



Characterization of filter photometer artefacts in soot and dust measurements – laboratory and ambient experiments using a traceably-calibrated aerosol absorption reference

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Abstract. A novel reference absorption instrument, based on photothermal interferometry - the PTAAM-2 λ , and scattering measurements are used to characterize filter photometer artefacts in measurements of absorption coefficients of soot and dust-dominated aerosol samples within laboratory and ambient campaigns.

The Aethalometer AE33 and the Continuous Light Absorption Photometer (CLAP) were characterized during a laboratory campaign where different soot-like and mineral dust samples were measured. Furthermore, ambient measurements during a campaign in Granada, Spain, were used to characterize the AE33 and MAAP (Multi Angle Absorption Photometer), a pseudo-reference absorption instrument.

The laboratory campaign showed significant wavelength dependence of the multiple-scattering parameter C . The C of AE33 at 450/808 was 4.08/3.95 and 6.25/5.27 for propane soot and diesel soot, respectively. For the CLAP the C was 5.10/4.26 and 6.79/5.80 for propane and diesel soot, respectively. For mineral dust, C at 450 nm ranged between 2.74 and 3.03 for the AE33 and between 2.50 and 2.80 for the CLAP. The ambient measurements showed an overall C of 4.72 at 450 nm and of 3.90 at 808 nm for the AE33. The results for both the AE33 and the CLAP show a dependence with the particle size, with fine particles having the highest C values, and as the aerosols become larger C levels off. Both the laboratory and the ambient measurements of the AE33 showed overlapping results.

The cross-sensitivity to scattering was smaller for the CLAP than for the AE33. The values of the cross-sensitivity parameter m_s at 450/808 nm were 3.0%/1.5% for the AE33 and 2.4/0.9% for the CLAP at 450/808 nm.

The intercomparison of the MAAP with the PTAAM-2 λ during the ambient campaign in Granada showed that the MAAP-derived absorption coefficients feature a 47% overestimation at 637 nm and a cross-sensitivity to scattering of 2.4%.



1 Introduction

20 Light-absorbing aerosols (LAA) are a major cause of atmospheric warming due to their direct effect on Earth's radiative budget and their semi-direct radiative effects in the Earth's atmosphere (Szopa et al., 2021). However, the short lifetime of aerosols and the aging processes that they undergo in the atmosphere as well as their heterogeneous emissions introduce a challenge for accurately determining their optical properties, spatial distribution and climate effects (Bond et al., 2013; Zanatta et al., 2016; Saleh et al., 2018; Samset et al., 2018). Indeed, the effect of aerosols is one of the largest uncertainties in climate models
25 (Szopa et al., 2021).

Of all LAA, black carbon (BC) is the most important aerosol warming agent due to its large mass absorption cross-section (Myhre et al., 2013; Szopa et al., 2021). The influence on the climate of absorbing organic LAA, either primary - emitted into the atmosphere, or secondary, produced in the atmosphere by physical and chemical processes, is also a large unknown (e.g. Laskin et al., 2015; Rovira et al., 2025). Moreover, mineral dust, which also absorbs sunlight and contributes most to
30 aerosol mass in the atmosphere, also features quite large uncertainties in its climate effects (e.g., Kok et al., 2017, 2021a, b). The highest uncertainties are associated with the variable distribution of particle size in the atmosphere and the mineralogical composition, both of which are important parameters in the influence of dust on the net radiative forcing (Di Biagio et al., 2020; Adebisi and Kok, 2020; Huang et al., 2020; Kok et al., 2021a; Li et al., 2021). Mineral dust absorption is mainly driven by the composition of iron oxides (Sokolik and Toon, 1999; Lafon et al., 2006; Balkanski et al., 2007; Caponi et al., 2017;
35 Di Biagio et al., 2019).

The most widely used instrumentation to determine aerosol absorption coefficients, especially in ambient measurements, are filter absorption photometers (FP) (Moosmüller et al., 2009): aethalometers (Hansen et al., 1984; Drinovec et al., 2015), particle soot absorption photometers (PSAP; Bond et al., 1999), continuous light absorbing photometers (CLAP; Ogren et al., 2017) and the multi angle absorption photometers (MAAP; Petzold and Schönlinner, 2004). Their working principle, albeit with
40 some design variations, is based on the accumulation of the aerosol sample on the filter, through which the attenuation of light in comparison with a blank filter is measured to derive absorption coefficients and the equivalent black carbon concentrations (eBC; Petzold et al., 2013). FP feature two main artefacts that influence the absorption determination: the filter loading effect (FLE) for which many correction schemes have been developed (Bond et al., 1999; Weingartner et al., 2003; Collaud Coen et al., 2010; Drinovec et al., 2015, 2017), and the cross-sensitivity to scattering of aerosol particles within the filter tape, which
45 can also be corrected for, but requires additional scattering measurements (Arnott et al., 2005; Ogren et al., 2017; Yús-Díez et al., 2021).

Filter photometers were designed for black carbon measurements (Gundel et al., 1984), but the measurement was interpreted at the same time as one of the absorption coefficient (Hansen et al., 1982, 1984). Eventually, filter photometers were used for dust measurements (Fialho et al., 2005). FP sensitivity depends on the depth at which particles are trapped in the filter matrix, as
50 taken into account by the MAAP using a simple radiative transfer model (Petzold and Schönlinner, 2004), or observed in other filter photometers as the dependence of the multiple-scattering parameter C on the size of the particles (Drinovec et al., 2022).

In order to use the filter photometers in dust measurements, the measurement needs to be characterized with a representative sample and its artefacts quantified.

Ultimately, reference absorption measurements are required to measure the absorption coefficient and correct for the FP artefacts. This is especially important since the new European Air Quality Directive (PE-CONS 88/24) requires the measurement of BC concentrations by optical absorption methods (European Commission, 2024). There are two main methods that provide direct measurements of the absorption coefficient and avoid artefacts introduced by FPs: photoacoustic spectrometry (PAS Arnott et al., 2003) and photothermal interferometry (PTI; Moosmüller and Arnott, 1996; Visser et al., 2020; Drinovec et al., 2022). Both are based on the heating of the aerosol sample by focusing light from an intensive source on it as it is drawn through a measurement chamber. The methods differ in the probe method: PAS uses an acoustic resonator and a microphone for detection, whereas PTI uses an interferometer and photodiodes. PAS can exhibit biases when the aerosol sample contains semi-volatile organic coatings or water, since the detected acoustic signal can be reduced by the latent heat of the material during evaporation of these substances upon heating (Arnott et al., 2003; Moosmüller et al., 2009). PTI and PAS have been found to agree when measuring soot particles coated with specific secondary organic matter (Kalbermatter et al., 2022). A novel traceably calibrated PTI instrument, the PTAAM-2 λ (Haze Instruments, Slovenia) has been shown to provide accurate and precise aerosol absorption measurements for different aerosol particle compositions (Drinovec et al., 2022).

Furthermore, another established manner to obtain the absorption coefficient, especially for laboratory experiments, is the extinction-minus-scattering method, which is usually performed by Cavity Phase Shift Extinction analyzers (CAPS; Massoli et al., 2010; Modini et al., 2021). Extinction-minus-scattering-obtained absorption coefficients are fairly robust and mainly feature low uncertainties, however for high single scattering albedo values, where the extinction is dominated by scattering, this methodology introduces very high uncertainties (Moosmüller et al., 2009; Singh et al., 2014).

Filter photometers therefore require the use of a co-located reference absorption measurement for their calibration. Some studies have used either the extinction-minus-scattering method (Bond et al., 1999; Di Biagio et al., 2017) or another filter photometer, such as the TAP in Laing et al. (2020) or more sophisticated filter photometers that measure the backscattering of light from the filter such as the MAAP (Yus-Díez et al., 2021) or the PP_UniMI (Bernardoni et al., 2021; Ferrero et al., 2021). Indeed, the MAAP is used as the reference instrument in the Aerosol, Clouds and Trace Gases Research InfraStructure (ACTRIS; ECAC-CAIS, 2022) guidelines to harmonize absorption coefficients from the dual-spot multi-wavelength aethalometer, the AE33 (Drinovec et al., 2015), across the ACTRIS network. However sophisticated these methods are, none truly measure the absorption coefficient, and all add an additional layer of uncertainty. Recently, Drinovec et al. (2022) used a traceably calibrated instrument based on PTI, the PTAAM-2 λ , to calibrate the AE33 and the CLAP and quantify their cross-sensitivity to scattering artefact for aerosol mixtures of fine soot and ammonium sulfate.

We use the novel PTAAM-2 λ and an integrating nephelometer Aurora 4000 to fully quantify the artefacts of widely deployed filter photometers: the AE33, the CLAP and the pseudo-reference MAAP. For this purpose, we have performed chamber experiments with a wide variety of aerosol types during a laboratory campaign, as well as ambient measurements in a polluted urban background atmosphere in Granada (Spain).



Table 1. Sample classification and number of measurements per sample type. The absorption Ångström exponent (AAE) was derived from the PTAAM-2 λ using 450 and 808 nm measurements, and the SSA at 450 and 808 nm as a combination of the PTAAM-2 λ and the nephelometer. Sediment mineral dust sample specific characteristics can be found in González-Romero et al. (2023, 2024a, b), while other samples are either commercially available or specifically generated for this study. The SSA and the AAE are characteristic of the generated aerosol mineral dust samples, produced from the sediment samples.

Group	Region/type	Samples	Latitude (°)	Longitude (°)	n	AAE	SSA _{450nm}	SSA _{808nm}
Mineral dust	Sahara (Morocco)	MOR Surf. random	29.83	-5.87	3	4.46	0.955	0.997
		MOR 31	29.82	-5.95	3	3.61	0.939	0.982
		MOR 40	29.70	-6.02	3	4.61	0.957	0.997
		MOR 108	29.93	-6.38	3	4.73	0.964	0.998
	Wadi Rum (Jordan)	JOR Surf. random	29.74	35.38	5	4.83	0.956	0.998
		JOR 50	32.49	38.03	3	4.77	0.965	0.997
		JOR 46	31.69	36.96	3	5.26	0.972	0.999
	Mojave (USA)	USA 21	35.54	-115.41	3	4.46	0.964	0.998
		USA 37	35.15	-116.06	3	3.30	0.972	0.997
		USA 49	35.14	-116.32	3	4.24	0.980	0.999
	Icelandic dust	ICE Surf. random	64.92	-16.78	4	1.94	0.971	0.992
		ICE 95	63.60	-18.35	4	2.31	0.949	0.988
		ICE 105	63.67	-19.93	3	2.16	0.944	0.987
		ICE 108	64.46	-20.86	3	2.47	0.970	0.990
	Quartz	Qua	-	-	3	-	1.000	1.000
Soot-like	Diesel soot	die	-	-	2	1.24	0.459	0.660
	Propane soot	pro	-	-	2	1.03	0.465	0.610

2 Methodology

This study showcases the analysis based on two distinct set-ups: a laboratory and an ambient campaign. Both campaigns had similar instrumental lay-outs, with the determination of particle absorption using filter photometers and a reference absorption measurement by the photo-thermal interferometer. Additional measurements of scattering and particle size distribution were performed at both campaigns. The experimental set-up at both campaigns can be found in Fig. 1.

2.1 Laboratory aerosol measurement

2.1.1 Samples: mineral dust, quartz, soot

Different samples of dust and soot-like aerosolized particles were measured in this study (cf. Table 1). In addition, pure quartz dust (Sigma-Aldrich 342890-100G) was used as a purely scattering dust reference sample. Finally, samples with high BC

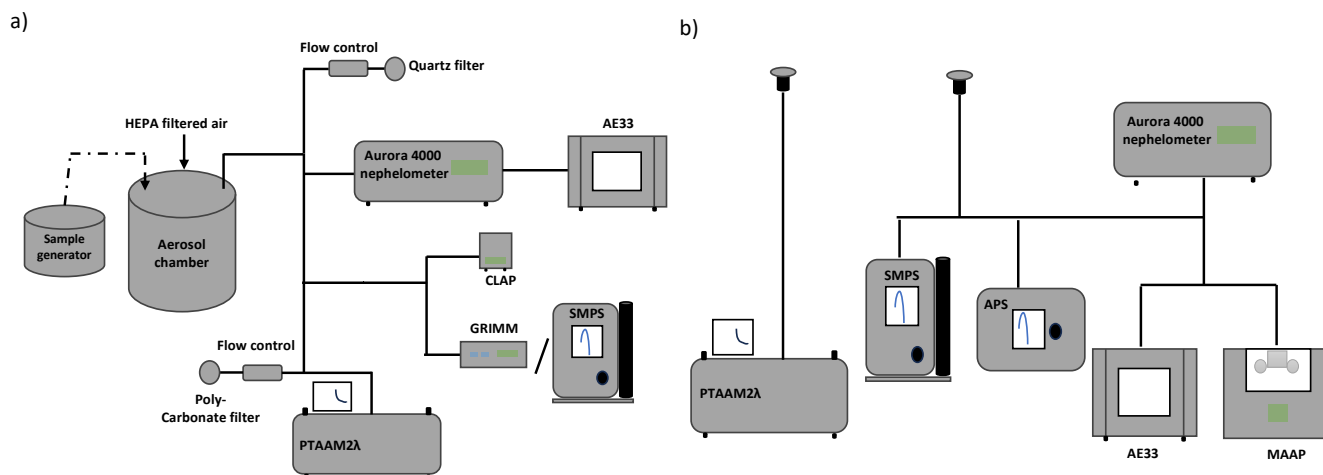


Figure 1. a) Laboratory set-up for measuring mineral dust airborne/combustion generated samples, and b) set-up at the UGR site for measuring ambient aerosol. The attenuation coefficients were measured by an AE33 (lab and ambient) and a CLAP (lab), reference absorption was measured with the PTAAM2 λ . Scattering coefficients were obtained by the Aurora 4000 nephelometer for both campaigns. Particle size distribution was obtained either by an OPS (GRIMM 11-D) for dust particles or by an SMPS for soot-like particles during the laboratory campaign, and by an SMPS and an APS during the ambient campaign. Offline quartz fiber and polycarbonate filters were sampled with different flows.

95 content: Euro-3 engine diesel soot and propane soot (Drinovec et al., 2022) were sampled. Fig. S1 shows the CLAP filter spots as a visual reference for the different samples.

Bulk mineral sediment samples were collected at different emission areas across mid- and high-latitude deserts which are sources of major dust emissions (Sahara, Middle-East, Mojave and Iceland). These samples were collected within the framework of the project FRontiers in dust minerAloGical coMposition and its Effects upoN climaTe (FRAGMENT; <https://cordis.europa.eu/project/rcn/214076/factsheet/en>). Mineral dust properties, such as size segregated composition and similar can be found in González-Flórez et al. (2023); Panta et al. (2023); González-Romero et al. (2023, 2024a, b). In this study, since the samples are collected with a relatively small spatial variability within a well-defined area of the broader dust emission area, we present the results for the overall emission area and not specifically for each of the individual samples.

105 Aerosol mineral dust samples were produced in the lab in a vortex generator similar to device described in (Moosmüller et al., 2012). Filtered compressed air was directed tangentially to produce a vortex. To remove the large particles the sample was collected from the center of the container. Mineral dust generated by several short bursts of compressed air was directed in a 120 l plastic barrel, which was then connected to the instruments and sampled until concentrations dropped to negligible levels. For each sample, between 2 and 4 experiments were performed with increasing concentrations.



2.1.2 Instrument set-up

110 Online reference absorption measurements were performed with a dual-wavelength photothermal aerosol absorption monitor
PTAAM-2 λ (Drinovec et al., 2022, ; Haze Instruments, Slovenia). PTAAM-2 λ is based on a folded Mach-Zender interferom-
eter which measures the difference in the optical path between the sample and reference interferometer arms. The resulting
phase difference is directly proportional to the absorption coefficient of the aerosol sample. For this study a newer version of the
instrument was used with pump lasers operating at 450 and 808 nm instead of 532 and 1064 nm used in Drinovec et al. (2022).
115 The calibration procedure was identical to the one described in Drinovec et al. (2022): the 450 nm channel was calibrated with
1 ppm NO₂, and for the 808 nm channel the calibration was transferred from 450 nm using polydisperse nigrosin particles and
an absorption ratio $b_{abs}(808)/b_{abs}(450) = 0.335$ calculated with a Mie model. For PTAAM-2 λ the sources of uncertainties at
450 nm are: NO₂ amount fraction, NO₂ absorption cross section at 450 nm, scattering and absorbing gases, and stability of
the instrument, resulting in the final absorption coefficient uncertainty 4.2% at 450 nm. At the longer wavelength, there are the
120 additional uncertainties of the Mie calculation due to the complex nigrosin refractive index and size distribution, resulting in
absorption coefficient uncertainty 6.2% at 808 nm. The uncertainty of the Ångström exponent is 10.4 % as it is independent
of the NO₂ amount fraction and the NO₂ absorption cross section at 450 nm. More details and references can be found in the
Table S1.

Measurements of attenuation were performed with two filter photometers, the Aethalometer AE33 (Magee Scientific/Aerosol,
125 USA/Slovenia) and the continuous light-absorbing photometer (CLAP, Haze Instruments, Slovenia). The AE33 measures at-
tenuation at seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) and compensates the filter loading effect through an
in-built algorithm (Drinovec et al., 2015). The CLAP measures the attenuation at 470, 529 and 653 nm, which were compen-
sated for the filter loading using a Ogren et al. (2017)-like algorithm scheme. Both instruments use glass-fibre filter, the M8060
filter tape in the AE33, and the Azumi 371M filter in the CLAP. The CLAP filter was changed for every set of experiments for
130 each sample. For comparison with the PTAAM-2 λ , the AE33 data was interpolated from 370 and 470 to 450 nm, and from 660
and 880 to 808 nm using the absorption Ångström exponent. The CLAP data was extrapolated to 808 nm using the absorption
Ångström exponent from CLAP measurements.

Aerosol particle size distribution measurements were performed using an optical particle spectrometer (GRIMM 11-D) for
mineral dust samples and with a scanning mobility parameter (TSI model 393L75) for soot-like particles (diesel, propane).
135 Scattering measurements were obtained with an Aurora 4000 polar integrating nephelometer (Ecotech, Australia) measuring at
450 nm, 525 nm and 635 nm. The instrument was set to measure total and back-scattering coefficients and angular truncation
errors were corrected applying Teri et al. (2022) correction schemes.

The single scattering albedo values were derived by using the total scattering coefficients from the Aurora 4000 integrating
nephelometer in combination with the PTAAM-2 λ absorption coefficient with the nephelometer data extrapolated to 808 nm
140 using the measured scattering Ångström exponent.



2.2 Ambient measurements

The ambient campaign was performed between 16 June and 24 August 2023 at the University of Granada (UGR) urban background station (37.18°N, 3.58°W, 680 m asl), which is part of the AGORA Observatory. It is located in the city of Granada in Southern Spain. The main local source of aerosol particles is road traffic (Titos et al., 2014, 2017), with sporadic presence of biomass burning aerosol, specially in autumn and winter seasons (Titos et al., 2017; Casquero-Vera et al., 2021). Air-mass stagnation episodes are relatively frequent and favor the accumulation of pollution near the surface (Lyamani et al., 2012; Patrón et al., 2017). The city is located 200 km away from the African continent, and is frequently influenced by long-range transport of Saharan dust (Lyamani et al., 2010; Valenzuela et al., 2012a, b).

Particle number size distributions at UGR were measured with a scanning mobility particle sizer (SMPS, TSI Mod. 3082) and an aerodynamic particle spectrometer (APS, TSI Mod. 3321). The APS aerodynamic diameters, in the range of ~ 0.5 to $20 \mu\text{m}$, were converted to mobility diameter following the procedure described by Khlystov and Pandis (2004). The aerosol particle density and shape factors used are based in those used for this station in previous studies (cf. Hess et al., 1998; Frank Wagner and Tegen, 2009; Sorribas et al., 2015). Total and back-scattering measurements were performed with the same Aurora 4000 nephelometer as during the laboratory measurements, to which the Teri et al. (2022) angular truncation correction was applied. The nephelometer was set up to measure at 7 different angles (0, 15, 30, 45, 60, 75 and 90°), of which only the total scattering coefficient is used in this study.

Online measurements of attenuation coefficients were performed with an AE33. As for the laboratory measurements, the filter tape used was the M8060 and FLE was compensated with the AE33 internal algorithm. Furthermore, absorption coefficients at 637 nm were obtained with a multiple-angle absorption photometer (MAAP; Petzold and Schönlinner, 2004), and the Müller et al. (2011) scheme was applied to correct for the difference between the reported and true instrument wavelength.

Reference absorption measurements were performed with a PTAAM-2 λ using the same configuration as the one used in the laboratory campaign. For this campaign, the instrument uncertainty is the same, a 4% at the blue channel (450 nm) and a 6% at the infra-red channel (808 nm).

As in the laboratory campaign, the AE33 measurements were interpolated/extrapolated to the PTAAM-2 λ wavelengths. Conversely, the PTAAM-2 λ absorption was interpolated using the AAE to the MAAP wavelength.

2.3 Filter photometer artefacts

We have applied the approach proposed by Yus-Díez et al. (2021) for the AE33, CLAP and MAAP. Yus-Díez et al. (2021) compensation scheme was an adaptation for the AE33 based of the Ogren et al. (2017) scheme for the CLAP, first developed by Bond et al. (1999).

Following these schemes, the FP measured attenuation coefficient is compensated for the scattering of light by particles in the filter matrix to derive the corrected attenuation coefficient:

$$b_{atn-cor} = b_{atn} - m_s \cdot b_{sp}, \quad (1)$$



where b_{atn} is the measured attenuation coefficient by the instrument, m_s is the scattering artefact describing the cross-sensitivity to scattering of the measurement, and b_{sp} is the scattering coefficient of the sampled aerosol.

175 The absorption coefficient derived from these attenuation measurements is compensated for the multiple-scattering of light in the filter matrix loaded with the sample using a single multiplicative multiple-scattering parameter C :

$$b_{abs} = \frac{b_{atn-cor}}{C} \quad (2)$$

180 Unlike in previous studies (e.g. Bernardoni et al., 2021; Yus-Díez et al., 2021), here we use as reference an instrument that does not suffer from the size and scattering artefacts – the PTAAM-2 λ (Drinovec et al., 2022). As a consequence, we were able to determine the correction parameters without any additional artefacts from the reference absorption measurements.

3 Results

Here we present the analysis of AE33, CLAP and MAAP artefacts from both the laboratory and the ambient campaigns.

As mentioned in section 2.1, the measurement of mineral dust samples during the laboratory experiment was performed as a two-stage process, first the chamber was filled with the airborne generated mineral dust sample, and then slowly emptied. 185 Figure 2a shows the decay of the AE33 and CLAP attenuation coefficients and the PTAAM-2 λ absorption coefficients as the chamber gets emptied. Due to the large initial concentrations, we maintain high values of the absorption/attenuation coefficients through-out the experiment – this is especially relevant for the 808 nm wavelength, since at this wavelength dust absorption is very low (Di Biagio et al., 2019). Nonetheless, the scatter of the data becomes quite large for absorption coefficients below 10 Mm^{-1} . Furthermore, Fig. 2b shows the typical temporal variability of the attenuation (AE33) and absorption (PTAAM- 190 2 λ) coefficients during two typical days of the ambient campaign in Granada during a Saharan dust event over the region. It features high AAE values (above 2), with prominent contributions from local sources at the urban site. This is shown by the increase in the absorption (and decrease of AAE) during peak local emission hours (06:00 and 20:00 UTC) and the otherwise low absorption values (5/1.5 Mm^{-1} at 450/808 nm) during midday due to the dilution within the atmospheric boundary layer.

3.1 Filter photometer cross-sensitivity to scattering

195 During the laboratory experiments, we analyzed the FP scattering cross-sensitivity artefact by measuring quartz as a reference purely scattering mineral dust sample. Therefore, any measured attenuation by the filter photometer is the result of the scattering artefact, which in Fig. 3a is obtained as the slope of the attenuation vs the scattering. The scattering artefact m_s at 450/808 nm for the AE33 was 3.0%/1.5% and 2.4%/0.9% for the CLAP (cf. Fig. 3a). Both the resuspended quartz and real-world mineral dust samples had similar particle size distributions (Fig. S2), so we have assumed that the magnitude of the multiple- 200 scattering artefact is the same for all samples. The scattering artefact values found herein for the AE33 were half the value and consistent with the decreasing trend found in Drinovec et al. (2022), where m_s was around 6% particles with a volume size mode of 122 nm, and within the range found in Drinovec et al. (2015) for the old filter type used in the AE33 (Pallflex

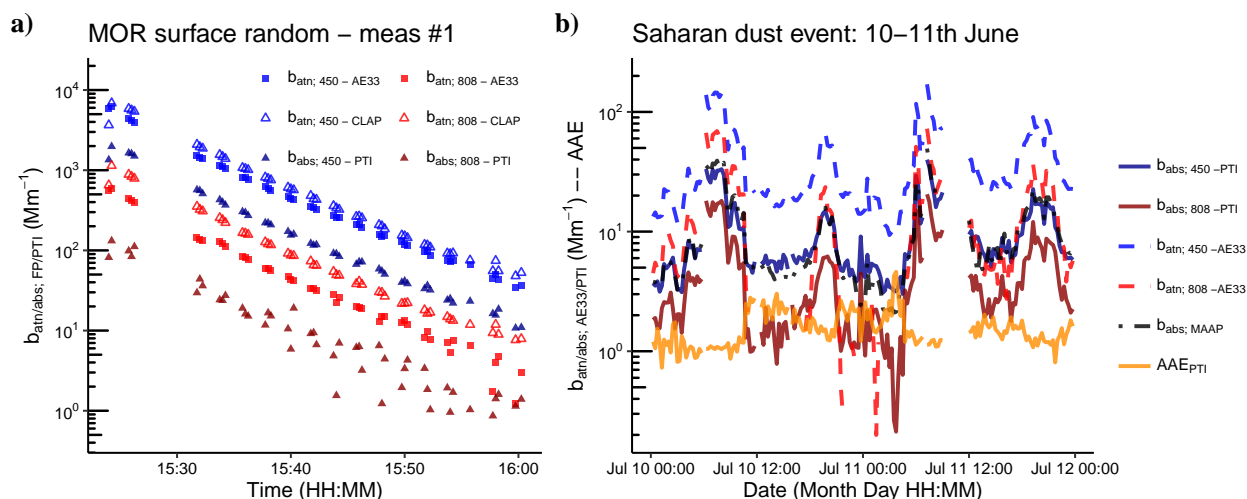


Figure 2. a) Example of the typical time evolution for a resuspended mineral dust sample, during the measurement of the *Morocco Surface Random sample* - the attenuation coefficient (b_{atn}) of the AE33 and CLAP, and the PTAAM-2 λ absorption coefficient, and b) the temporal evolution of the absorption from the PTI, the absorption Ångström Exponent, and the multiple-scattering parameter for 2 Saharan dust event days during the campaign in Granada (time in UTC). Note the logarithmic scale on the y-axis.

“Fiberfilm” T60A20), 1.2-3.4%. The average CLAP scattering artefacts at 450 nm (2.4%) and at 808 nm (0.9%), were close to the wavelength-independent value 1.64% determined by Ogren et al. (2017). The values are summarized in Table 2.

205 The ambient campaign showed a highly heterogeneous aerosol composition with a multi-mode particle size distribution (Fig. S2), thus preventing the scattering artefact to be assumed to be the same as for the laboratory quartz samples. Consequently, to correct for the cross-sensitivity to scattering the AE33 data we have followed the approach proposed by Yus-Díez et al. (2021), where the scattering artefact is obtained from the fit of the ratio of attenuation, measured by the AE33, to the reference absorption coefficient, measured by PTAAM-2 λ , as a function of single scattering albedo. Unlike for dust sample measurements, the
 210 presence of BC results in significant absorption in the infrared, resulting in a good fit not only at 450 nm but also at 808 nm. Figure 3(b) shows that the scattering artefact, obtained from the fit of the curve following eq. (9) in Yus-Díez et al. (2021), is 6.1% for 450 nm and 6.7% for 808 nm. These values were higher than those found in this study for the dust laboratory experiments, however similar to the ones found in Drinovec et al. (2022) for the 120 nm particles investigated therein. Compared with literature values, we have found considerably higher scattering artefact values for AE33 than at other measurement
 215 sites. Indeed, Yus-Díez et al. (2021) showed scattering artefacts of 1.6-4.9%. We attribute this difference to the use here of a direct reference instrument (the PTAAM-2 λ) with no artefacts instead of MAAP as done in Yus-Díez et al. (2021)(see Fig. S3 and section 3.3 below). Another reason for, smaller, discrepancies could be the different aerosol composition and particle size among different sites.

Although we have shown that the scattering artefact was higher during the ambient campaign than for the dust samples
 220 during the laboratory campaign, Fig. 4 shows that the relative contribution introduced by the scattering artefact to the total

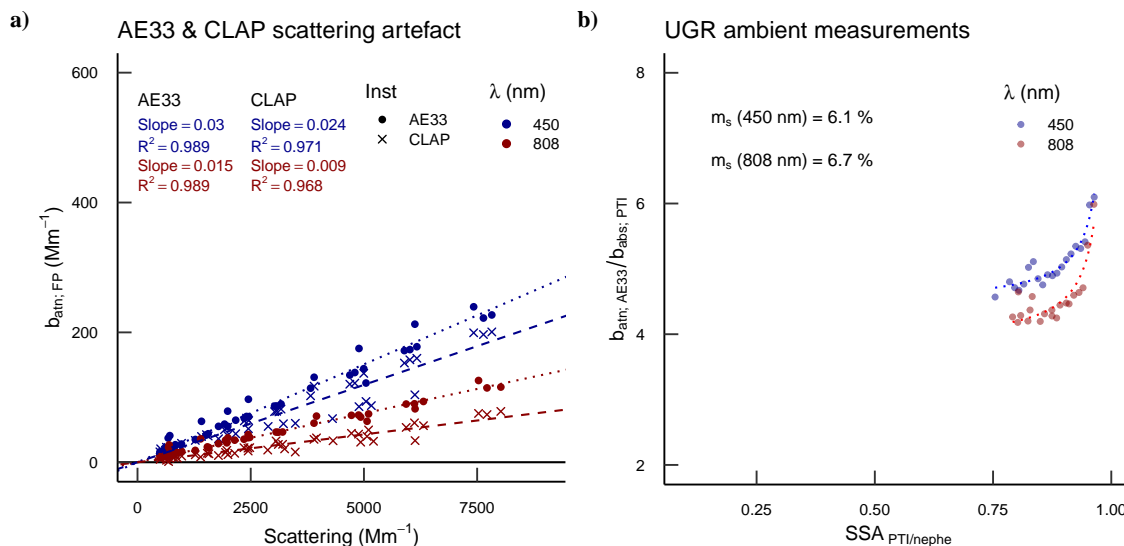


Figure 3. The filter photometer scattering artefact shown as a) the dependence of attenuation on the scattering coefficient in AE33 and CLAP during the laboratory campaign using resuspended non-absorbing Quartz samples; and b) during the the summer 2023 AGORA campaign in Granada as the fit of the ratio of the AE33 attenuation coefficient to PTAAM-2 λ absorption coefficient vs. SSA at 450 nm and 808 nm - parameters from fit as in Yus-Díez et al. (2021).

attenuation measured by filter photometers is higher for the pure dust samples in laboratory experiments, where SSA is higher (cf. Table 1). Furthermore, Fig. 4 shows that the relative contribution of the scattering artefact for pure dust samples increased with the wavelength, to values over 40% at the red (cf. Fig. S7). The increase with the wavelength is associated with the very low absorption by dust particles in the red and infrared wavelengths compared to scattering (Caponi et al., 2017). Fig. S6(a) shows that the relative contribution of scattering to attenuation remained constant through-out the experiment. The higher increase of the relative contribution of scattering to attenuation at 808 nm, although not as apparent, it was also a feature in Granada under low concentration scenarios (cf. Fig. S6(b)). However, during the ambient campaign, soot-like particles, which were the main absorbing particles, have a higher MAC, hence this effect was only observed for very low concentrations, when the absorption coefficient was below 1 Mm^{-1} (cf. Fig. S6(b)), which is more frequent for the 808 nm wavelength.

230 3.2 Multiple-scattering parameter for filter photometers

Table 3 shows the multiple-scattering parameter, C, after correcting the attenuation coefficients for the cross-sensitivity to scattering for each of the sample types. The values have been obtained for each of the sample groups as the slope of the orthogonal fit of the attenuation coefficients corrected for the cross-sensitivity to scattering $b_{atn-cor}$ (cf. eq. 1) vs the reference absorption coefficients from the PTAAM-2 λ (cf. Figs. S8-S24). It should be noted that due to the effect of interpolation/extrapolation from/to the FP or PTAAM-2 λ wavelengths the C values reported at the FP wavelengths vs the PTAAM-2 λ could vary. This



Table 2. Scattering artefact, m_s , for both the Aethalometer AE33 (laboratory and ambient campaign) and the CLAP (only laboratory campaign) obtained as the dependence of attenuation on the scattering coefficient in AE33 and CLAP during the laboratory campaign using resuspended non-absorbing quartz samples; and during the the summer 2023 AGORA campaign in Granada as the fit of the ratio of the AE33 attenuation coefficient to PTAAM-2 λ absorption coefficient vs. SSA at 450 nm and 808 nm - parameters from fit as in Yus-Díez et al. (2021). Figures with the fits for the AE33 and CLAP wavelengths can be found in Figs. S4-S5.

	λ (nm)	m_s (%)	
		Quartz dust	UGR ambient
AE33	370	4.2	6.7
	450	3.0	6.5
	470	2.8	7.2
	520	2.5	8.4
	590	2.2	8.1
	660	1.9	7.4
	808	1.5	6.4
	880	1.4	7.0
	950	1.3	6.6
CLAP	450	2.4	
	470	1.9	
	529	1.8	
	653	1.3	
	808	0.9	

effect is expected to be the highest for dust, since the AAE between 370 and 660 nm for dust is higher than between 660 and 950 nm.

The C of the dust samples at 450 nm was between 2.50 and 2.80 for the CLAP and between 2.74 and 3.13 for the AE33. The C for the AE33 is smaller than those provided by Di Biagio et al. (2017) for dust samples for aethalometer AE31. Table 3 shows that for all the mid-latitude deserts, the C decreases slightly between 370 and 660 nm, with an increase in the C value and its variability above 808 nm. Iceland dust, however, does not follow this pattern, and the C for 808, 880 and 950 nm, although slightly higher, had a lower variability (cf. Tables S2 and S3). This behavior is linked with the high contribution of the scattering artefact to the total absorption for all the dust samples, which is lower for Icelandic dust (Fig. S7), which could be linked to its higher absorption at these wavelengths (Baldo et al., 2023).

This high variability of the fit is shown by the low R^2 and the 95% confidence interval range for these samples, as can be seen in Tables S2 and S3. Therefore, the C values obtained for these higher wavelengths (808, 880 and 950 nm) cannot be relied on for the dust samples, since the high contribution of the scattering artefact to the total AE33 signal and the low correlation

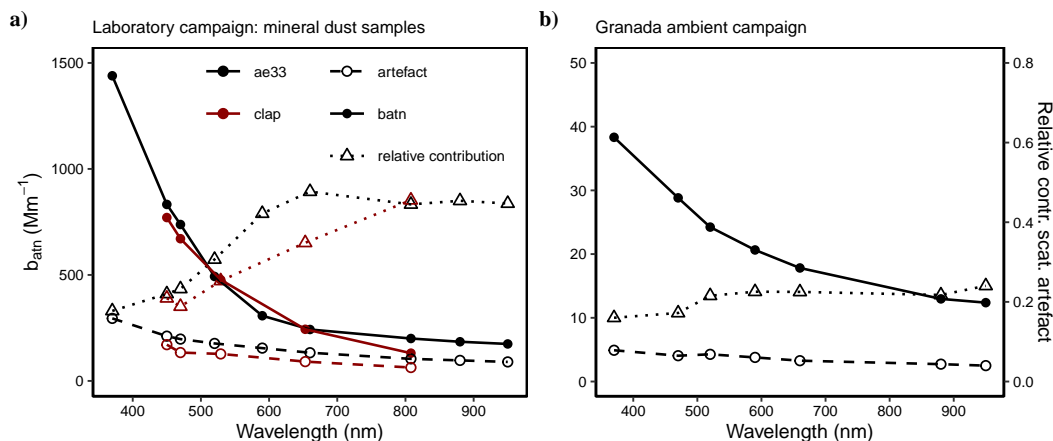


Figure 4. Dependence on the wavelength of the attenuation coefficient, the scattering artefact, and the relative contribution of the scattering artefact to the total attenuation measured by the AE33 and the CLAP (inter/extrapolated to 450 and 808 nm) for a) the laboratory campaign, and b) the ambient campaign in Granada.

Table 3. Multiple-scattering parameter C for both the Aethalometer AE33 (laboratory and ambient campaign) and the CLAP (laboratory campaign) obtained as the orthogonal fit of their attenuation coefficients corrected for cross-sensitivity to scattering vs. the PTAAM-2 λ absorption coefficients (Figs. S8-S24). Values have been reported at the reporting wavelengths of each FP (by inter- and extrapolation) and the wavelengths of the PTAAM-2 λ at which they were measured (marked in bold). The lower and upper 95% confidence interval of the C and the R^2 of the attenuation and absorption measurements used in the fit are shown in Tables S2 and S3.

Sample	AE33									CLAP				
	370	450	470	520	590	660	808	880	950	450	470	529	653	808
Icelandic dust	3.01	2.96	2.56	2.21	2.16	2.22	3.55	3.22	3.80	2.80	3.06	2.61	2.57	3.06
Wadi Rum (Jordan)	2.04	3.03	2.90	2.32	1.56	1.69	7.50	6.08	8.03	2.50	2.70	3.32	2.40	5.08
Sahara (Morocco)	1.83	3.13	3.10	2.90	1.92	1.93	5.39	4.29	5.46	2.54	2.49	2.78	2.00	1.92
Mojave (USA)	2.37	2.74	2.61	2.20	1.81	2.02	4.26	3.99	4.75	2.58	2.49	2.57	2.37	3.29
Propane Soot	3.55	4.08	4.04	3.92	4.03	3.75	3.95	3.83	3.77	5.10	5.22	5.19	4.68	4.26
Diesel soot	5.73	6.25	6.01	5.61	5.48	5.25	5.27	5.19	5.23	6.79	7.68	6.30	5.83	5.80
UGR - average	4.85	4.72	4.72	4.25	4.12	3.99	3.90	3.81	4.30					
UGR - urban	4.83	4.79	4.74	4.30	4.21	3.74	4.08	3.89	4.39					
UGR - urban + dust	4.93	4.59	4.69	4.15	3.90	3.74	3.76	3.59	4.12					
UGR - urban + wildfires	4.55	4.13	4.45	3.98	3.85	3.50	3.71	3.47	4.17					



is related to the limitations of the compensation model (eq. 1 and 2). Here we link this with a probable a non-linearity in the cross-sensitivity to scattering due to second-order effects associated with the particle properties and the instrument design, visible only at the lower absorption values and when the relative scattering contribution to the measured attenuation is the highest.

Di Biagio et al. (2017) performed a similar study, albeit for an aethalometer AE31 (Magee Scientific, USA) which has been shown to have C values lower than those of the AE33 (Drinovec et al., 2015; Savadkoobi et al., 2023), where they (Di Biagio et al., 2017) measured reference dust absorption using a CAPS PMex at 450 nm, and the extrapolated MAAP absorption coefficients at 660 nm. Di Biagio et al. (2017) multiple-scattering parameter for dust ranged between 3.64 and 5.12 for 450 nm and between 3.56 and 4.04 at 660 nm. Values of this parameter for dust at 450 nm in our study are much smaller: 2.74 to 3.13 at 450 nm and 1.69 to 2.22 at 660 nm. They also exhibit lower variability which is a combination of the lower variability of the particle size distribution in this study for dust, lower uncertainty due to cross-sensitivity to scattering due to different filter tape material (AE31 uses reinforced quartz tape) and the lower noise of the reference absorption instrument, since the extinction-minus-scattering method can introduce high systematic uncertainties in the case of high SSA (Modini et al., 2021).

Two types of soot were analyzed: propane (mean volume particle diameter, D_p , of 341 nm) and diesel soot (D_p of 177 nm), for which the C for the AE33 at 450/808 nm was 4.08/3.95 and 6.25/5.27, respectively (cf. Table 3). The CLAP featured C values at 450/808 nm of 5.10/4.26 for propane soot and 6.79/5.80 for diesel soot. These C values are consistent with the values found in Drinovec et al. (2022) for similar diesel and propane soot samples, between 5.3 and 3.2 at 532 nm, and between 4.2 and 2.6 at 1064 nm, for 100 and 500 nm volume size mode, respectively. There are some differences due to the difference in the measuring wavelength and the different flow used in the compared studies for the AE33. The C value of 1.44 recalculated for the CLAP from parameters in Ogren et al. (2017), based on the experiments with a PSAP from Bond et al. (1999) on aerosol mixtures of pure nigrosin with pure ammonium sulfate using also a reference extinction minus scattering measurement, is clearly lower than the nigrosin and/or any soot mixture measured in this study and in Drinovec et al. (2022).

During the ambient measurements at UGR, we found AE33 C values at 450/808 nm of 4.72/3.90 (cf. Table 3). These values were very similar to the propane soot measurements, and higher than those found by Drinovec et al. (2022) during a winter ambient campaign in Ljubljana, which were 3.28/2.57 at 532/1064 nm. Furthermore, the C did not vary with the type of event affecting the site — urban background pollution, fresh urban pollution, a dust event, or globally transported wildfire smoke. Dust events were determined using the Collaud Coen et al. (2010) methodology, dust forecasts and PM concentration levels measured on-site. Wildfire events were identified through the CAMS global forecast tool. Figure S2 shows that the volume particle size distribution during the urban background and the dust events (there were no APS measurements during the wildfire event) did not change significantly between the periods for the fine fraction below 400 nm, as previously shown in the measured urban background station (Casquero-Vera et al., 2020). However, the coarse fraction increases significantly during the dust events, associated to the advection of coarse particles from the Saharan dust outbreaks (Casquero-Vera et al., 2020). It is the combination of the absorbing soot-like particles and the dust particles present during the campaign that influenced the C , but the changes of their relative contribution did not have an effect on the resulting C values.



The C parameter found in the literature for ambient AE33 measurements compared to an offline polar filter photometer (Yus-Díez et al., 2021; Bernardoni et al., 2021; Ferrero et al., 2021), showed values at 520 and 660 nm ranging between 2.2 and 3.6, which were lower than those found in this study during the ambient campaign. The ACTRIS guidelines recommend a wavelength independent C parameter for AE33 ambient measurements using the M8060 filter of 2.44, using the MAAP (an online polar filter photometer) as the pseudo-reference. Indeed, when using the MAAP as a pseudo-reference instrument, following Yus-Díez et al. (2021) approach we have obtained a C parameter at 637 nm of 2.62 via the fit of eq. (1) and of 2.68 through an orthogonal fit, with the 95% confidence interval ranging between 2.55 and 2.81 (cf. Fig. S3). Here, as in Bernardoni et al. (2021) and Drinovec et al. (2022), the shorter wavelengths also showed higher C values. The systematic differences between direct measurements with the PTAAM-2 λ and the MAAP are discussed below in section 3.3.

3.2.1 Dependence on the particle size

Most remarkable features of Table 3 are the higher C values found for soot-like particles in comparison with dust particles. As shown in Drinovec et al. (2022), this is related with the size of the particles, with smaller particles showing higher C values. Figure 5 shows the variation with the size of the C values for the different sample types and the UGR campaign. It should be noted that the particle size of the dust samples was measured with an OPC, which measures the optical diameter, whereas for diesel and propane soot particle size was measured with an SMPS, which measures the mobility diameter. During the UGR summer campaign particle size was obtained as the combination of the SMPS and the APS size distributions, reported in mobility diameter. Therefore, any comparison on the behavior with the particle size between dust and non-dust samples in Fig. 5 should be performed while keeping in mind the differences among these different diameters.

Figure 5 shows that both the AE33 and CLAP feature C values that, albeit higher, exhibit the same trend as those found in Drinovec et al. (2022), where it was shown in the sub- μm particle mobility diameter region, that CLAP measurements had a higher C than AE33 for the smaller particles, and vice-versa for the coarser particles. Here, we see that there is a considerable decrease of the C at 450/808 nm within the fine mode for soot, from 6.41/5.39 at 178 nm volume size mode (D_p) to 4.77/4.76 at 341 nm for the AE33 and from 7.64/5.73 to 5.32/4.23 for the CLAP. At Granada, the C for the AE33 ranged between 4.64/4.47-3.97/3.55 at 450/808 nm, very similar to the coarser diesel soot particles measured in the laboratory. As the particles become coarse ($>1 \mu\text{m}$), there is a significant decrease and consequent leveling-off in the parameter C, with values around 2.6-3.0 for both AE33 and CLAP at 450 nm.

It should be noted that at 808 nm, the C shows a high variability, as also shown in Table 3 for wavelengths over 660 nm. As can be observed in Fig. S25, the higher variability of the C for dust particles is mainly driven by the variability between the different samples and the higher relative contribution of the scattering by particles to the attenuation measured by the FP. Figure 5 shows that the CLAP features a higher dependency of C on the particle size, with higher values at smaller diameters (below 500 nm) and similar values for the broader region of larger diameters. It should be noted that the ambient Granada aerosol sample is an external mixture of a multi-mode aerosol particle size distribution (cf. Fig. S2) of fine soot-like particles, coarse dust particles and other fine and coarse particles. Therefore, D_p is only a proxy of the size of the total mixture that averages the effect of the absorbing fine soot-like with higher C values and the coarser dust particles with lower C values.

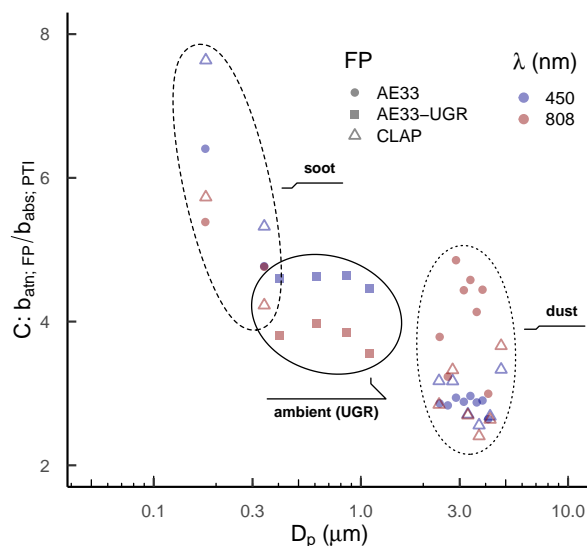


Figure 5. Dependence of the FP multiple-scattering parameter C , for AE33 and CLAP, on the volume particle diameter (D_p). C is obtained as the ratio of the FP attenuation coefficient, corrected for the scattering artefact, and the PTAAM- 2λ absorption coefficient at 450 and 808 nm. AE33 and CLAP refer to the laboratory measurements, whereas AE33-UGR refers to the ambient measurements performed in Granada, Spain.

3.3 MAAP artefacts

The MAAP is considered in many studies a pseudo-reference instrument that has been used to characterize AE31 and AE33 artefacts (e.g., Di Biagio et al., 2017; Yus-Díez et al., 2021; ECAC-CAIS, 2022). It measures the backscattering of light from the sample-laden filter at two angles that are used to parametrize the angular distribution of light, and correct for the (multiple) scattering of the light by particles within the filter through an internal algorithm (Petzold and Schönlinner, 2004). However, as shown by Valentini et al. (2020), the MAAP design limitations (only two angles measuring backscattered radiation) hinder its ability to fully represent the angular distribution of light scattered by the sample-laden filter. Valentini et al. (2020) shows that the MAAP-derived absorption coefficient differs by 14% from the one provided by the higher angular resolution PP_UniMI (Vecchi et al., 2014).

Figure 6 shows the ratio of the MAAP-derived absorption coefficient and the the absorption coefficient measured by PTAAM- 2λ changing with the ambient SSA, all at 637 nm. It shows overestimation by the MAAP relative to the traceably-calibrated PTAAM- 2λ : a clear deviation of the absorption coefficients ratio from 1 at low SSA, as well as a large increase for the MAAP absorption coefficient relative to the PTAAM- 2λ at SSA values above 0.95. The result of the fit showcases an artefact – cross-sensitivity to scattering, by the MAAP of m_s of 2.4%. Although it is 2.5 times lower than what has been found for the same period for the AE33 (Fig. 3b) and Table 2, it is still remarkable, since for the highest SSA measured, the

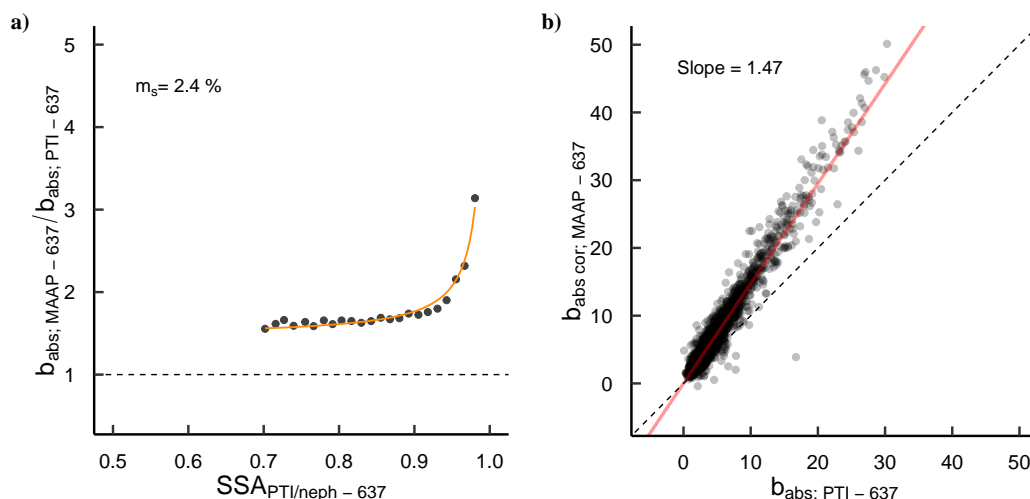


Figure 6. a) Evaluation of the cross-sensitivity to scattering for the MAAP, through the ratio of the MAAP and PTAAM-2 λ absorption dependence on SSA, and b) relationship between the MAAP absorption corrected for the scattering artefact vs the PTAAM-2 λ absorption.

MAAP absorption was more than double the one measured with the PTAAM-2 λ . After correcting the MAAP absorption for its cross-sensitivity to scattering (following eq. (1) but for $b_{\text{abs}}(\text{MAAP})$ instead of b_{atn}), Fig. 6b shows a 47% overestimation of the MAAP absorption values in comparison with the PTAAM-2 λ reference measurements.

Similarly to the AE33 and the CLAP, a dependence on the particle size in the ratio between MAAP-derived absorption coefficient and the reference one was observed. However, the particle diameter did not present such a wide size range as during the laboratory experiments, hence conclusions are limited (Fig. S26). In this regard, Fig. S5 from Romshoo et al. (2022) showed that the MAAP and AE33 (harmonized to the MAAP) absorption ratio to extinction-minus-scattering measurements as a reference also had ratio between 1 and 1.5 for aerosols with a volume mean diameter below 100 nm, and that it converged towards 1 with increasing particle size, especially for particles with a volume mean diameter above 100 nm. Similar as in our study, Romshoo et al. (2022) shows that the decrease of the ratio with size is more pronounced for the AE33 than for the MAAP.

The presented cross-sensitivity to scattering artefact and multiple-scattering parameter values for MAAP and AE33 have large implications for the reporting of absorption coefficient in ambient measurement networks. For example, the MAAP deployed at Granada was part of Savadkoohi et al. (2023) as an effort to harmonize equivalent Black Carbon (eBC) AE33 measurements across Europe following the ACTRIS guidelines (Müller and Fiebig, 2021). These guidelines used the MAAP as a pseudo-reference instrument, and measured the deviation between the AE33 and MAAP eBC under different scenarios. This deviation was parametrized with the ACTRIS harmonization factor ($H^*=1.76$; ECAC-CAIS, 2022), a multiplicative parameter to compensate the AE33 measurements across different measurement sites across the ACTRIS network. This H^* factor is computed as: $H^* = \text{eBC}_{\text{AE33}} * \sigma_{\text{AE33}} / b_{\text{abs,MAAP}}$, where eBC_{AE33} and σ_{AE33} are the AE33 reported black carbon



350 concentration and the default AE33 MAC, respectively, interpolated to the MAAP wavelength, and $b_{\text{abs,MAAP}}$ is the absorption coefficient provided by the MAAP.

Following the methodological approach proposed by the ACTRIS guidelines (ECAC-CAIS, 2022), we have obtained a Granada-specific $H^* = 1.89$ (Fig. S27), and 95% confidence interval ranging between 1.86 and 1.92, which is about a 7% higher than the ACTRIS value. This difference can be explained by the different aerosol sample measured as well as by the instrument
355 inter-variability. Therefore, to properly correct the AE33 measurements using the MAAP as a pseudo-reference instrument, the additional deviation factor of 1.47 between the MAAP and the PTAAM-2 λ has to be introduced into the harmonization factor to account for the artefacts of both instruments, increasing the H^* to $1.76 \cdot 1.47 \simeq 2.6$. The variability of this additional factor needs to be investigated, as we have shown it is dependent on the correction for SSA effects.

This overestimation by the MAAP, then propagated to the AE33 measurements (cf. Fig. S3), results in a significant over-
360 estimation of the eBC, Mass Absorption Cross-section (MAC) and absorption enhancement (E_{abs}) values reported for measurements that used either instrument (e.g. Zanatta et al., 2016; Yus-Díez et al., 2022; Savadkoochi et al., 2023). Indeed, the MAC reported in Zanatta et al. (2016) for MAAP was higher (1.47 on average) than with the PSAP-ITM, however the sites backgrounds were not strictly comparable.

An overestimation of the MAAP measurements when compared with the PTAAM-2 λ absorption of 1.41 has also been found
365 during the laboratory experiments performed within the framework of the stanBC project (European Partnership on Metrology; <https://stanbc.com>), which aims to provide a methodology to standardize the measurement of the aerosol absorption coefficient, BC and eBC from FP measurements and the manuscript is in preparation. In addition to laboratory experiments, further ambient campaigns where the PTAAM-2 λ is deployed should be performed to provide further details on the effects that influence the sensitivity and performance of filter photometers in general, and the MAAP as a pseudo-reference in particular.

370 4 Conclusions

In this study we have characterized the multiple-scattering artefacts of the most widely used filter photometers (FP) during a laboratory and an ambient campaign where the traceably-calibrated reference instrument for absorption, the dual-wavelength photothermal interferometer, PTAAM-2 λ was deployed. The investigated FPs were: an aethalometer AE33 and a continuous light absorbing photometer (CLAP) during the laboratory campaign, and an AE33 and a multi-angle absorption photometer
375 (MAAP) during the ambient campaign. Mineral dust and soot-like samples were analyzed during the laboratory campaign, whereas the ambient campaign was performed during summer at an urban station in Granada (UGR) with varying composition of urban and regional aerosols, rich in soot and dust.

We have determined cross-sensitivity to scattering due to coarse dust samples using a non-absorbing quartz sample. The parameter m_s is lower for the CLAP than the AE33, which is linked with the instrument design, with values range 3.0/1.5%
380 at 450/808 nm for the AE33 and 2.4/0.9% at 450/808 nm for the CLAP. Furthermore, we have derived the cross-sensitivity to scattering at the urban background station, where the AE33 values 6.1/6.7% at 450/808 nm but with larger values around 8%



in the mid-visible range. The larger scattering artefact found at the UGR is associated to the presence of finer particles, which result in a higher cross-sensitivity to scattering.

The sample variability in this study has enabled the study of the dependence of the multiple-scattering parameter, C , across a wide range of particle sizes. We have found that the multiple-scattering for both CLAP and AE33 decreases sharply with size within the fine fraction, and levels-off for coarse particles. As a result, the finer samples feature C values at 450/808 nm for the AE33(CLAP) that range between 4.08/3.95 (5.10/4.26) for propane soot and 6.25/5.27(6.79/5.80) for diesel soot. For the mineral dust samples, the C ranged between 2.74 and 3.13 for the AE33 and between 2.50 and 2.80 for the CLAP at 450 nm. At UGR, the aerosol particle composition is a mixture of local/regional emissions and dust, which results in a particle size in-between the soot and the mineral dust lab samples with a value of 4.72/3.95 at 450/808 nm.

Overall, the multiple-scattering parameters C , found in this study for both the AE33 and the CLAP were higher than those presented in previous studies, where filter photometers as the MAAP or similar are used as a reference. Here, thanks to the co-located MAAP and PTAAM-2 λ measurements during the ambient campaign in Granada, we have shown that the MAAP absorption coefficients overestimate the absorption by a 47%. Moreover, despite the MAAP's more complex design and absorption retrieval scheme, we have found a cross-sensitivity to scattering of 2.4%.

This study provides a comprehensive analysis on the multiple-scattering compensation parameter, C , for the widely used AE33 and CLAP filter photometers, as well as first characterization of the pseudo-reference MAAP. It provides the platform for accurately compensating the FP-derived aerosol absorption coefficients, highlights the importance of correcting for scattering artefact, and the need for the knowledge of the particle sizes. Finally, it showcases the need for reference absorption measurements and the re-evaluation of previous reports on absorption coefficients and mass absorption cross-sections based on the MAAP.

Author contributions. LD, GM, GT, LAA and JYD conceived and designed the study. LD performed the laboratory campaign measurements. AGR, JYD, AA, XQ and CPGP were involved in the field campaigns that collected the bulk mineral dust samples. GT, LAA, EB, DP, AC, LD, GM, JYD were involved in the installation, operation, maintenance and calibration of the instruments before, during, and/or after the campaigns. JYD processed the data, and LD, GM and JYD prepared the manuscript. All co-authors contributed to the preparation and the scientific discussion of the manuscript, and reviewed it.

Competing interests. LD and GM are employed by Haze Instruments d.o.o., the manufacturer of PTAAM-2 λ . Other authors declare no potential conflict of interest.



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