Good 2

Scientific Quality – Fair 4

Presentation Quality - Fair

Over all English language presentation:

There is a general non-standard usage of comma separators, and a few general awkward English syntax constructions. However, it seems most intended meanings are clear. The paper could use revision of grammatical and syntactical usage to make the reading flow more smoothly.

Over all Scientific Presentation:

General lack of the definition and standard used for the terms accuracy and precision. There should be at least an equation presented for the calculation assumed in each measure. It is important as the system of closure for the complete instrumental experimental circuit depends not only upon the accuracy and precision of each individual instrument, but the data path through all of them.

The study could be enhanced by a true presentation of error propagation by classical form differential error analysis. The assumption of normally [Gaussian} distributed error seems perhaps unfounded in such a complicated closure strategy.

The work has merit and should be published conditional upon appropriate revisions and additions.
Specifics:

Table1: the mixture of AS+AD is assumed to have an SSA λ=630nm of 0.6 for the study case, but lacks details in discussion of how the mixture of the standard substances was to be controlled.

The SSA of the mixture containing AS and AD, was controlled by the online measured SSA measured by the CAPS $PM_{ssa.}$ See: Line 80-81 in the revised version.

…. The SSA of the mixture containing AS and AD, was controlled by the online measured SSA measured by the CAPS PMssa. …

We also added to the head of the table 1) that this is an expected/estimated value.

Lines 100 – 105:

Perhaps some calibration data could be presented, as well as plot of Scattering Channel signal vs. Extinction Channel signal. This could provide insight into baseline fluctuations and possible instrumental bias.

We have added Figure 4 (new) to the revised version.

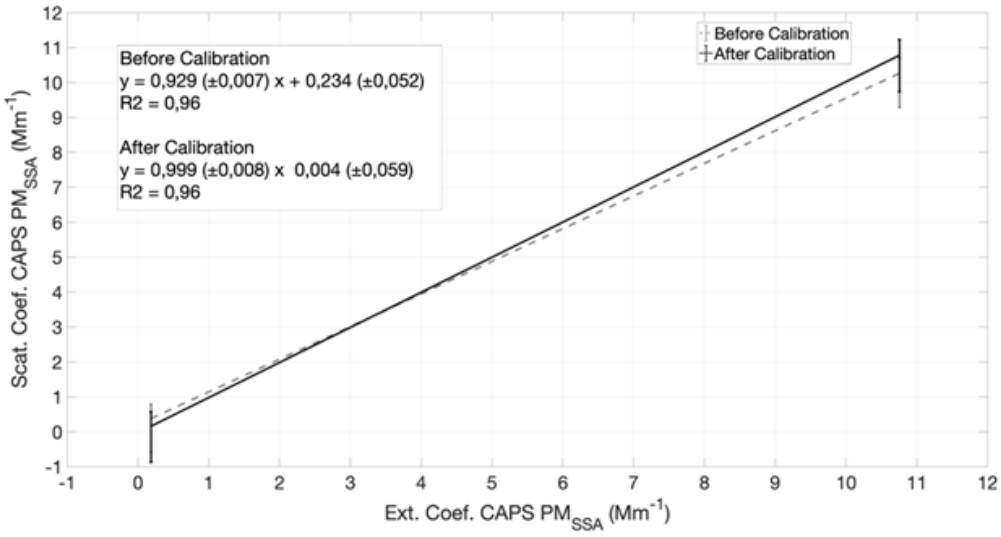

Figure 4. Scatterplot of the correlation of the extinction and scattering channel of the CAPS PMssa before and after the calibration using CO2.

Section 2.1.2

Lines 115 -120 “The instrument measures.....”

We have complemented the section.

Section 2.1.3 CAPS PMext configuration

It might be beneficial to include a figure as nicely detailed as that of Figure 2. For the CAPS PMssa configuration.

Figure 2 (new) has been added – although we still have to check the rights to publish a figure from another journal.

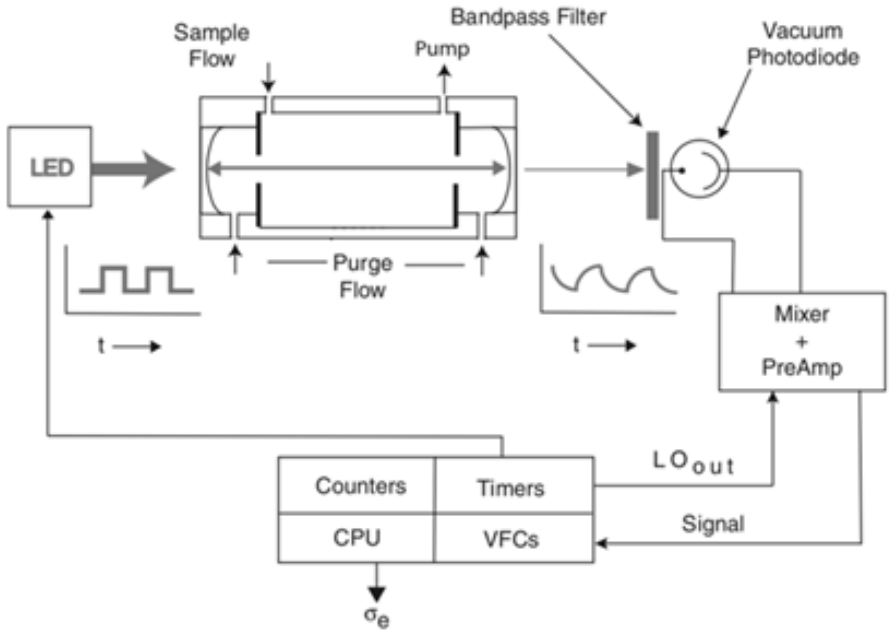

Figure 2. Overview of the main components and operation principle of the CAPS PMex instrument (Massoli et al., 2010)

Lines 140-145

One of the unique features of the CAPS PMssa set-up is the integrating sphere. The glass tube that passes through the sphere needs a bit more detailed information as it is inside the integrator. Some specifics as to the thickness of the wall, any coating it may have, it's optical properties should be characterized or listed somewhere from the manufacturer or supplier - if not determined during calibration of the instrument itself.

We have added more information in the lines 185 – 193

Petzold at al. (2013) showed that this purge flow shortens the measurement path and dilutes the sample and requires a correction factor. As done for the CAPS PM_{ex}, a new correction factor was developed, by using monodisperse polystyrene spheres (PSL) of know size, for the CAPS PM_{ssa}. Due to the cell geometry, the new correction factor was slightly larger than the one found for the extinction monitor, 1.37 and 1.27, respectively (Onasch et al., 2015). The noise of the instrument, trunction angle and instrument uncertainty have also been studied by Onasch et al. (2015). The values found were all below 1 Mm^{-1} for the noise levels (1σ, 1s) for all wavelengths. For the case of this particular instrument (630 nm), the truncation correction was determined below 4% for typical ambient conditions. The uncertainty was estimated at ±0.03 for SSA equal to 1 (PSL and ammonium sulphate) and decreases to ±0.01 as the SSA goes down. (630 nm), the truncation correction was determined below 4% for typical ambient conditions. The uncertainty was estimated at ±0.03 for SSA equal to 1 (PSL and ammonium sulphate) and decreases to ±0.01 as the SSA goes down.

For more details we have to reference Onasch et al. (2013).

[A general Question: Were any other wavelengths considered or tested for the calibration standard? - No]

Section 3 discussion:
Some of the sentences could be divided into shorter more clear constructions Lines 210 – 215:

1) I think these critical figures could be sized up a bit

We have improved the resolution of the figures, but the final aspects will be determined by the layout from AMT, thus we will wait until the final version (format wise) is done to work on this issue.

2) There seems to be a general assumption that the standard deviation is the most reliable measure of experimental uncertainty. This reviewer is not sure this is a completely valid assumption.

The Reviewer is right, as long as the standard deviation relays on the assumption that the statistical population follow a Gausian distribution. We have not proven the assumption by applying a Chi square fitting test, but the assumption is not too bad considering the frequency of occurrence diagram (Figure 8 in the revised version) . We have chosen the standard deviation and not a quartile distance (used as distribution free measure) because most readers are used to- and will ask for it.

3) The reference to PSAP-NEPH extinction measurements being similar to those of Petzold 2013: this unfortunately requires the reader to find the other paper to validate this statement of event or know what the expected result was. A simple sentence could clarify this. [Yes, as one of the contributing authors it is perfectly acceptable to cite their own previous research articles, but perhaps a bit much to expect the reader of this article to be familiar with the result of that work.]

We have added lines 268-269 :

…similar to the one found by Petzold et al. (2013), in which an excellent correlation (slope of 0.99) was found for the laboratory comparison between the same instruments using highly absorbing aerosol, exclusive scattering aerosol and mixtures of both"

Section 3.2

Line 217: " There is no systematic error found neither in the average nor in the standard deviation of the measured values." Although the internal reference is to a table included in the supplemental material, it is a mathematically unsupported assertion. A calculation or insight into how this statement is evidenced might make a stronger case for its inclusion.

The reviewer is right. We have changed the sentence

Within the error bars of the two instruments we could not observe a systematic deviation of both either in the average or in the standard deviation of the measured values.

Section 3.3

Figure10: is problematic on multiple levels: although the notion of overlying *timeseries* into a single track representing the CAPS PMssa and PSAP for the three type of aerosol particles testing is a good idea, the diagram has flaws. [same comments apply to Figure 7 on the scattering channel;]

1) The figures do not expand into full size charts and are presented TOO SMALL to intuit any scientific sense from their visual examination. [This may be a display result after the Copernicus online system was revamped for their paper display] But the authors could simple make a much larger figure.

We have improved the resolution of the figures, but the final aspects will be determined by the layout from AMT, thus we will wait until the final version (format wise) is done to work on this issue.

2) The horizontal axis has numbers on a scale with no mention or label as to their units. Are they "seconds" after the calibration sequence has finished? Are they minutes?

The Unit (seconds) has been added to the axis label.

3) Even if the individual axis numbers align, there is not a mention to assure the reader they were simultaneously measured.

We have clarified this in the Figure caption.

4) These figures as a set need to be amplified in the vertical scale so as to make visible Any regions in time where the CAPS PMssa signal fluctuations and spikes might not be synchronous to those of the supposed time coincident signal of the PSAP.

Same answer about the figures.

5) Expanding the horizontal time axes will allow the reader to view regions where the signals might not be precisely time correlated and any instrumental fluctuations as "noise."

This would require an interactive zooming option. Unfortunately this is not supported by AMT.

Discussion of this diagram is not complete. No mention is made to the significance of the regions where the traces converge over time to a common point in the AD and BC examples. No mention of the significance, if any, of time intervals where the signals step down, or step-up in sigma (σ).

This increase/decrease is seen in figure 12 for the absorption. The explanation is added:

"The increase in the absorption coefficient observed in Figure 12 for the higher levels of AD and BC, is related to the transmission decay of the filter in the PSAP and the correction algorithm chosen for this study."

NOTE: as mentioned prior it is not sufficient to cite a method "data correction" (Ogren 2010) without explaining why it is appropriate in this situation and how it fundamentally treats the data. Forcing the reader to find another paper to understand what is going on in this paper is not exercising good

scientific communication skill. There is nothing wrong with the citation of Ogren 2010, simply the authors here should explain how and why is it used, as well as it's importance to the data collected in this research.

We have added all correction functions to the text for completeness. But we did not motivate them in all the details.

I would like to state that it is good practice to reference a data correction algorithms used to the paper without explaining them in all detail as long as they are commonly rated as best practice. If we would have chosen a new/or exotic algorithm then the referee is right. Scientific papers relay on the referencing system. Otherwise the wheel has to be invented again and again- and articles would be more like textbooks. It is no argument that it is some work for a reader to search for a paper. This is part of our job! On the other hand, reading the original literature gives the original authors the credit they should get.

It should also be noted that without a time series analysis proper [lag correlation, etc. as an example] there is not a reliable method to indicate how the static correlation coefficients presented in the table evolve over time as the instruments run. Correlation coefficients are important as measures, but should state clearly they might not reveal complex interrelationships between data signals as the instruments run over time.

In particular the lag correlation (auto- and cross-correlation) does not help for this kind of lab studies. We did not test the instruments dynamically for transfer- response- or relaxation-times. This would be part of an extra paper. A detailed time series analysis is needed for field measurements. The way we address this is to do the test for several runs repeatedly. Thus the reported correlation is not just a snapshot.

Section 3.4
This is the key portion of the research and should be strongly emphasized. Generally well done.

Line 281: does the statement "....expected values for each aerosol type" directly refer to table 1? If so, reiterate that. If not, please summarize the expected values directly here.

We have added the internal reference (Table 7).

THIS IS ENOUGH TO WORK ON FOR now

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Laboratory Validation ~~and Field Deployment~~ of a Compact Single-Scattering Albedo (SSA) Monitor

Julia Perim de Faria[1], Ulrich Bundke[1], *, Andrew Freedman[2], Timothy B. Onasch[2], and Andreas Petzold[1]

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Abstract. An evaluation of the performance and accuracy of a Cavity Attenuated Phase-Shift Single Scattering Albedo Monitor (CAPS PM_{ssa}, Aerodyne Res. Inc.) was conducted in an optical closure study with proven technologies: Cavity Attenuated Phase-Shift Particle Extinction Monitor (CAPS PM_{ex}, Aerodyne Res. Inc.); 3-wavelengh Integrating Nephelometer (TSI Model 3563); and 3-wavelength filter-based Particle Soot Absorption Photometer (PSAP, Radiance Research). The evaluation was conducted by connecting the instruments to a controlled aerosol generation system and comparing the measured scattering, extinction, and absorption coefficients measured by the CAPS PM_{ssa} with the independent measurements. Three different particle types were used to generate aerosol samples with single-scattering albedos (SSA) ranging from 0.4 to 1.0 at 630 nm wavelength. The CAPS PM_{ssa} measurements compared well with the proven technologies. Extinction measurement comparisons exhibited a slope of the linear regression line for the full data set of 0.96 (-0.02/+0.06), an intercept near zero, and a regression coefficient R^2>0.99; whereas, scattering measurements had a slope of 1.01 (-0.07/+0.06), an intercept of less than +/-2×10^{-6} m^{-1} (Mm^{-1}), and a coefficient R^2~1.0. The derived CAPS PM_{ssa} absorption compared well to the PSAP measurements at low levels (< 70 Mm^{-1}) for the small particle sizes and modest (0.4 to 0.6) SSA values tested, with a linear regression slope of 1.0, an intercept of -4 Mm^{-1}, and a coefficient R^2=0.97. Comparisons at higher particle loadings were compromised by loading effects on the PSAP filters. For the SSA measurements, agreement was highest (regression slopes within 1%) for SSA = 1.0 particles, though the difference between the measured values increased to 9% for extinction coefficients lower than 55 Mm^{-1}. SSA measurements for absorbing particles exhibited absolute differences up to 18%, though it is not clear which measurement had the lowest accuracy. For a given particle type, the CAPS PM_{ssa} instrument exhibited the lowest scatter around the average. This study demonstrates that the CAPS PM_{ssa} is a robust and reliable instrument for the direct measurement of the scattering and extinction coefficients and thus SSA. This conclusion also holds as well for the indirect measurement of the absorption coefficient with the constraint that the accuracy of this particular measurement degrades as the SSA and particle size increases.

Keywords: CAPS PM_{ssa}, optical closure, single scattering albedo.

1 Introduction

Airborne aerosols impact climate directly though the interaction with incident solar light by scattering, generating a cooling effect, or by absorbing it and reemitting infrared radiation, having a heating effect. ~~According to Haywood and Shine (1995)~~According to Haywood and Shine (1995), the effect of aerosols on the atmospheric radiation budget in the visible spectral range depends on the aerosols optical depth (AOD), the single-scattering albedo (SSA), and the backscattered fraction (BF). The radiative forcing efficiency (RFE) describes the resulting aerosol direct forcing per unit AOD (~~Andrews et al., 2011;~~ Haywood and Shine, 1995; Andrews et al., 2011; Sheridan et al., 2012) and is widely used to describing the

radiative impact of a given aerosol type. As an aerosol intensive parameter the RFE value depends only on SSA and BF. As is stated in the latest IPCC report ~~(Boucher et al., 2013)~~(Boucher et al., 2013), uncertainties in SSA and the vertical distribution of aerosol contribute significantly to the overall uncertainties in the direct aerosol radiative forcing, while AOD and aerosol size distribution are relatively well constrained.

The measurement of SSA requires the simultaneous but independent observation of two parameters since, by definition, the SSA is the ratio of the scattering to the extinction coefficient (where extinction is the sum of the scattering and absorption – see Equation (1) and (2); the index p refers to the contribution of aerosol particles to overall light extinction, which has also a contribution by gas molecules, identified by the index g not shown in the equation).

$$\sigma_{ep} = \sigma_{ap} + \sigma_{sp} \quad (1)$$

$$SSA = {}^{\sigma_{sp}}/_{\sigma_{ep}} \quad (2)$$

Measuring all three aerosol optical parameters independently allows for the closure of optical properties and thus the determination of uncertainties of the involved instruments.

The aerosol optical parameters are typically measured *in-situ* by instruments such as Integrating Nephelometers (NEPH) for the scattering coefficient ~~(Heintzenberg and Charlson, 1996)~~(Heintzenberg and Charlson, 1996); photoacoustic (see e.g., ~~Lack et al. (2006)~~Lack et al. (2006); Arnott et al. (2006)) and filter-based methods such as the Particle-Soot Absorption Photometer (PSAP; ~~Bond et al. (1999)~~Bond et al. (1999)), the Multi Angle Absorption Photometer (MAAP; ~~Petzold and Schönlinner (2004)~~Petzold and Schönlinner (2004)) and more recently the Tricolor Absorption Photometer (TAP~~; Ogren et al. (2017)~~/CLAP; Ogren et al. (2017)) for the absorption coefficient; and for the extinction coefficient, the Cavity Ring Down (CRD) technology ~~(Moosmüller et al., 2005)~~(Moosmüller et al., 2005) or, since 2007, the Cavity Attenuated Phase Shift Particle Extinction Monitor (CAPS PM_{ex}) (Massoli et al., 2010). To measure the SSA using the optical closure approach involves separate instruments with different principles and uncertainties, leading to potential sources of significant errors and biases.

A novel instrument based on cavity attenuated phase-shift technology and incorporating an integrating sphere was recently developed by Aerodyne Research, Inc. This novel instrument represents a major step forward in the observation of aerosol optical properties since it simultaneously measures two of the three aerosol optical parameters from the same air sample, reducing the potential sources of sampling biases (Onasch et al., 2015). The two main applications of the CAPS PM_{ssa} instrument, apart from the direct measurement of scattering and extinction coefficients, are the indirect measurement of the aerosol absorption coefficient and the measurement of the single-scattering albedo. A few recent *in-situ* application studies of the CAPS PM_{ssa} instrument are already available (Han et al., 2017;Corbin et al., 2018~~; Han et al., 2017~~). The present optical closure study intends to quantify uncertainties in the measurement of the primary aerosol optical properties and the resulting SSA by the CAPS PM_{ssa} for several types of laboratory aerosol by applying a full set of established instrumentation for measuring the extinction (CAPS PM_{ex}), absorption (PSAP), and scattering (Integrating Nephelometer TSI Model 3563) coefficients at multiple wavelengths.

2 Instruments and Methods

2.1 Instrumental Set-up

The laboratory study was conceived to evaluate the operational principle of the CAPS PM_{ssa} and its performance and accuracy when compared to proven technologies. The instrumental set-up used is shown in Figure 1.

In this study, similar to previous work (Massoli et al., 2010;–Petzold et al., 2013); two collision-type aerosol generators (TSI Model 3076) were used; one containing a solution of deionized water and purely scattering aerosol, Ammonium Sulphate (AS), and a second containing absorbing aerosol, water-soluble colloidal graphite (Aquadag – AD – from Agar Scientific) or Black Carbon (REGAL 400R Pigment Black – BC – from Cabot Corporation). The SSA of the dispersed aerosol ranged from approximately 0.4 (pure AD or BC) to 1.0 (pure AS), with the modal value of the particle size distribution being below 100 nm in all cases. A drying tube filled with silica gel was positioned after each particle generator in order to reduce the relative humidity below 30%. Once the samples were passed through the dryer, they entered a mixing chamber where effective ensemble particle SSA values of 0.4 < SSA < 1.0 could be produced by mixing aerosol flows containing both absorbing and scattering aerosols. The aerosol generation set-up specifications are shown in Table 1, whereas Table 2 compiles the information about the applied instrument and correction schemes. The SSA of the mixture containing AS and AD, was controlled by the online measured SSA measured by the CAPS PM_{ssa}.

Three mass flow controllers (MFC), one at each generator's head and a third after the mixing chamber, supplied particle-free compressed air to the sample to both reach the desired humidity and particle number concentration and to make-up the flow required by the instruments. The particle number concentration was measured by a condensation particle counter (CPC).

Table 1. Type of generated aerosol, targeted SSA (630 nm), and targeted max. aerosol extinction values

Aerosol type	Estimated /Expected SSA	Run 1 200 Mm^{-1}	Run 2 150 Mm^{-1}	Run 3 100 Mm^{-1}	Run 4 50 Mm^{-1}	Run 5 25 Mm^{-1}
Aquadag (AD)	0.4	x	x	x	x	x
Black Carbon (BC)	0.4		x	x	x	x
Mixture (AS+AD)	0.6			x	x	x
Ammonium Sulphate (AS)	1.0		x	x	x	x

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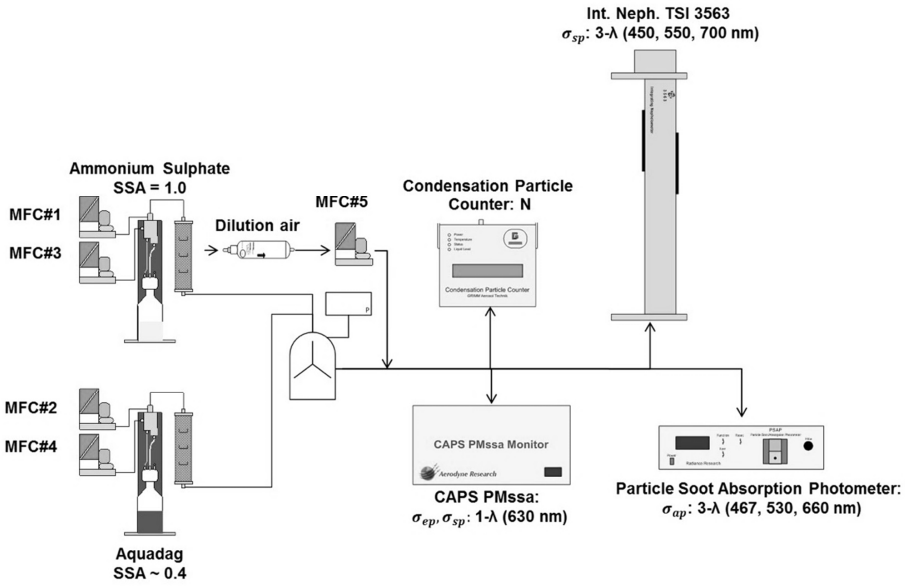

Figure 1. Instrumental set-up applied in the optical closure study

Table 2. List and specifications of optical instrumentation and applied correction algorithms

Instrument	Manufacturer	Property	λ (nm)	Aerosol	Correction Algorithm
CAPS PM_{ssa}	Aerodyne Research Inc.	σ_{sp}, σ_{ep}	630	AS, AD, BC, MIX	Mie Amigo (Aerodyne) for σ_{sp} truncation correction (Onasch et al., 2015)
CAPS PM_{ex}	Aerodyne Research Inc.	σ_{ep}	630	AS, AD, BC, MIX	No correction required
NEPH	TSI Inc.	σ_{sp}	450, 550, 700	AS	~~Müller et al. (2009), Anderson and Ogren (1998)~~
				AD, BC, MIX	Müller et al. (2009), Anderson et al. (1998) Massoli et al. (2009)
PSAP	Radiance Research Inc.	σ_{ap}	467, 530, 660	AS, AD, BC, MIX	~~Ogren (2010) and Virkkula (2010)~~Ogren (2010) and Virkkula (2010)

Field Code Changed

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The samples were produced at up to five nominal concentration levels, as shown in Table 1, defined by the aerosol extinction. This was achieved by holding the aerosol generation system constant (MFC#~~1~~3 and MFC#~~2~~4) and regulating the make-up air MFCs (MFC#~~3~~1, MFC#~~4~~2 and MFC#5). Extinction coefficient levels were varied from ~10 up to 200 Mm^{-1}. For each level, a sampling time of at least 5 minutes was sustained.

To ensure an isoaxial, isokinetic sampling by all instruments, special sampling tips made of stainless steel were designed such that the sample air extraction tips were each concentrically placed along the centre line of the sample tube of 1 inch inner diameter. The inlet nozzles diameters are dimensioned such that the flow velocities in the sample tube and inside extraction tip nozzles match. Distances between the extraction points for the different instruments were 20 cm.

~~All scattering instruments were~~The nephelometer was calibrated using CO_2 (high span gas) and particle-free air (low span gas), before starting the experiments. ~~This~~The calibration procedure includes also, as recommended by the manufacturers, the calibration of scattering channel of the CAPS PM_{ssa}, against the extinction channel of the instrument. For the filter-based absorption instruments, no calibration is necessary since they both operate with a blank filter in parallel as reference (see description in the subsections below).

The optical instruments were placed downstream of the generation system, measuring simultaneously, as shown, and will be described in more detail in the following subsections.

2.1.1 Integrating Nephelometer

In this optical closure study, an integrating nephelometer (NEPH) of the type TSI Model 3563 was used. The NEPH collects scattering measurements both in the forward and backscatter directions at three wavelengths 450, 550, and 700 nm (Heintzenberg et al., 2006).

The NEPH data was corrected for truncation angle effects using the approach proposed by Massoli et al. (2009) for strongly light-absorbing ~~aerosol and~~aerosols (equations 3 and 4 and Table 3). For predominantly light-scattering aerosols, the approaches proposed by Anderson et al. (1996) and ~~Müller et al. (2009)~~Müller et al. (2009) ~~for predominantly light-scattering aerosols.~~were used (equation 5 and Table 4).

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$$C = MAX\{1.0, v_0 + v_1 \exp(v_2 * (3.25 - å)) + C(n)\} \quad (3)$$

where å is the Ångstrom exponent (equation 7), $C'(n)$ is an optional correction for submicron distributions. $C(n)$ is equal to 0 for å ≥2.8, and to

$$C(n) = v_3(2.8 - å) * (\frac{1}{(n-1)} - \frac{1}{0.48}) \quad (4)$$

and v_0, v_1, v_2 *and* v_3 are given in Tab and n is the real part of the refractive indices.

Table 3. Coefficient values for v_0, v_1, v_2 *and* v_3 for equations 3 and 4 (Massoli et al., 2009).

	v_0	v_1	v_2	v_3
698 nm sub-µm	0.8627	0.1423	0.1816	0.0306
554 nm sub-µm	0.8511	0.1589	0.2153	0.0439
453 nm sub-µm	0.8863	0.1327	0.2758	0.0610
698 nm all	0.9869	0.0182	0.7980	
554 nm all	0.9948	0.0152	0.8951	
453 nm all	1.0072	0.0118	1.0036	

$$C = a + b * å \quad (5)$$

Table 4. Values for a and b for equation 5 for Anderson and Ogren (1998) and Müller et al. (2011).

	Blue (450 nm)	Green (550 nm)	Red (700 nm)

Ångstrom exponent		å(B/G)		Å(B/R)		Å(G/R)	
		a	b	a	b	a	b
Anderson et al. (1998)	No cut	1.365	-0.156	1.337	-0.138	1.297	-0.113
	Sub-µm	1.165	-0.046	1.152	-0.044	1.120	-0.035
Müller et al. (2011)	No cut	1.345	-0.146	1.319	-0.129	1.279	-0.105
	Sub-µm	1.148	-0.041	1-137	-0.040	1.109	-0.033

2.1.2 Particle-Soot Absorption Photometer

The PSAP is a filter-based three wavelength (467, 530, 660 nm) instrument, manufactured by Radiance Research, that provides continuous measurement of the light absorption coefficient. The instrument uses two spots on a quartz fibre filter; one receives the particle containing sample, and the second clean air. The instrument measures ~~then~~ the difference in the transmission of light between a loaded and a blank filter spot ~~(Bond et al., 1999)~~(Bond et al., 1999). ~~Absorption coefficient data were determined using the approach proposed by Ogren (2010).~~Two absorption coefficient data correction were used and evaluated: Ogren (2010) and Virkkula (2010). The best fitting corrections is the one shown in each result subsection.

In his approach, Ogren (2010) furthers the corrections from Bond et al. (1999), considering the filter area correction and wavelength adjustment, as shown in equation 6, for the complete absorption coefficient measurement.

$$\sigma_{ap} = 0.85 \left(\frac{Q_{PSAP}}{Q_{meas}}\right)\left(\frac{A_{meas}}{A_{PSAP}}\right)\frac{\sigma PSAP[\lambda]}{K_2} - \frac{K_1}{K_2}\sigma_{sp}[\lambda] \quad (6)$$

where σ_{ap} is the absorption coefficient of the desired wavelength, Q_{PSAP} is the flow recorded by the instrument, Q_{meas} is the measured flow, A_{meas} is the real area of the filter, A_{PSAP} is the manufacturer supplied area of the filter, σ_{PSAP} is the measured absorption coefficient at a certain wavelength (λ), K_1 and K_2 are constants given (0.02 ± 0.02 and 1.22 ± 0.20, respectively) and σ_{sp} is the scattering coefficient measured at the same wavelength as σ_{PSAP}.

Virkkula (2010) derives a new correction from a field campaign, including as a function factor the single scattering albedo, as shown in equation 7.

$$\sigma_{ap} = (k_0 + k_1(h_0 + h_1\omega_0)\ln(Tr))\sigma PSAP[\lambda] - s\sigma_{sp}[\lambda] \quad (7)$$

where σ_{ap} is the absorption coefficient of the desired wavelength, k_0, k_1, h_0, h_1 and s are constants given (Table 5), ω_0 is the single scattering albedo, Tr is the measured transmission, σ_{PSAP} is the value measured by the PSAP and σ_{sp} is the scattering coefficient measured at the same wavelength as σ_{PSAP}.

Table 5. Constant values given by Virkkula (2010) for equation 7

Constant	467 nm	530 nm	660 nm
k_0	0.377 ± 0.013	0.358 ± 0.011	0.352 ± 0.013
k_1	-0.640 ± 0.007	-0.640 ± 0.007	-0.674 ± 0.006
h_0	1.16 ± 0.05	1.17 ± 0.03	1.14 ± 0.11
h_1	-0.63 ± 0.09	-0.71 ± 0.05	-0.72 ± 0.16
s	0.015(0.009, 0.020)	0.017(0.012, 0.023)	0.022(0.016,0.028)

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2.1.3 The CAPS PM_{ex}

The CAPS PM_{ex} system, described in detail and assessed in several studies, such as Massoli et al. (2010), Petzold et al. (2013) and ~~Perim de Faria et al. (2017)~~Perim de Faria et al. (2017) measures light extinction by determining the change in signal phase shift caused by the introduction of particles into an optical cavity. The use of high reflectivity mirrors (reflectivity approx. 99.99%) in the optical cavity creates the long measurement path of approx. 2 km required to measure very low values of light extinction (LOD of 1-2 Mm^{-1} in 1 second sample period).

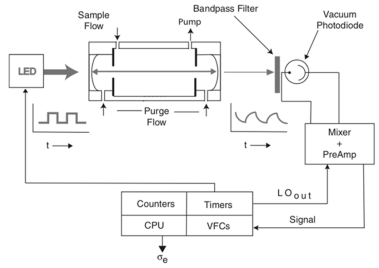

Figure 2. Overview of the main components and operation principle of the CAPS PM_{ex} instrument (Massoli et al., 2010)

2.1.4 The CAPS PM_{ssa}

The CAPS PM_{ssa} (Onasch et al., 2015), uses the same principle to measure light extinction as the CAPS PM_{ex}, but it also contains, located at the centre of the measurement cell, a 10 cm diameter integrating sphere capable of measuring light scattering on the same aerosol sample, as shown in ~~Figure 2.~~Figure 3. The integrating sphere acts as an integrating nephelometer, which measures the scattering of light by particles at all angles, only excluding the near 0 and near 180° angles since ~~at these directions~~ the opening of the extinction chamber is located in these directions, allowing the sample and light beam to pass through. The sphere shows 98-99% Lambertian reflectance efficiency due to its high reflectivity coating (Avian D from Avian Technologies). The usage of an integrating sphere increases the collection of scattered light at the photomultiplier compared to a traditional cosine corrected detector arrangement.

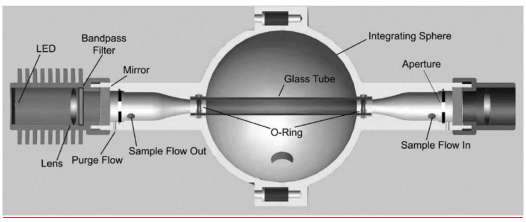

Figure 3. CAPS PMssa components and set-up (Onasch et al., 2015).

The scattering channel is calibrated against the extinction channel using either small particles (<250 nm) that have SSA=1.0~~,~~ or CO_2 (as done in this ~~case ammonium sulphate,~~study, shown in Figure 4 – slope, offset and R^2 calculated using the 1-second resolution data; the scatter plot shows the average and standard deviation) and set equal to the extinction measurement. Thus, the monitor should be thought of as providing separate extinction and SSA values with the scattering channel a derived measurement. This calibration procedure also allows the user to prove monitor linearity over a wide

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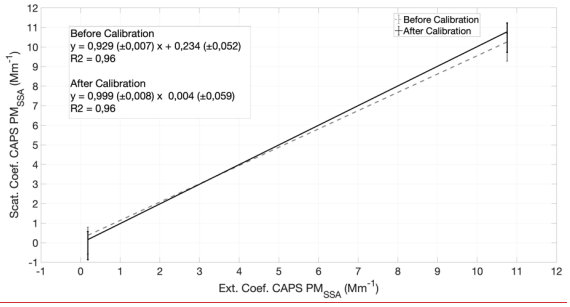
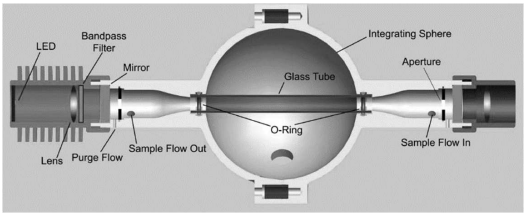
range of optical extinctions without the limitation of using individual gases with sometimes not particularly well-known Rayleigh scattering coefficients.



~~As in the CAPS PM$_{ex}$, the sample flow~~

Figure 4. Scatterplot of the correlation of the extinction and scattering channel of the CAPS PM$_{ssa}$ before and after the calibration using CO^2.

The sample flow in the instrument is set to 0.85 lpm and is controlled by a critical orifice. The measurement sample enters the chamber in one end and exits through an opening located in the other end flowing through a glass tube inside the integrating sphere (~~Figure 2). The mirrors are kept particle-free by a continuously flowing purge flow (25 cm^3 min^{-1}).~~Figure 3). The mirrors are kept particle-free by a continuously flowing purge flow (25 cm^3 min^{-1}). Petzold at al. (2013) showed that this purge flow shortens the measurement path and dilutes the sample and requires a correction factor. As done for the CAPS PM$_{ex}$, a new correction factor was developed, by using monodisperse polystyrene spheres (PSL) of know size, for the CAPS PM$_{ssa}$. Due to the cell geometry, the new correction factor was slightly larger than the one found for the extinction monitor, 1.37 and 1.27, respectively (Onasch et al., 2015). The noise of the instrument, trunction angle and instrument uncertainty have also been studied by Onasch et al. (2015). The values found were all below 1 Mm^{-1} for the noise levels (1σ, 1s) for all wavelengths. For the case of this particular instrument (630 nm), the truncation correction was determined below 4% for typical ambient conditions. The uncertainty was estimated at ±0.03 for SSA equal to 1 (PSL and ammonium sulphate) and decreases to ±0.01 as the SSA goes down.



~~**Figure 2. CAPS PMssa components and set-up (Onasch et al., 2015).**~~

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The baseline determination system is identical to the one used in the CAPS PM$_{ex}$, in which filtered and thus particle-free sample air fills the measurement chamber and is used to quantify contributions of gas molecules to the instrument response by Rayleigh scattering and potential absorption of light, and to determine interferences of system components. Both the CAPS PM$_{ex}$ and CAPS PM$_{ssa}$ used in this study operate at a wavelength of 630 nm and thus show minimal interference from absorption by ambient gaseous species like NO_2 and H_2O.

2.2 Data Treatment

All multi-wavelength instruments were adjusted to match the other instruments' wavelengths for the intercomparison by using the Ångström exponent approach; see Equation (~~3~~8) and (~~4~~9),

$$å = -\left. log\frac{\sigma_x}{\sigma_y} \right/ log\frac{x}{y} \quad (~~3~~8)$$

$$\sigma_w = \sigma_y \times (^{w}/_{y})^{-å} \quad (~~4~~9)$$

where å is the Ångström exponent, σ is the optical property measured (extinction, scattering or absorption coefficient), x and y are the operating wavelengths of the instrument, and w refers to the wavelength, to which the property should be adjusted. For a better understanding of the wavelength adjustment, the complete description is given in Figure 3 from Petzold et al. (2013).

All instruments provide 1 second resolution data. Data was collected over 5 minutes for each experimental point to remove any effect of differences in response times and fluctuations in the aerosol generation system. The data was averaged for each extinction/scattering/absorption level, and the standard deviation was calculated from the mean.

Standard linear regression analysis was performed for the mean values of each level. For the cases with the standard deviation of the intercept value being higher than the value itself, the regression model interception was forced to zero intercept, since the intercept value shows no significant difference to zero.

2.3 Measurement uncertainties

This paper does not address in any explicit way, nor was it designed to address, the question of the absolute uncertainties of the different measurement techniques. It was designed to address the question of how well they correlate. Thus, the results are given in correlation coefficients (slope and intercept) and their statistical uncertainties.

For this reason, this section compiles the reported absolute errors by the relevant instrument papers.

Table 6 Measurement uncertainties for the different instruments as reported by the relevant instrument papers.

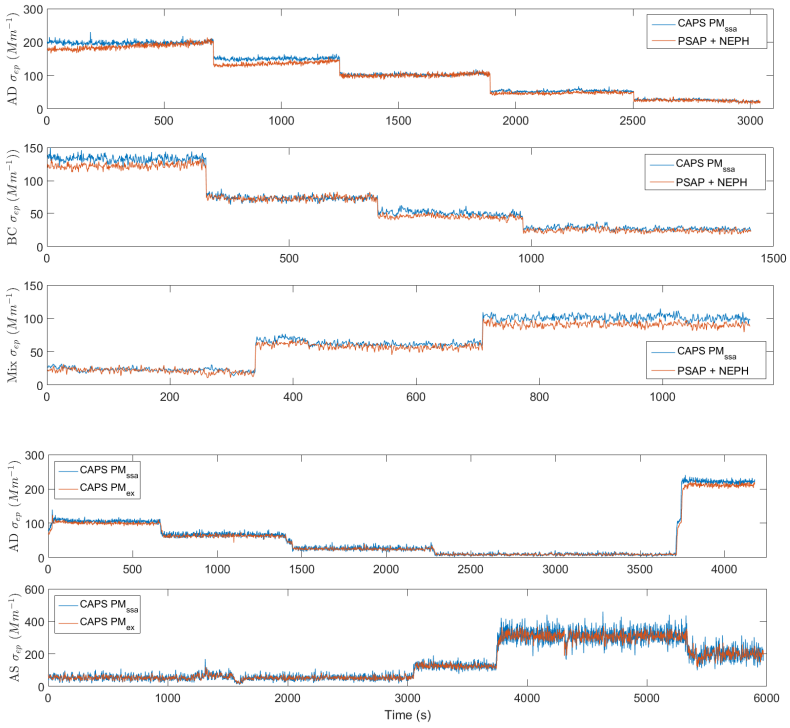
Instrument	sep	ssp	sap	SSA	Reference	Comments
CAPS PMssa	5%	8%	13% (SSA=0.5) and 5%(SSA=1.0)	3%	(Onasch et al., 2015)	Estimates for polydispers aerosol. Absorption uncertainty is dependent upon the SSA value
NEPH		<10%			(Anderson et al., 1996)& (Massoli et al., 2010)	for sub micrometer particles
CLAP			8%		(Ogren et al., 2017)	
PSAP			8%		(Muller et al., 2014)	
NEPH+PSAP	7%				(Petzold et al., 2013)	(3-sigam) obtained for the test aerosol inversion of NEPH+PSAP data

3 Results and Discussion

In this section, we present the results and relevant discussion of findings for the optical closure study. All the measurements presented here were corrected to the CAPS PM_{ssa} operational wavelength of 630 nm.

3.1 Extinction Coefficient

The extinction coefficient measured by the CAPS PM_{ssa} was analysed in comparison with proven technologies. On the direct measurement of σ_{ep}, we compared the two CAPS systems for AS and AD (Petzold et al., 2013). The direct measurement of σ_{ep} from the CAPS PM_{ssa} was also compared with the indirect measurement given by the sum of the absorption coefficient measured by the PSAP with the scattering coefficient measured by the NEPH for BC, AD, and MIX (as defined in Table 1~~).~~) – shown as PSAP+NEPH. For AS with the measured SSA value of 1.0, extinction coefficients provided by the CAPS extinction channels and scattering coefficients provided by the CAPS scattering channel and the NEPH instrument are used for the evaluation of the light scattering measurements in the next subsection. The time series for the extinction channels are shown in Figure 5 and the averages and standard deviations for each test point are shown in Table A1 in the supplemental information. The higher variability observed in the last plot of the figure is due to particle load fluctuations from generation system when operating at very high loads.



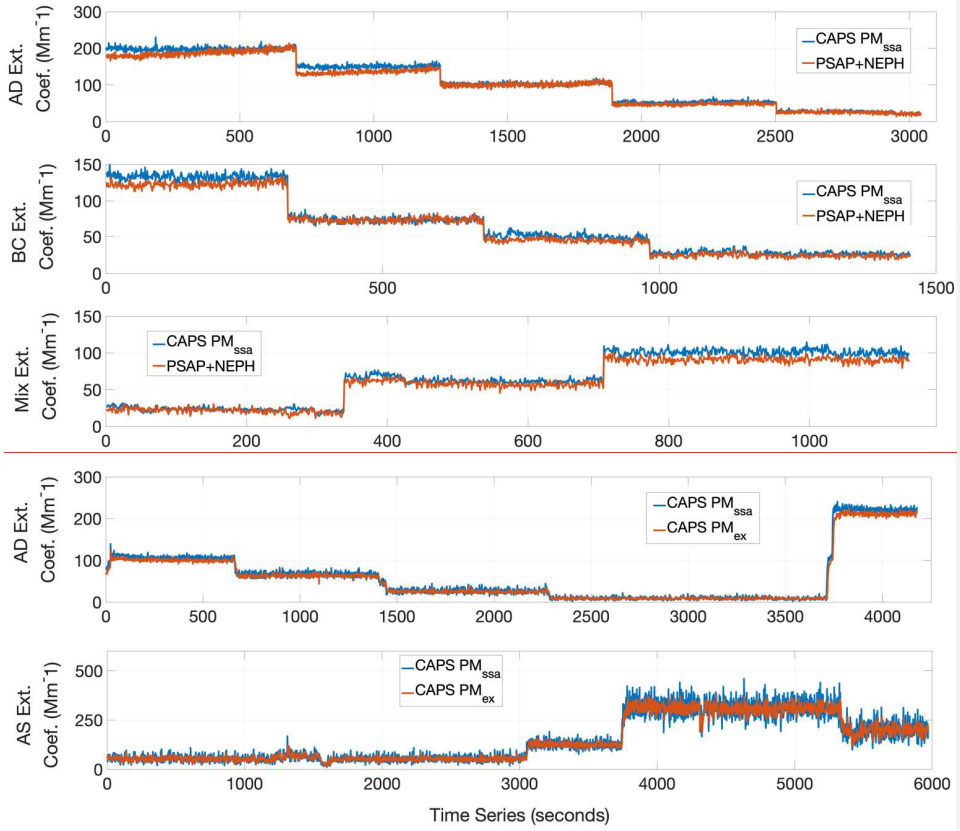


Figure 5. ~~**Time series of the measurements by the extinction channel.**~~

~~**Figure 4.**~~ **Time series of the parallel measured extinction coefficients by the different instruments. Instruments used : CAPS PM_{ex}, CAPS PM_{SSA} and the sum of absorption- and scattering coefficients measured by PSAP and NEPH as noted in the legend for the individual subplots. The test aerosolsused are noted in the caption of the y-axis of the individual subplots.**

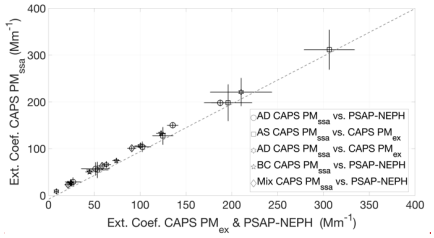
Figure 6 shows the scatter plot of the measured extinction coefficient for the two CAPS systems for AD and AS and the comparison with the sum of the NEPH and PSAP for AD and BC. The best results for the AD and BC were found when applying the Massoli et al. (2009) correction with the assumption, that no particle size cut has been used for the inlet system (no-cut approach) to the NEPH data, and ~~Virkkula (2010)~~Virkkula (2010) for strongly light-absorbing aerosols AD and BC to the PSAP data. For the mixture, the applied corrections were ~~Anderson et al. (1996) for the NEPH data and Ogren (2010) for the PSAP data.~~Anderson et al. (1998) for the NEPH data and Ogren (2010) for the PSAP data. The extinction channels from the two CAPS and the sum of the NEPH and PSAP (PSAP-NEPH) signals show a good agreement for all aerosol types, with linear regression slopes (m) between 0.94 and 1.02 and correlation coefficients above 0.99 (all regression analysis data for the averaged values of each level is presented in Table 7 together with their standard deviation). For the linear regression analysis of the full data set including all types of aerosols, the slope found was 0.96

(R^2=0.99) for the comparison of the CAPS PM_{ssa} extinction data with the sum of NEPH and PSAP data, and 0.97 (R^2=1.00) for the comparison of the CAPS PM_{ssa} and CAPS PM_{ex} extinction data. The slopes of the regression analysis and their standard deviation are shown in ~~Figure 5~~Figure 7 as a function of the sampled aerosol single-scattering albedo. As it can be seen there is no systematic difference in the slope with increase or decrease of the aerosol SSA.

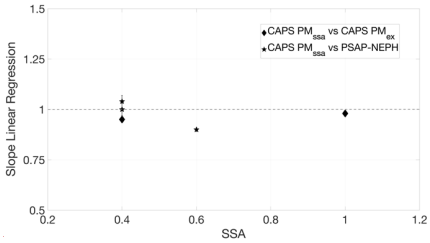
Table 7 Linear regression parameters including the slope (M), standard deviation of the ~~mean,~~slope (Std m), intercept~~,~~ (B), standard deviation of intercept~~,~~ (std b), and linear regression coefficient (R^2) for the comparison of the CAPS PM_{ssa} extinction channel with proven technologies

Aerosol	Reference Instrument	Estimated SSA	M	Std m	B	Std b	R^2
AD	PSAP~~-~~+NEPH	0.4	0.94	0.01	0.00	< 0.01	1.00
BC	PSAP~~-~~+NEPH	0.4	1.00	0.01	0.00	< 0.01	1.00
MIX	PSAP~~-~~+NEPH	0.6	1.02	0.00	0.00	< 0.01	1.00
ALL	PSAP~~-~~+NEPH	NA	0.~~96~~95	0.01	0.00	< 0.01	0.99
AD	CAPS PM_{ex}	0.4	0.95	0.00	0.00	< 0.01	1.00
AS	CAPS PM_{ex}	1.0	1.00	0.00	0.00	< 0.01	1.00
ALL	CAPS PM_{ex}	NA	0.97	0.00	0.00	< 0.01	1.00

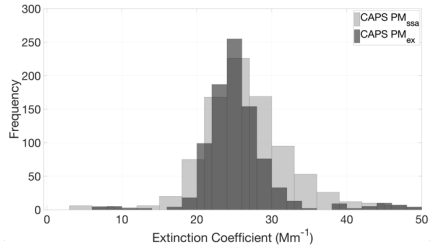
It is worth noting that for the particular instruments used in our study, the standard deviation for the extinction data of the CAPS PM_{ssa} is larger than for the extinction data provided by the CAPS PM_{ex} (horizontal error bars). This finding is shown in the histogram of the extinction channel from one measurement level (in this case the used dataset refers to the 25 Mm^{-1} target-level for AD aerosol) for both equipment (~~Figure 6).~~Figure 8). Thus, the precision of this particular CAPS PM_{ssa} is lower than the precision of the CAPS PM_{ex}. Regarding the precision of the CAPS PM_{ssa} in comparison with proven technologies, the standard deviation found in this study for both cases are comparable. The precision in the CAPS PM_{ex} and PSAP~~-~~+NEPH extinction measurements found in this study are very similar to the one found by Petzold et al. (2013)~~,~~ in which an excellent correlation (slope of 0.99) was found for the laboratory comparison between the same instruments using highly absorbing aerosol, exclusive scattering aerosol and mixtures of both.



~~Figure 4. Comparison result of the extinction channel of the CAPS PM_{ssa} with the CAPS PM_{ex} and the PSAP-NEPH for for the different aerosol types.~~



~~Figure 5. Slope values of the linear regressions of measured particle extinction as a function of nominal aerosol SSA for the different instrument intercomparison.~~



~~Figure 6 Frequency of extinction coefficient measurement for the CAPS PM_{ssa} and PM_{ex} systems at the nominal 25 Mm^{-1} (level 5) test point for AD.~~

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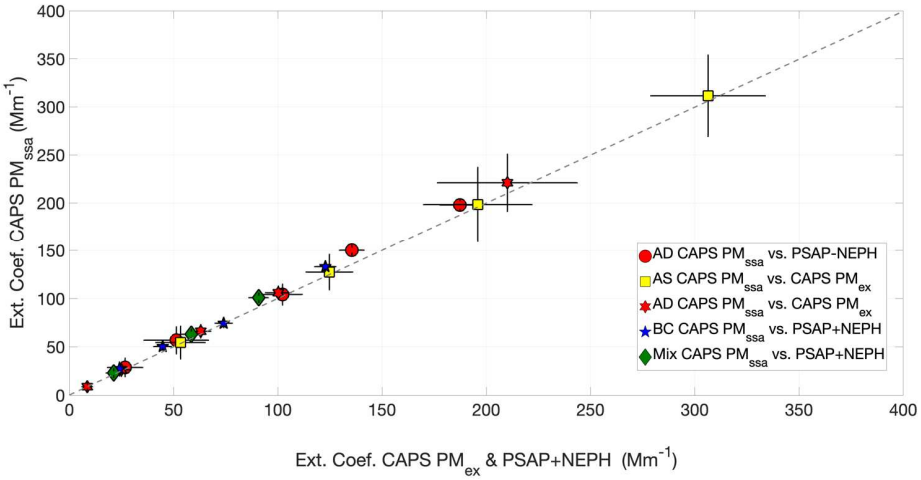

Figure 6. Comparison result of the extinction channel of the CAPS PM_{ssa} with the CAPS PM_{ex} and the combination PSAP+NEPH for for the different aerosol types (dashed line is the identity line (i.e., 1:1 line)).

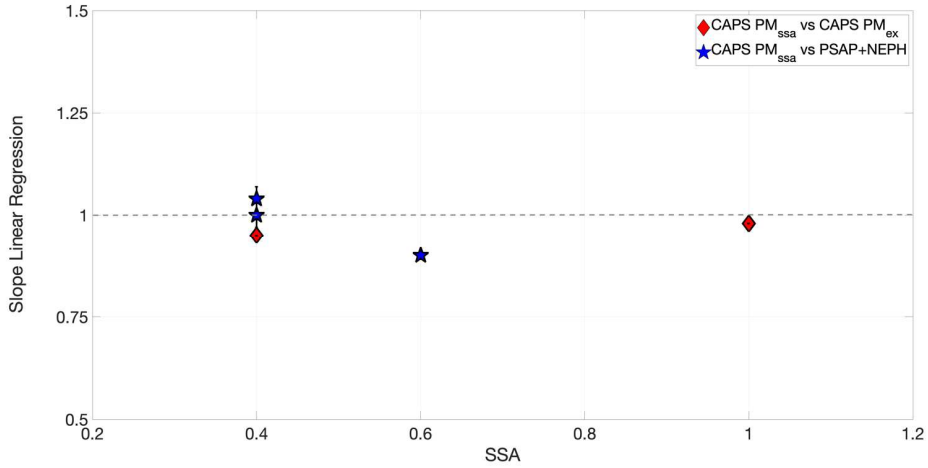

Figure 7. Slope values of the linear regressions of measured particle extinction as a function of nominal aerosol SSA for the different instrument intercomparison.

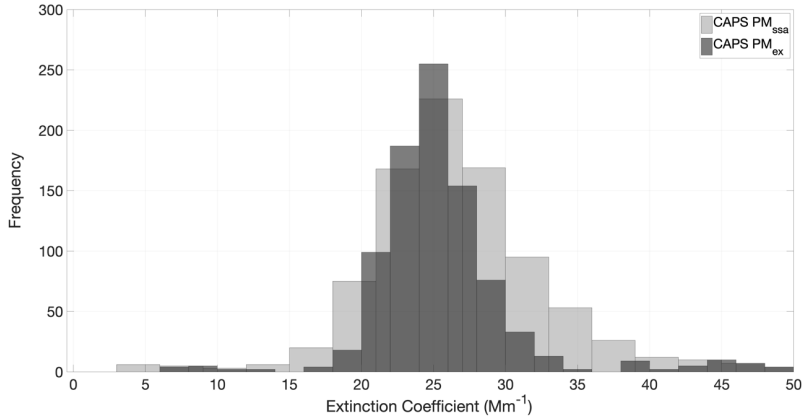

Figure 8. Frequency of extinction coefficient measurement for the CAPS PM_{ssa} and PM_{ex} systems at the nominal 25 Mm^{-1} (level 5) test point for AD.

3.2 Scattering Coefficient

The scattering channel of the CAPS PM_{ssa} was evaluated in comparison to the NEPH measurements for AD, BC, AS, and MIX (Table 1). The time series of scattering coefficient data for the various aerosol runs is shown in Figure 9. Supplemental Table A2 shows the average and 1-σ standard deviation obtained for the targeted scattering coefficient levels. ~~There is no~~Within the error bars of the two instruments we could not observe a systematic ~~error found neither~~deviation of both either in the average ~~nor~~or in the standard deviation of the measured values. The precision of both instruments for the measurement of scattering coefficient is very similar.

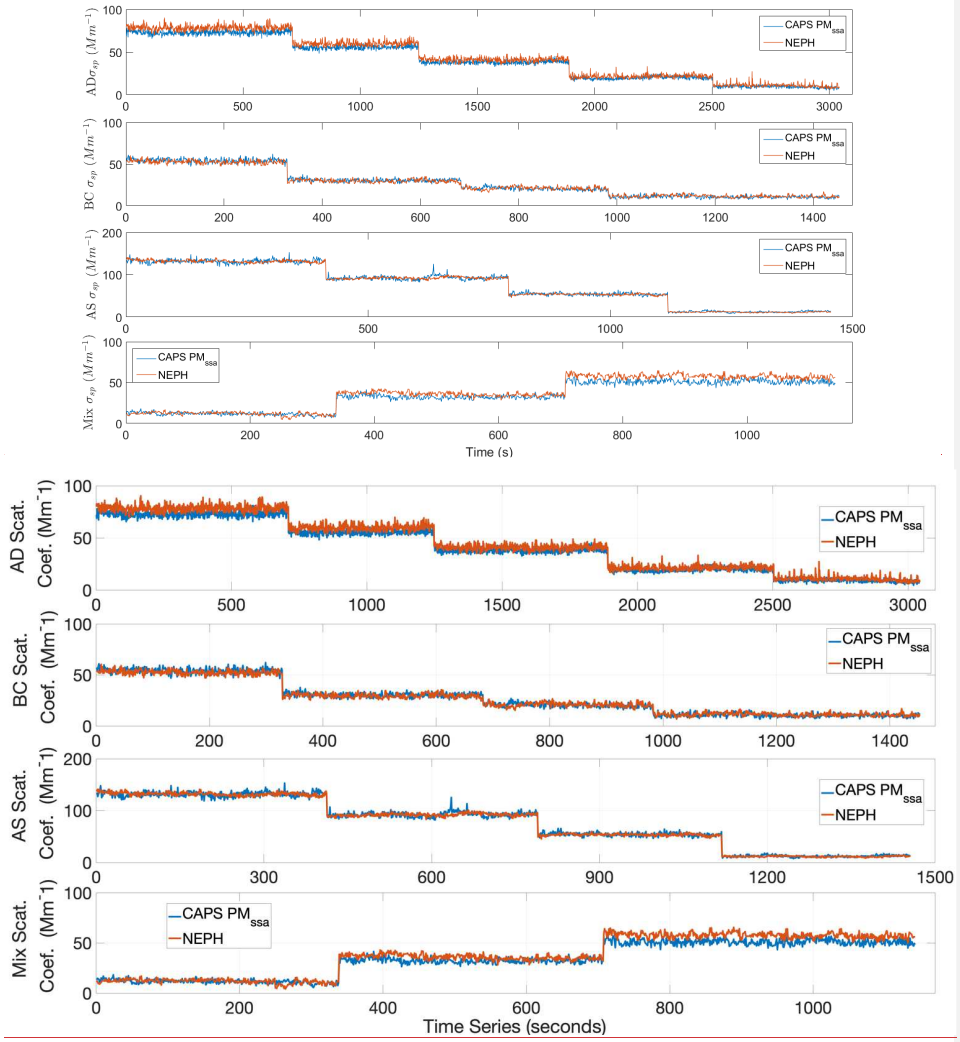

Figure 9. Time series of the ~~measurements by the~~ scattering ~~channel.~~coefficients parallel measured by the different instruments: CAPS PM_{SSA} and NEPH for the different aerosol types (BC (top-) AS(middle-) and Mix (bottom-figure).

~~Figure 8~~Figure 10 shows the scatter plot of the 1-second average and standard deviation of the CAPS PM_{ssa} against NEPH. As it can be seen from ~~Figure 8~~Figure 10 and the data compiled in ~~Table 4~~Table 8, the agreement with the NEPH measurements is excellent, with less than 8% difference in the slope, offset smaller than 2.00 Mm^{-1} and correlation coefficient of 1.00 for all aerosol types. The slope value and standard deviation as a function of SSA is shown in ~~Figure 9~~ Figure **11**. For the AD, BC and Mix cases, the NEPH data was corrected with the Massoli et al. (2009) approach. For the AS

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case both the ~~Anderson et al. (1996) and Müller et al. (2009)~~Anderson et al. (1998) and Müller et al. (2011) were applied and the results given were practically the same, less than 2% in the slope and less than 1.00 Mm^{-1} difference in the offset. For the overall measurement linear regression model, including all types of aerosols, the slope found was 1.01 (R^2=1.00) for the comparison of the CAPS PM_{ssa} with the NEPH.

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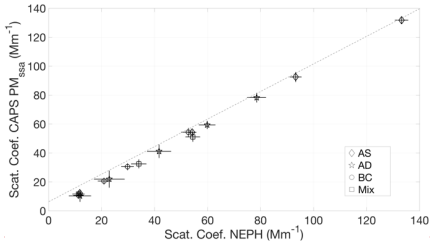
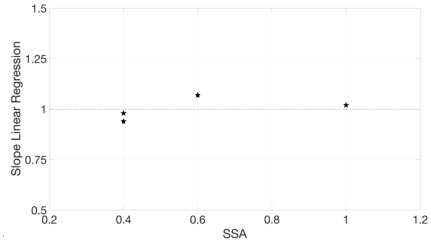
Table 8. Linear regression parameters including the slope (M), standard deviation of the ~~mean,~~slope (Std m), intercept~~,~~ (B), standard deviation of intercept~~,~~ (std b), and linear regression coefficient (R^2) for the comparison of the CAPS PM_{ssa} scattering channel with NEPH

Aerosol	Reference Instrument	SSA	m	Std m	b	Std b	R^2
AS	NEPH	1.00	1.02	0.00	-0.72	0.14	1.00
AD	NEPH	0.40	0.98	0.00	1.48	0.18	1.00
BC	NEPH	0.40	0.94	0.01	1.22	0.28	1.00
MIX	NEPH	0.60	1.07	0.01	-0.55	0.50	1.00
ALL	NEPH	NA	1.01	0.01	0.00	0.00	1.00



~~**Figure 8. Comparison result of the scattering channel of the CAPS PM_{ssa} with the measurements from the NEPH for the different aerosol types.**~~



~~**Figure 9. Slope values of the linear regression as a function of expected aerosol SSA for CAPS PM_{ssa} and NEPH; uncertainty of the slopes is below the resolution of the symbols; see Table 4.**~~

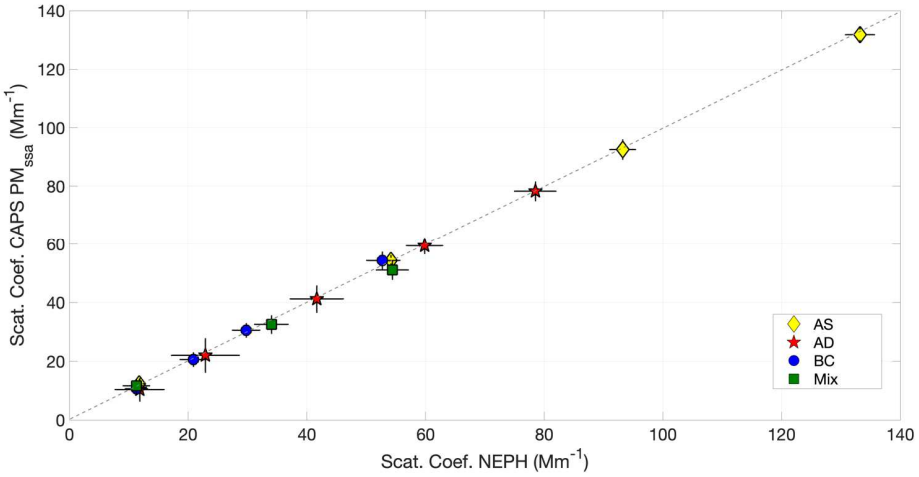


Figure 10. Comparison result of the scattering channel of the CAPS PM$_{ssa}$ with the measurements from the NEPH for the different aerosol types (dashed line is the identity line (i.e., 1:1 line)**).**

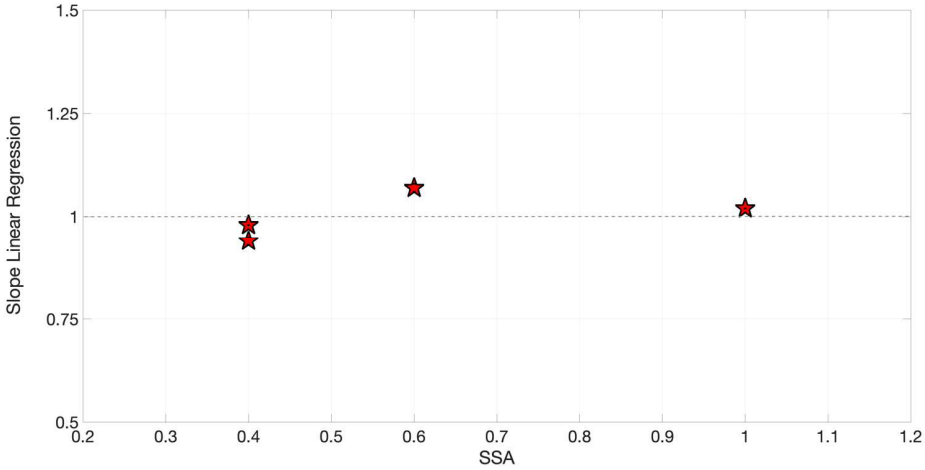


Figure 11. Slope values of the linear regression as a function of expected aerosol SSA for CAPS PM$_{ssa}$ and NEPH; uncertainty of the slopes is below the resolution of the symbols; see Table 8.

3.3 Absorption Coefficient

In spite of the fact that the CAPS PM$_{ssa}$ is not capable of directly measuring the absorption coefficient, the values can be derived as the difference of the extinction and the scattering coefficients; see Equation (1). From the difference of the two CAPS PM$_{ssa}$ channels the calculated absorption coefficients were compared to the direct measurement by the PSAP. In this

analysis, when operating with a mixture of AS and AD, the PSAP data were treated using the correction from ~~Ogren (2010).~~Ogren (2010). The time series for the measurement of the different aerosols are shown in Figure 12 whereas Supplemental Table A3 shows the average and 1-σ standard deviation obtained for the targeted absorption coefficient levels.

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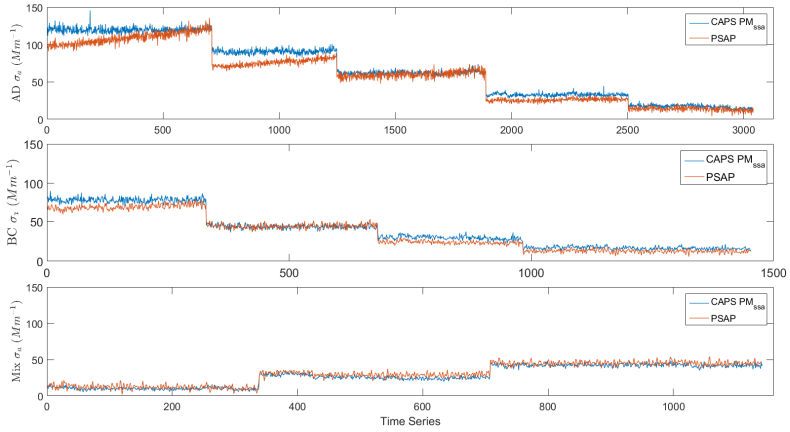

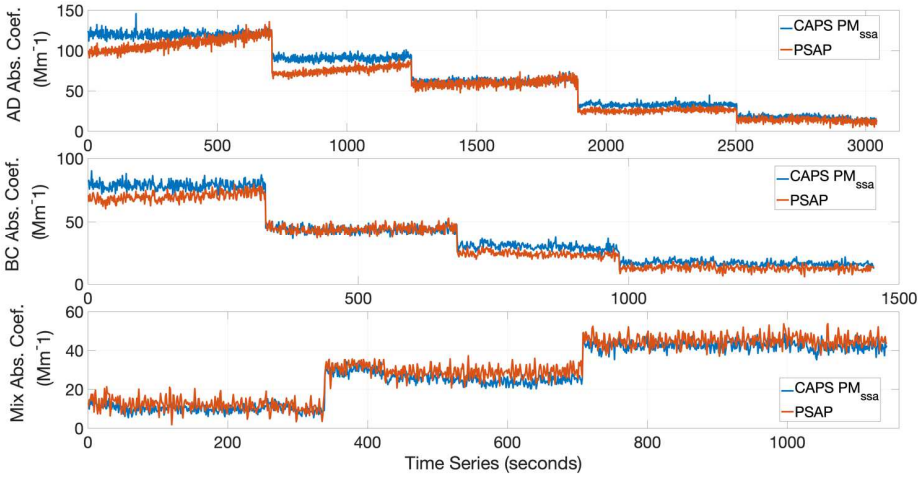

Figure 12. Time series (in seconds) of the parallel measurements ~~by~~of the absorption ~~channel.~~coefficient for the different test aerosols (TOP (Aquadag AD, Middle BC, Butttom (Mix) by the PSAP and the CAPS PM_{SSA} (as a result of the substraction of the scattering coefficient from the extinction coefficient).

The scatter plot for the average measured values from both methods for all levels is shown in Figure 13, whereas the results of the linear regression analysis are compiled in Table 9. The agreement between the methods is good, with deviations below 11% in the slope, and offsets less than 2.0 Mm^{-1}. The correlation coefficient is above 0.98 for all cases. For the full data set of CAPS PM_{ssa} and PSAP absorption coefficient data including all types of aerosols, the slope is 0.91 with a correlation coefficient of R^2=0.98. Figure 13 demonstrates that for higher absorption coefficients, the two methods deviate more strongly than for lower absorption coefficients. This is mainly caused by the correction algorithm applied to the PSAP data (also seen on Figure 12); filter loading corrections are significantly larger for higher absorption coefficient levels than for lower absorption coefficient levels. If the three data points for higher absorption coefficient data (σ_{ap} > 70 Mm^{-1}) are removed from the regression analysis, the slope value increases to 1.00 (R^2=0.97), although with an offset of -3.64. The increase in the absorption coefficient observed in Figure 12 for the higher levels of AD and BC, is related to the transmission decay of the filter in the PSAP and the correction algorithm chosen for this study. This finding proves that, although the CAPS PM_{ssa} cannot directly measure aerosol light absorption, it provides a rather reliable measurement of the absorption coefficient of the sampled aerosol, at least for the small particle sizes and intermediate SSA values sampled in this study. The accuracy of absorption measurements by the two channels of the CAPS PM_{ssa} may be significantly reduced for weakly absorbing but large-sized and irregularly shaped mineral dust particles.

Table 9. Linear regression parameters including the slope (M), standard deviation of the ~~mean,~~slope (Std m), intercept, (B), standard deviation of intercept, (std b), and linear regression coefficient (R^2) for the comparison of the CAPS PM_{ssa} and the PSAP instruments.

Aerosol	Reference Instrument	m	Std m	b	Std b	R^2
AD	PSAP	0.89	0.01	0.00	0.00	1.00
BC	PSAP	0.90	0.00	0.00	0.00	0.99
MIX	PSAP	1.02	0.04	2.02	1.16	0.99
ALL	PSAP	0.91	0.02	0.00	0.00	0.98
ALL (σ_{ap} < 70 Mm^{-1})	PSAP	1.00	0.07	-3.64	2.33	0.97

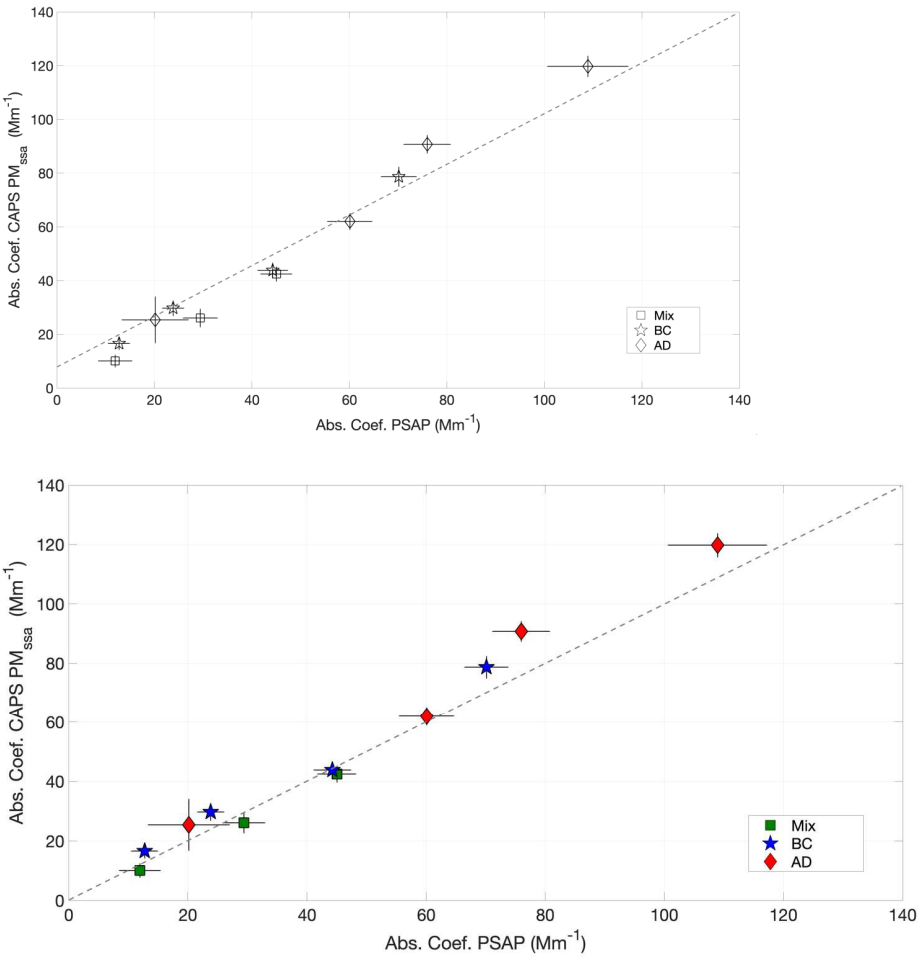

Figure 13. Comparison result of the absorption indirect measurement by the CAPS PM_{ssa} with the measurements from the PSAP for AD, BC and Mixture. (dashed line is the identity line (i.e., 1:1 line)).

3.4 Single Scattering Albedo Measurement

The ultimate property targeted by the CAPS PM_{ssa} is the aerosol single-scattering albedo. Figure 14 shows the average and standard deviation of the SSA measured by the CAPS PM_{ssa} and the applied proven technologies for each aerosol type containing a light-absorbing fraction, at the different extinction coefficient levels. The values for each level are also compiled in Supplemental Table A4.

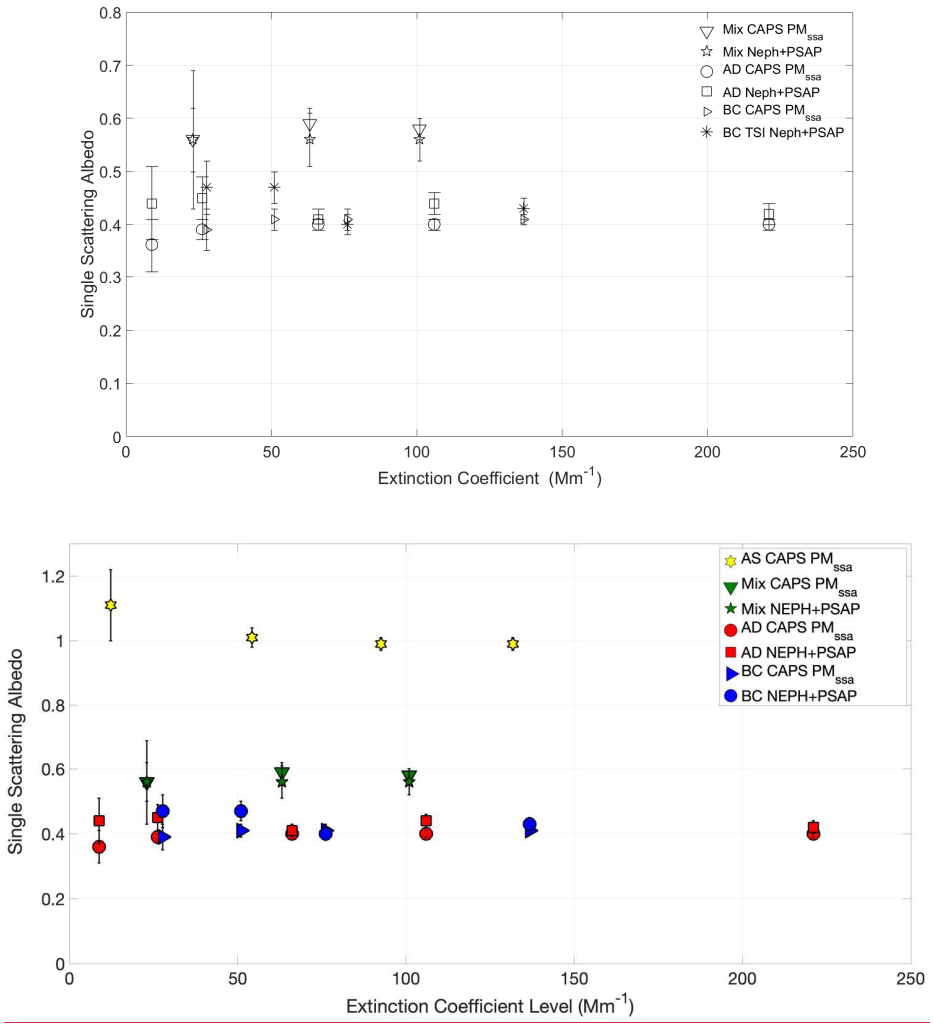

Figure 14. Average and standard deviation of the measured Single Scattering Albedo as a function of extinction coefficient level for the different aerosols and technologies.

For the absorbing aerosols, we found maximum deviations between the different SSA values of 0.09, or 18%, with the deviations being randomly distributed around zero. For a single aerosol type, the SSA provided by the CAPS PM_{ssa} shows less scatter around the average value compared to the values derived from PSAP and NEPH data. The measurements by the CAPS PM_{ssa} are more robust in terms of stability in comparison with the values measured by the PSAP+NEPH combination, with an average of the standard deviation for the different aerosol types of 0.01 for the CAPS PM_{ssa} and 0.02

for the PSAP-NEPH combination. It is worth noting that even though there are differences found in the measurements, all measured SSA values fall within the range of values expected for each aerosol type. (as measured and detailed in section 3.2 - Table 8).

Analysing the error propagation for the measured parameters (extinction and scattering coefficients), the increase of the uncertainty at the lower extinction coefficient levels is also visible for both CAPS PM_{ssa} and proven technologies; see Table 6 for details. From the experimental set-up, it was observed that the particle generation system was lightly unstable when operating at lower extinction/scattering levels, resulting in higher variations of the absolute values, which could explain such higher error propagation. This supports the previous findings that the CAPS PM_{ssa} accuracy is very good and comparable to the proven technologies.

Table 6. Absolute uncertainty of the SSA measurement for given aerosol types and applied instrument combinations

Aerosol	Instrument	Run 1	Run 2	Run 3	Run 4	Run 5
AS	CAPS PM_{ssa}	0.04	0.07	0.14	0.19	NA
AD	CAPS PM_{ssa}	0.05	0.05	0.14	0.34	0.46
	PSAP-NEPH	0.06	0.06	0.14	0.38	0.45
BC	CAPS PM_{ssa}	0.05	0.07	0.11	0.17	NA
	PSAP-NEPH	0.05	0.07	0.11	0.16	NA
Mix	CAPS PM_{ssa}	0.22	0.11	0.07	NA	NA
	PSAP-NEPH	0.25	0.11	0.06	NA	NA

4 Summary and Outlook

An optical closure study has been performed using different types of aerosols (pure scattering, strongly absorbing, and mixture) to evaluate the performance and accuracy of the recently launched Cavity Attenuated Phase-Shift Single Scattering Albedo Monitor.

The results from the instrument intercomparison with proven technologies (CAPS PM_{ex}, NEPH, and PSAP) show a very good agreement for all aerosol types, with accuracy of 96% and 99% for the extinction coefficient and scattering coefficient channels, respectively, for all aerosol types. The small deviation of 4% observed in the extinction channel between the CAPS PM_{ssa} and PSAP-NEPH combination originates from the applied correction algorithm to the PSAP data, since it is a logarithmic function of the filter transmission leading to deviations in the dataset. For the evaluation of the performance for each aerosol individually, the extinction channel shows accuracy between 94% and 98%; and the scattering channel, between 94% and 98%. These values are very similar to those found by Petzold et al. (2013) for the CAPS PM_{ex}.

Regarding the application of the CAPS PM_{ssa} for the measurement of the absorption coefficient and single-scattering albedo, the instrument has shown good performance on both sides. The accuracy of the absorption coefficient measurement by the CAPS PM_{ssa} in comparison with the PSAP was 91%, as obtained for the linear regression analysis for all investigated aerosol types and aerosol loadings. The large difference observed here comes from the correction scheme applied to the PSAP data at high loadings, as stated earlier. It is possible to observe that the higher deviations occur at high absorption coefficient, also where the transmission of the filter has a steeper decrease. Once the linear regression analysis excludes the points where the average absorption coefficient was higher than 70 Mm^{-1}, the slope approaches 100%

agreement between the two technologies. For the measurement of SSA, the CAPS PM_{ssa} showed a very good stability for all measured σ_{ep} levels, better than the PSAP-NEPH combination. The measured values are within what is expected for the different types of aerosols (0.4 for strongly absorbing aerosols and 1.0 for purely scattering aerosols).

The results reported from our study demonstrate that the CAPS PM_{ssa} is a very robust and reliable instrument for the direct measurement of the scattering and extinction coefficient, as well as for the indirect measurement of the absorption coefficient and single scattering albedo.

5 Author Contributions

JP, UB, and AP designed the study and prepared the manuscript, with contributions from all co-authors. AF and TO provided technical details of the instrumentation and contributed to the interpretation of the study results.

6 Competing Interests

The authors declare that they have no conflict of interest.

7 Acknowledgements

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9 Appendix A

SUPPLEMENTAL INFORMATION

Table A1. Extinction coefficient mean and 1-σ standard deviation of the mean measured by the CAPS PM_{ssa} extinction channel and proven technologies

			Run 1	Run 2	Run 3	Run 4	Run 5
AS	CAPS PM_{ssa}	Av	54.62	127.43	311.65	198.31	NA
		Std	0.29	0.66	1.04	1.50	NA
	CAPS PM_{ex}	Av	53.39	124.78	306.40	195.94	NA
		Std	0.21	0.41	0.68	1.01	NA
AD	CAPS PM_{ssa}	Av	221.04	105.98	66.16	26.25	8.84
		Std	1.34	0.23	0.22	0.20	0.08
	CAPS PM_{ex}	Av	210.15	100.22	63.08	24.93	8.66
		Std	1.53	0.22	0.16	0.14	0.05
AD	CAPS PM_{ssa}	Av	198.00	150.09	104.15	56.88	28.85
		Std	0.20	0.17	0.39	0.53	0.37
	PSAP-+NEPH	Av	187.37	135.55	102.30	51.34	26.78
		Std	0.33	0.23	0.36	0.61	0.34
BC	CAPS PM_{ssa}	Av	136.77	76.16	50.99	27.73	NA
		Std	0.26	0.20	0.22	0.13	NA
	PSAP-+NEPH	Av	134.98	81.59	48.51	26.28	NA
		Std	0.22	0.18	0.16	0.29	NA
Mix	CAPS PM_{ssa}	Av	23.05	63.14	100.94	NA	NA
		Std	0.17	0.25	0.20	NA	NA
	PSAP-+NEPH	Av	21.28	58.47	90.83	NA	NA
		Std	0.19	0.23	0.18	NA	NA

Table A2. Scattering coefficient mean and 1-σ standard deviation of the mean measured by the CAPS PM_{ssa} and NEPH

			Run 1	Run 2	Run 3	Run 4	Run 5
AS	CAPS PM_{ssa}	Av	131.79	92.57	54.29	12.31	NA
		Std	0.11	0.16	0.08	0.06	NA
	NEPH	Av	133.22	93.22	54.18	11.77	NA
		Std	0.11	0.10	0.08	0.04	NA
AD	CAPS PM_{ssa}	Av	78.29	59.42	41.18	21.98	10.32
		Std	0.11	0.10	0.16	0.22	0.15
	NEPH	Av	78.50	59.86	41.70	22.93	11.87
		Std	0.12	0.12	0.17	0.22	0.17
BC	CAPS PM_{ssa}	Av	54.33	30.54	20.58	10.66	NA
		Std	0.14	0.11	0.11	0.08	NA
	NEPH	Av	52.71	29.81	20.91	11.31	NA
		Std	0.14	0.11	0.12	0.08	NA
Mix	CAPS PM_{ssa}	Av	11.66	32.52	51.09	NA	NA
		Std	0.11	0.14	0.14	NA	NA
	NEPH	Av	11.32	34.05	54.43	NA	NA
		Std	0.11	0.14	0.12	NA	NA

Table A3. Absorption coefficient mean and 1-σ standard deviation of the mean measured by the CAPS PM_{ssa} (extinction minus scattering) and PSAP

			Run 1	Run 2	Run 3	Run 4	Run 5
BC	CAPS PM_{ssa}	Av	78.69	43.78	29.73	16.57	NA
		Std	0.18	0.13	0.14	0.09	NA
	PSAP	Av	70.13	44.27	23.85	12.74	NA
		Std	0.19	0.16	0.12	0.09	NA
AD	CAPS PM_{ssa}	Av	119.75	90.76	62.02	25.40	18.53
		Std	0.14	0.13	0.24	0.32	0.23
	PSAP	Av	108.92	75.97	60.09	20.16	14.92
		Std	0.31	0.19	0.23	0.40	0.20
Mix	CAPS PM_{ssa}	Av	10.09	26.09	42.44	NA	NA
		Std	0.10	0.16	0.11	NA	NA
	PSAP	Av	11.95	29.42	45.03	NA	NA
		Std	0.18	0.17	0.14	NA	NA

Table A4. Single Scattering Albedo average value and standard deviation for CAPS PM_{ssa} and proven technologies

	Scat/Ext		Run 1	Run 2	Run 3	Run 4	Run 5
AS	CAPS PM_{ssa}	Av	0.99	0.99	1.01	1.09	NA
		Std	0.02	0.02	0.03	0.11	NA
AD	CAPS PM_{ssa}	Av	0.40	0.40	0.40	0.39	0.36
		Std	0.01	0.01	0.01	0.02	0.05
	PSAP-+NEPH	Av	0.42	0.44	0.41	0.45	0.44
		Std	0.02	0.02	0.02	0.04	0.07
BC	CAPS PM_{ssa}	Av	0.40	0.40	0.40	0.38	NA
		Std	0.01	0.02	0.02	0.04	NA
	PSAP-+NEPH	Av	0.39	0.37	0.43	0.43	NA
		Std	0.02	0.02	0.03	0.05	NA
Mix	CAPS PM_{ssa}	Av	0.51	0.52	0.51	NA	NA
		Std	0.06	0.03	0.02	NA	NA
	PSAP-+NEPH	Av	0.53	0.58	0.60	NA	NA
		Std	0.13	0.05	0.04	NA	NA