

EGUSPHERE-2025-2823: “The role of size in the multiple scattering correction C for dual-spot aethalometer: a field and laboratory investigation”

Answers of the authors to Reviewer#1

While the reviewer’s comments are given in **black bold**, our answers are given below in [blue letters](#). Additionally, we added the changes made in the revised manuscript in *blue italic letters*.

The manuscript deals with an important and often discussed topic, the multiple scattering correction factor “C” and related size effects reported for aethalometers. The authors present new data from laboratory and field experiments. The data analysis is well-founded and reproducible. The results are discussed in detail and the relevant literature is well reviewed.

The results are important for the community to compare light absorption or equivalent soot content across measurement networks and also as a basis for calibrating the Aethalometer AE33.

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. Here we provide some major considerations. Both reviewers raised important points regarding the suitability of the filter-based instruments used as reference methods for ambient measurements. We have addressed this issue carefully by expanding the discussion in the results, incorporating recent literature, and revising comments related to the interpretation of our results. Additionally, both reviewers through different comments emphasised the need to clarify and deepen the analysis of the dependence of the multiple scattering correction factor (C) on particle size further, as this is a key aspect of the manuscript. They also recommended integrating the discussion of results obtained under controlled chamber conditions and in ambient more closely.

Accordingly, we have significantly revised and expanded in particular Sections 3.1.2 and 3.2.4 in the results, providing additional detail and context, and also improved Abstract, Introduction and Conclusion to better clarify the aim of the present study.

General comment:

MWAA and MAAP are used as references. As this is crucial to the work, further information is needed to justify the suitability of these instruments as references, since they are filter-based techniques. Specifically, has it been demonstrated that the size dependence is negligible for their use as references? Please comment on this.

Both reviewers emphasized the importance of this aspect, which is critical for interpreting results obtained for atmospheric observations at CMN where the MAAP and the MWAA were used as reference instruments. To fully address this common point, we provide a more complete literature study and implications in the main text.

As noted, the reliability of filter-based instruments for measuring the absorption coefficient is debated in the literature. A recent study by Yus-Díez et al. (2025) shows that MAAP can overestimate absorption by up to 47% when compared to PTAAM-2A, particularly for aerosols with high single scattering albedo ($SSA > 0.9$). On the other hand, Heuser et al. (2025) reported an excellent comparison (less than 10% differences) when comparing filter-based techniques (MAAP, MWAA, PP-UniMi) with EMS for the highly absorbing CS particles from laboratory experiments in this study with $D_{m,vol}$ between 130 and 450 nm. Earlier work by Romshoo et al. (2022) reported good agreement between MAAP and the EMS technique for particles with a volume mean diameter $D_{m,vol} > 100$ nm, but MAAP slightly overestimated absorption (~20%) for smaller particles ($D_{m,vol} < 100$ nm). Similarly, Kalbermatter et al. (2022) demonstrated that MAAP tends to overestimate absorption measured with other non filter-based techniques by at least 20%, especially for particles with a high total mass-to-BC mass ratio. Finally, Moteki et al. (2010) highlighted that the two-stream model on which the MAAP is based may underestimate its size dependent bias, when stratification occurs in the filter, in particular for particles smaller than 200 nm.

Together, these studies indicate that filter-based reference instruments such as MAAP and MWAA may be subject to systematic biases, but the magnitude of these biases depends on particle properties and the reference technique used, being larger for: high SSA (> 0.9), small particle diameters (< 200 nm) and BC particles mixed with secondary organic materials.

Although further investigation is needed, using these references in our study could lead to a possible general underestimation of the C factor in ambient measurements. If PTAAM-2 λ or photoacoustic techniques are considered as more accurate references, the underestimation could reach up to 47%, particularly for high SSA and mixed particles. This affects not only the absolute C values but also the observed dependencies of C on SSA and particle size. At CMN, the C dependencies we report may therefore be underestimated for SSA > 0.9 and diameters < 200 nm, while estimates for SSA < 0.9 and diameters > 200 nm are expected to be more reliable.

To better include this discussion, we extended the text in **Section 3.2.1, 3.2.3 and 3.2.4**. In particular, we added these paragraphs:

- the text in **Section 3.2.1**

...*"The lower ambient values may be partly due to the different reference techniques used. The MAAP can exhibit a positive bias in absorption when compared with non-filter-based reference techniques as the EMS, particularly at relatively high SSA (>0.9), small particles (diameter <200 nm) and for aged aerosol (Yus-Díez et al., 2025; Kalbermatter et al., 2022; Romshoo et al., 2022). Such bias would lead to higher MAAP absorption values and, consequently, to lower C values in ambient measurements at CMN compared to those obtained in CESAM."*...

- the text in **Section 3.2.3**

...*"Moreover, we should consider that this dependence, estimated using the MAAP as a reference, may have been underestimated. As shown by Yus-Díez et al. (2025), this instrument is also affected by cross-sensitivity to scattering when compared with a non-filter-based technique."*...

- the text in **Section 3.2.4**

...*"This attenuation may be partly related to the use of the MAAP. Its positive bias compared to non filter-based techniques could be more relevant for smaller particles (Moteki et al., 2010; Romshoo et al., 2022). As a result, the influence of particle size on C under ambient conditions could be partially masked by the measurement bias inherent to the reference technique used."*...

Added reference:

Romshoo, B., Pöhlker, M., Wiedensohler, A., Pfeifer, S., Saturno, J., Nowak, A., Ciupek, K., Quincey, P., Vasilatou, K., Ess, M. N., Gini, M., Eleftheriadis, K., Robins, C., Gaie-Levrel, F., and Müller, T.: Importance of size representation and morphology in modelling optical properties of black carbon: comparison between laboratory measurements and model simulations, *Atmospheric Measurement Techniques*, 15, 6965–6989, <https://doi.org/10.5194/amt-15-6965-2022>, 2022.

Section 2.1. Experimental setup

The manuscript focuses primarily on observation rather than a detailed discussion of the reasons for size dependence. According to Moteki et al. (2010) and Nakayama et al. (2010), the reason for size dependence is the penetration depth of the particles, which is a function of sampling face velocity. The datasets collected at CMN and CESAM use data from AEE collected at different flow rates (see Table 2). Could the differences in flow rate influence the C factors of CMN and CESAM?

Thanks to the reviewer's comment, we further explored this aspect based on the findings of Moteki et al. (2010), Huang et al. (2013), and the filtration theory described by Hinds (1999). These studies demonstrate that particle penetration and its dependency on particle size changes as the face velocity, and therefore the flow, varies; this has an impact on the relationship between the absorption coefficient measured with filter-based instruments and the real absorption coefficient.

More specifically, in the work of Huang et al. (2013, Figure 1), increasing the face velocity from 10 to 30 cm/s for specific filter characteristics result in 20% more penetration for particles with a diameter of 200 nm, and a more pronounced dependence of the penetration on particle size.

According to Moteki et al. (2010), where the absorption coefficient measured by a filter-based instruments is modelled through two-stream and four-stream approximation, the ratio between the absorption coefficient measured by filter-based technique $B_{\text{abs_filter}}$ and the true absorption B_{abs} , is strongly affected by the particle penetration in the filter. $B_{\text{abs_filter}}/B_{\text{abs}}$ increases with penetration up to a certain depth for then decreasing again Moteki et al. (2010, Figure 2). Since most of the particles in the filter are usually collected in the first layers of the filter and smaller particles penetrate more deeply into it (Moteki et al., 2010; Figure 3), based on this study we expect to observe a decrease of C with particle size. At the same time an increase in the flow can significantly affect the penetration of all the particles causing more particles to better penetrate the filter matrix, lowering the $B_{\text{abs_filter}}/B_{\text{abs}}$ and changing its size dependence. According to Moteki et al. (2010, Figure 4), $B_{\text{abs_filter}}/B_{\text{abs}}$ decreases for increasing flow rates from 300 to 900 cm^3/min . And the size dependence of this ratio is more pronounced at the lower flow for particles bigger than 200 nm and less pronounced at the lower flow for particles smaller than 200 nm.

A detailed quantification of the implications of a different flow rate for our specific conditions would require applying filtration theory specifically to the M8060 aethalometer filter as well as to the MAAP filter, with the corresponding flow rates, and it is beyond the scope of the present study. However, the decrease in C and a more smoothed dependence on size for particles diameters in the range of our ambient observations seems to be in agreement with the results of Moteki et al. (2010). It is thus likely that the difference in the flow rates between chamber and ambient measurements could explain part of the difference observed in the two conditions.

To better include this aspect in the discussion, we extended the text in **Section 3.2.1 and 3.2.4**. In particular, we added these paragraphs:

- the text in **Section 3.2.1**

... "In addition, the aethalometers operated at different flow rates in the ambient and chamber setups (5 and 2 L min^{-1} , respectively). As shown by Moteki et al. (2010) and Huang et al. (2013), variations in flow rate and face velocity can affect particle penetration and stratification by size within the filter. These effects may modify both the absolute value of C (which is expected to decrease with increasing flow rate) and its dependence on particle size. A detailed quantification of this effect is beyond the scope of the present study, but it may represent one of the factors contributing to the lower absolute C values observed in ambient measurements. "...

- the text in **Section 3.2.4**

... "Differences in sampling flow of the aethalometer between the ambient and chamber measurements may also have contributed to this weaker dependence, since different flow rates are expected to affect the penetration of the particles as well as the resulting C dependence on size (Moteki et al., 2010; Huang et al., 2013). "...

3.1.1 Loading correction, lines 310 to 319

Since the DUAL spot correction factors require some time to adapt to new aerosol types, it is advantageous when changing aerosols to first load one spot and then analyse only the following spots. Another different approach used in this study is data post-processing. In addition to the existing compensation methods for single spot instruments, the dual spot aethalometer method could be used with a constant compensation factor determined by an adjustment procedure. This would have the advantage of not requiring the introduction of a new correction

function. The best-fitting compensation parameter should also be given, as it could be helpful for future studies on the dependence of the compensation parameter on the aerosol type.

We agree with the reviewer that using the dual spot model of the aethalometer would be the ideal post-processing approach, and it is indeed the approach we initially tried to apply before proceeding with a different fit. Unfortunately, however, this approach did not lead to complete compensation for ATN dependence, so we preferred to test a different function and perform specific fits for each filter spot. This ensured optimal compensation but is certainly not a method that can be extended to other contexts. For the sake of completeness, however, we also add the Dual Spot method to those discussed in the Supplementary Information and report the corresponding average compensation parameters (k) for each type of aerosol studied.

The text in Section 3.1.1 was changed to add the Dual Spot method.

- the text in **Section 3.1.1**

... "In these cases, the AE33 data can be re-corrected for loading effect. With this aim, the AE33 can be treated like a single-spot aethalometer and the loading correction schemes from W2003 and C2010, or the Dual Spot aethalometer method itself with a constant compensation parameter, be applied." ...

The text in Supplementary Information was modified to include the description of the Dual Spot method and the comments to the corresponding results:

- the text in **Text S1**

... "- the Dual Spot method defined in Drinovec et al., (2015; equations 3 and 4) applied, using a constant compensation parameter, to b_{atn}/b_{REF} instead of BC as follows:

$$\frac{b_{atn}}{b_{REF}} = \left(\frac{b_{atn}}{b_{REF}} \right)_0 (1 + RS \cdot ATN), \quad (S3)$$

$$RS = \left(\frac{b_{REF}}{b_{atn}} \right)_0 \cdot \frac{d \left(\frac{b_{atn}}{b_{REF}} \right)}{dATN}. \quad (S4)$$

The RS and $\left(\frac{b_{REF}}{b_{atn}} \right)_0$ were obtained through a linear fit applied to each filter spot and used to obtain the k values for correcting b_{atn} according to Drinovec et al., (2015; equation 7). Table S4 summarizes the averaged k values for each CS type. "...

- the text in **Text S2**

... "We examined the dependence of C on attenuation using data corrected for the loading effect through the other methods described in Sect. 2.2.1: W2003, C2010, Dual Spot and the fitting method. Results for these different correction algorithms are also shown in Fig. S4. The C2010 method (Fig. S5b) introduces a significant bias, with a spread of up to 45% for C, particularly below an ATN of 20. The W2003 method (Fig. S5c) reduces this bias to approximately 35%, but still leaves notable inaccuracies, especially at low attenuation values (<10). The Dual Spot method reduce the bias but introducing inaccuracies for high attenuation (>50), without solving the jumps between spots Fig. S4." ...

- the text in **Text S3**

... "The internal correction and Dual Spot methods both yield a 71% increase, similar to the 77% obtained with the fitting method developed in this study." ...

- Table S4 was included to report the values of the averaged k as requested by the reviewer.

Table S4: Mean k values derived applying the dual spot method to all filter spot for each CS type.

	CS1	CS2	CS3	CS4	CS5
k mean	0.0101	0.0097	0.0112	0.0125	0.0224

- Table S3 was modified to include the Dual Spot correction.

Table S3: Average C values, and their standard deviations, determined from one-minute resolution attenuation coefficient and absorption coefficient values, corrected for the loading effect using different methods; AE33 internal correction, W2003, C2010, Dual Spot, and an experiment fitting specifically obtained for this study.

Method	Internal correction	W2003	C2010	Dual Spot	Fitting
Mean (SD)	3.16 (0.50)	2.77 (0.3)	2.88 (0.56)	3.40 (0.64)	3.54 (0.6)

- Figure S4 was changed to include the Dual Spot correction. The modifications include: i) adding the line for dual spot correction; ii) different colouring to improve readability; iii) adjustment of axis labels and legend.

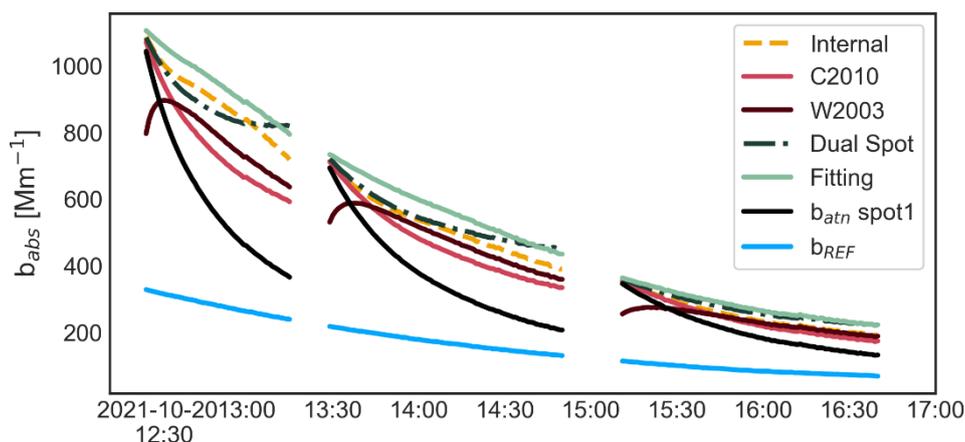


Figure S4: Variation of b_{atn} and b_{abs} at one-minute resolution during a representative chamber experiment. The periods of interruption in the series correspond to the aethalometer filter changes. The lines represent the reference measurement, the attenuation coefficient uncorrected for loading measured in spot 1 of the aethalometer at 660 nm, the loading-corrected attenuation coefficient using the C2010, W2003, Internal method, Dual Spot and Fitting methods, respectively.

- Figure S5 was changed to include the Dual Spot correction. The modifications include: i) adding a subplot for dual spot correction; ii) adjustment of axis labels and legend.

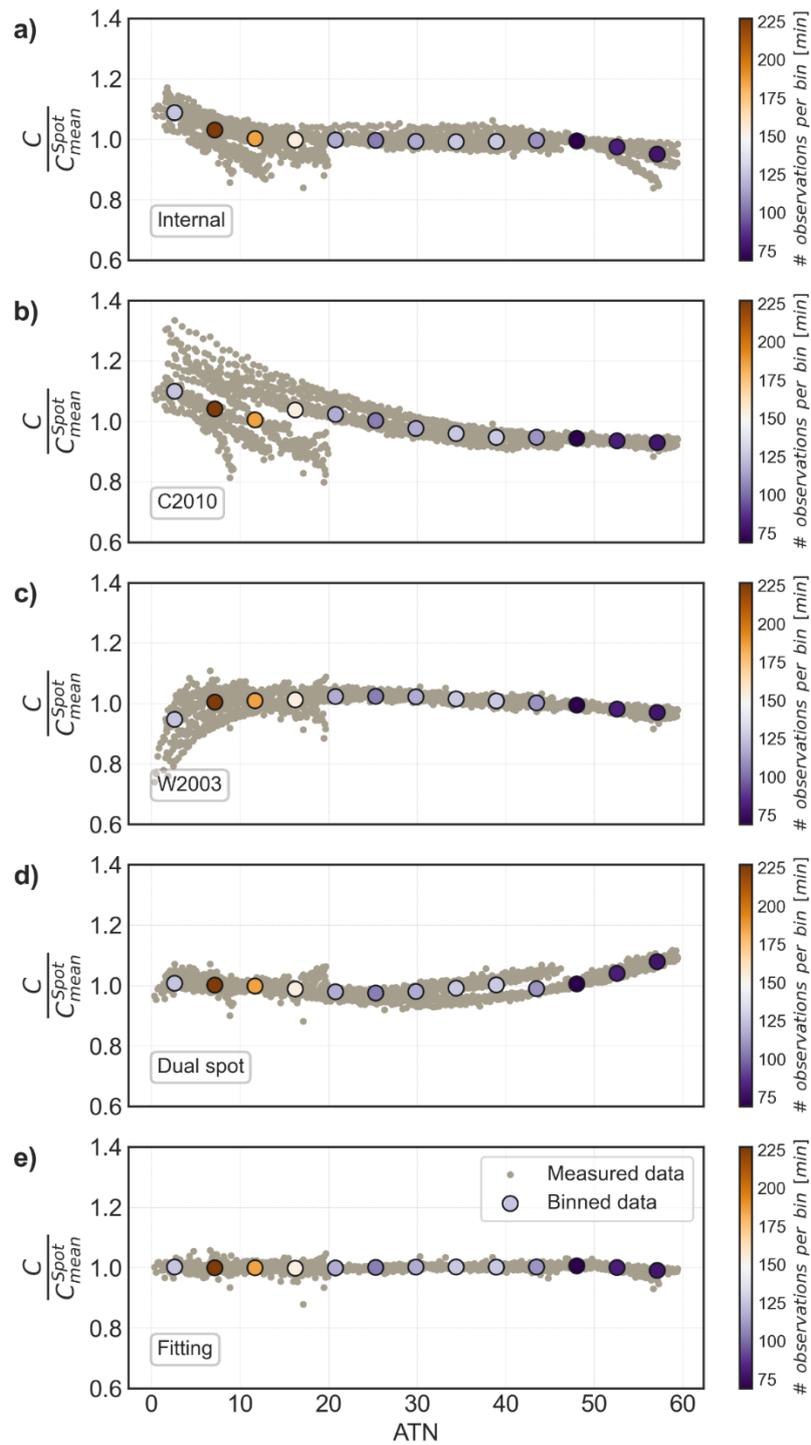


Figure S5: Variability of C values normalized for their average in each filter spot with attenuation at 660 nm. The grey points represent the one-minute resolution C values, while the colored points indicate the median values for each attenuation bin. The color intensity corresponds to the number of data points within each bin, providing a clear visualization of the data distribution. Data correspond to all CS1-CS5 experiments.

- Figure S6 was changed to include the Dual Spot correction and to use the volume mean diameter $D_{m,vol}$ instead of the geometric mean diameter as detailed in the last comment. The modifications include: i) adding a line for dual spot correction; ii) modifying the lines for using the $D_{m,vol}$ instead of the geometric mean diameter; iii) adjustment of axis labels and legend.

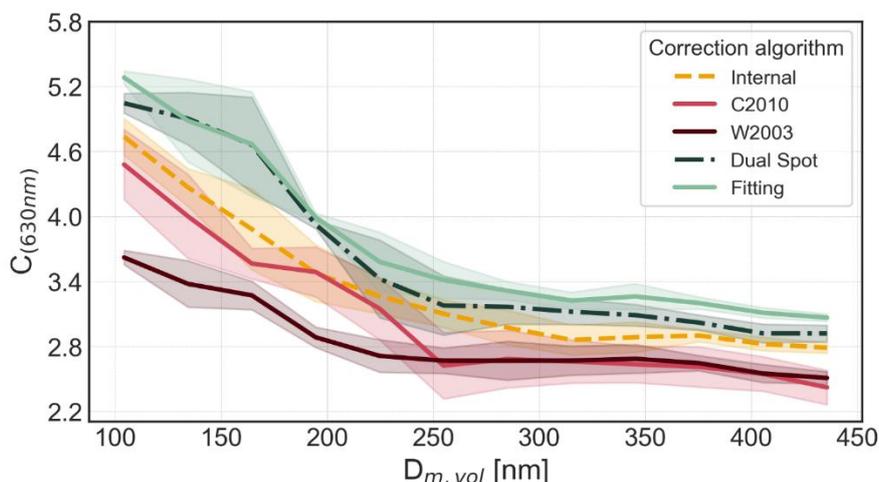


Figure S6: C dependence on particle size in laboratory studies. One-minute resolution C values are binned on the volume mean diameter ($D_{m,vol}$). The line represents the mean values in each bin the coloured area the mean \pm the standard deviation. One line is displayed for the C values derived from $b_{am,LC}$ loading corrected using different methods (Internal, C2010, W2003, Dual Spot and the 'Fitting' method).

Line 432:

A single scattering albedo of equal to 1.0 is probably due to measurement noise. Values below the lower limit of babs for C determination should be removed.

We agree with the reviewer. To note however that 1.00 is the upper limit we set, but the real maximum value is slightly lower (0.99), in particular after removing b_{abs} values below 0.06 Mm^{-1} . To avoid inaccuracies, we have rephrased the text in line 432.

- The text in results **Section 3.2.3**.

... "The C values measured during the period considered in this study, are binned to the SSA values (Figure 4)."...

Figure 5 and related text: Is the geometric mean diameter of the number size distribution the best indicator of "size"? Are there other moments of the size distribution that better capture the proportion of soot in the total aerosol? These could possibly be more consistent with the laboratory results.

We thank the reviewer for this insightful comment. The geometric mean diameter despite being widely used as a metric in aerosol studies, it's not necessarily the best metric. A moment that would better capture the proportion of soot in the total aerosol and assure a more consistent comparison between laboratory and field could be the volume mean mobility diameter, or the mean diameter weighted based on the volume size distribution. In ambient measurements, the number distribution is mainly shifted toward the smallest particles that contribute less to the total mass and optical properties, the volume diameter instead emphasises the contribution of the size range of soot particles. Following this suggestion, we replaced the geometric mean diameter with the volume mean mobility diameter ($D_{m,vol}$).

This change has significantly expanded the range of diameter variability, shifting it toward larger diameters. In chamber experiments this just resulted in points shifted towards larger sizes. In ambient conditions the increase in C as particle size decreases becomes clearer and more pronounced, with a 25% difference between the two extremes.

To account for these changes, we modified:

- the text in **Abstract**

... "Both ambient and chamber experiments highlight the dependence of the C factor on particle size, with C increasing as particle diameter decreases below 200 nm. This size dependence is relatively small (within 25%) under ambient conditions dominated by mostly scattering aerosols, but it leads to changes greater than 60% for highly absorbing soot particles. "...

- the text in method **Section 2.3.2**

... "The size distribution measured by the SMPS was used to derive the volume mean mobility diameter ($D_{m,vol}$). This parameter was derived from hourly averaged size distributions at CMN and to both 3-minutes size distributions and size distribution averaged on each filter spot of the aethalometer in chamber experiments.

To notice that in the field the size distribution is multi-modal and include both absorbing and non-absorbing particles, in chamber experiments they were mono-modal and including only absorbing aerosols. "...

- the text in **Section 3.1.2**

... "The analysis shows that the C factor increases from 2.75 to 4.45 as the volume mean particle diameter of BC generated particles decreases from 444.8 nm to 124.9 nm. This is particularly evident for particles smaller than 200 nm, while at larger sizes the trend is not significant when considering the associated uncertainties. The points with the highest C values and smallest sizes correspond to experiments involving CS5 particles. These particles are not only smaller but also differ in composition, with a negligible fraction of EC in spite of dominating OC composition. This compositional difference significantly affects the optical properties, increasing the SSA from 0.05-0.09 for C1-CS4 to 0.27 for CS5 at 630 nm and the AAE from around 1.27-1.88 to 3.79 (Heuser et al., 2024). However, focusing on a subset of points with limited size variability (230-300 nm, highlighted in the grey box), it becomes evident that at similar diameters, C values are comparable for CS5 particles and CS1-CS4 particles. Moreover, for CS5 particles, C increases significantly from 3.15 to 4.45 as the volume mean diameter decreases from 247.9 nm to 124.9 nm. "...

- the text in **Section 3.2.4**

... "In this section, we examine the dependence of the C factor on particle size under ambient atmospheric conditions, where isolating the influence of individual aerosol properties is not straightforward. The objective is to evaluate whether particle size remains a significant driver of C variability. The analysis was performed by binning C values according to the aerosol volume mean diameter and computing the median together with the 25th and 75th percentiles within each bin, following the same procedure adopted for SSA.

During the observation period at CMN, the hourly $D_{m,vol}$ of total aerosol particles ranged from 134.5 nm to 738.9 nm. The resulting C -size relationship shows an increase of approximately 25% in C as $D_{m,vol}$ decreases from 732.1 nm to 141.4 nm (Figure 5). This trend is consistently observed across the median as well as the interquartile range, indicating a clear dependence of C on particle size. "...

- Figure 1 was changed to modify the $D_{m,geo}$ in $D_{m,vol}$. The modifications include: i) modification of all points to use the new variable; ii) adjustment of axis label and subtitle.

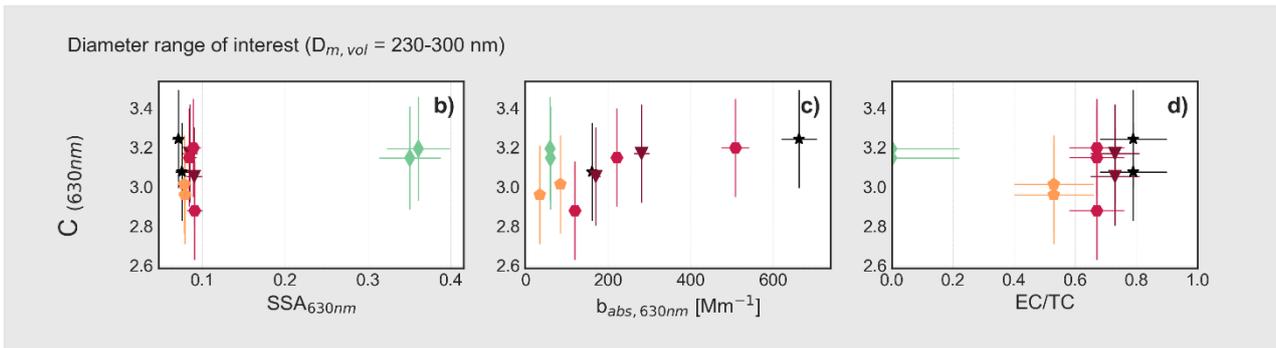
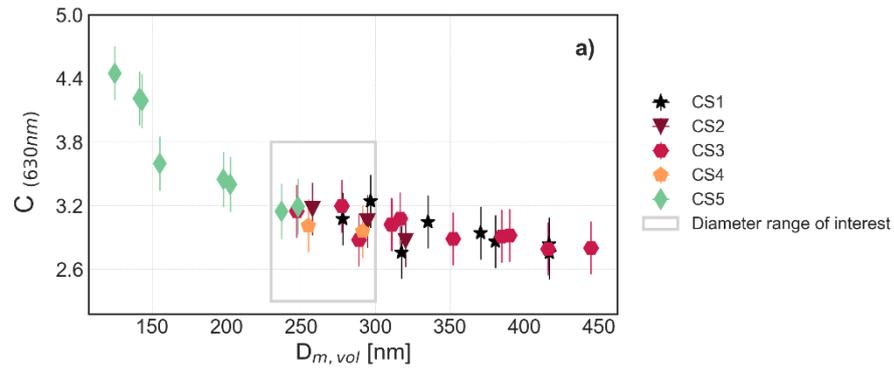


Figure 1: C dependence on particle size in laboratory studies measuring the properties of different freshly emitted particles (CS1-CS5). The top panel shows the C dependence on the size ($D_{m,vol}$). The bottom panels show the dependence of C values with the SSA, b_{abs} (measured by the EMS technique) and the EC/TC ratio, in experiments where the diameter was in the range 230-300 nm (grey rectangle in the top panel).

- Figure 5 was changed to modify the $D_{m,geo}$ in $D_{m,vol}$. The modifications include: i) all values in the plot were changed to use the modified variable; ii) adjustment of axis label and legend.

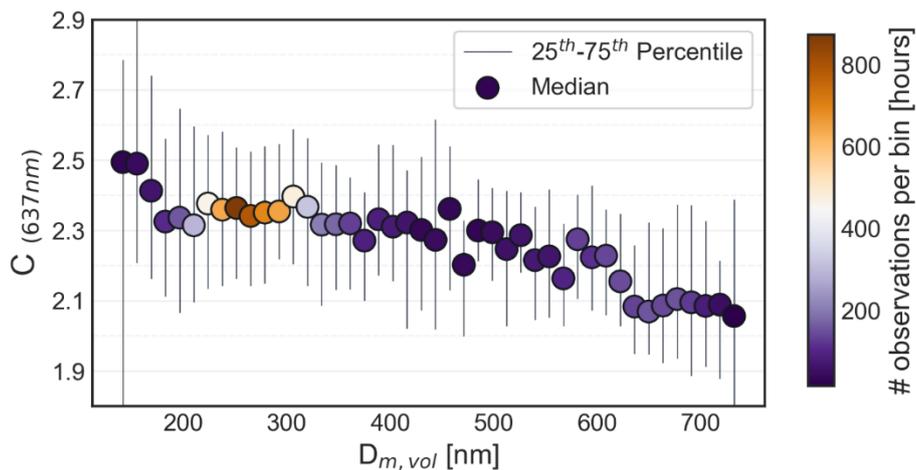


Figure 5: C (637 nm) dependence on the geometric mean diameter $D_{m,vol}$. Reference absorption instrument: MAAP. Dots represent the mean values in each $D_{m,vol}$ bin, the gray lines the 25th and 75th percentile. The dots colours represent the number of observations used in each bin for calculating the median.

- Figure S2 was changed to modify the $D_{m,geo}$ in $D_{m,vol}$. The modifications include: i) modification of all points to use the new variable; ii) adjustment of axis label and subtitle.

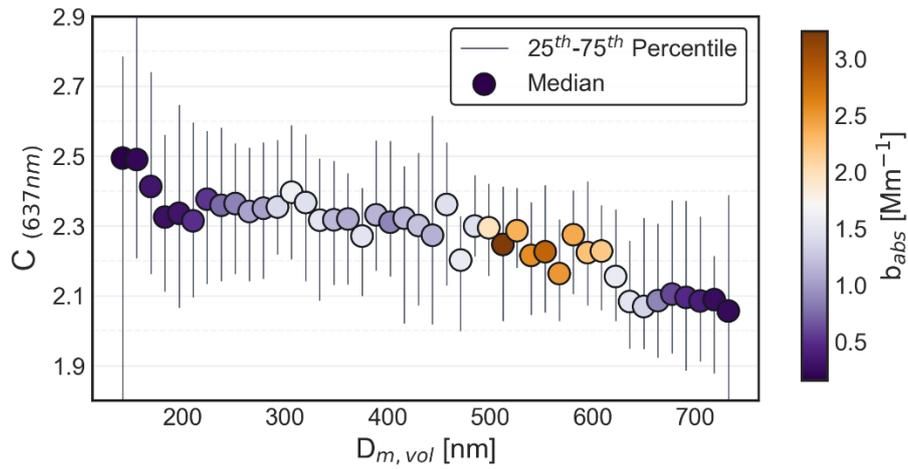


Figure S2 Cross-sensitivity of the C-size dependence on absorption coefficient measured by the MAAP. The color of the dots follows the variability of the median absorption coefficient in each size bin.

- Figure S3 was changed to modify the $D_{m,geo}$ in $D_{m,vol}$. The modifications include: i) modification of all points to use the new variable; ii) adjustment of axis label and subtitle.

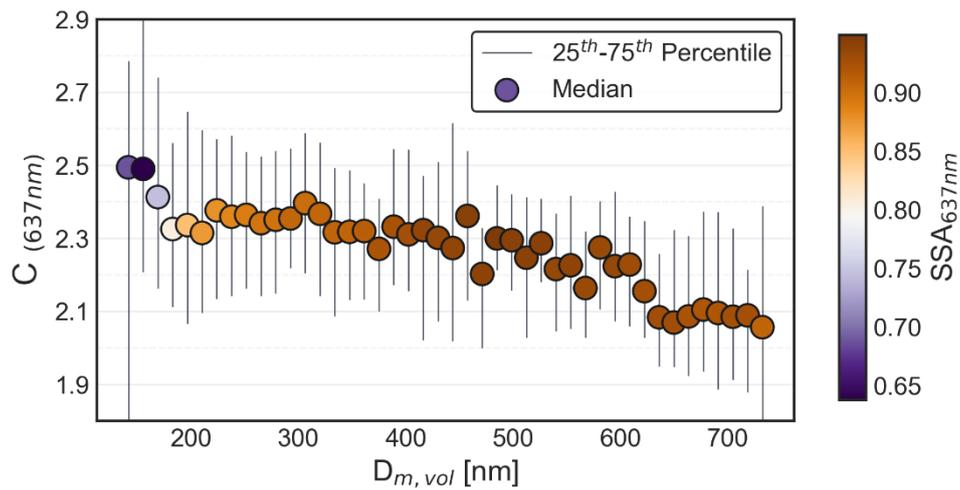


Figure S3 Cross-sensitivity of the C-size dependence on the SSA. The color of the dots follows the variability of the median SSA in each size bin.

- Table 3 was changed to modify the $D_{m,geo}$ in $D_{m,vol}$. The modifications include: i) modification of all $D_{m,vol}$ [nm] values; ii) adjustment of column name and legend.

Table 3: Average C values for each soot type (CS1-CS5) and associated particles properties. In particular, the average volume mean diameter ($D_{m,vol}$), absorption coefficient b_{abs} and single scattering albedo (SSA). These values were derived averaging the results obtained for all aethalometer filter spots corresponding to the same CS type. Values obtained in this study are shown together with the ratio between elemental and total carbon mass concentration (EC/TC) and the Absorption Ångstrom Exponent (AAE) derived in Heuser et al. (2024).

	C (630 nm) (this study)	$D_{m,vol}$ [nm] (this study)	b_{abs} [Mm^{-1}] (this study)	SSA (630 nm) (this study)	AAE (Heuser et al., 2024)	EC/TC (Heuser et al., 2024)
CS1	2.89 ± 0.03	351.5 ± 15.9	450 ± 30	0.08 ± 0.01	1.27 ± 0.13	0.79 ± 0.11
CS2	3.00 ± 0.03	291.0 ± 18.2	179 ± 11	0.09 ± 0.01	1.36 ± 0.21	0.73 ± 0.08
CS3	2.96 ± 0.03	339.9 ± 18.8	400 ± 30	0.09 ± 0.01	1.59 ± 0.22	0.67 ± 0.09
CS4	2.99 ± 0.03	273.2 ± 18.3	59 ± 4	0.08 ± 0.01	1.88 ± 0.31	0.53 ± 0.13
CS5	3.90 ± 0.06	181.2 ± 16.5	63 ± 6	0.29 ± 0.03	3.79 ± 0.33	0.00 ± 0.22

Answers of the authors to Reviewer#2

While the reviewer's comments are given in **black bold**, our answers are given below in [blue letters](#). Additionally, we added the changes made in the revised manuscript in *blue italic letters*.

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. Here we provide some major considerations. Both reviewers raised important points regarding the suitability of the filter-based instruments used as reference methods for ambient measurements. We have addressed this issue carefully by expanding the discussion in the results, incorporating recent literature, and revising comments related to the interpretation of our results. Additionally, both reviewers through different comments emphasised the need to clarify and deepen the analysis of the dependence of the multiple scattering correction factor (C) on particle size further, as this is a key aspect of the manuscript. They also recommended integrating the discussion of results obtained under controlled chamber conditions and in ambient more closely.

Accordingly, we have significantly revised and expanded in particular Sections 3.1.2 and 3.2.4 in the results, providing additional detail and context, and also improved Abstract, Introduction and Conclusion to better clarify the aim of the present study.

This manuscript addresses an important and timely topic regarding filter absorption photometers, which is of considerable interest to the atmospheric science community. The study is well contextualized, and the authors justify their results with relevant literature. The focus on the influence of particle size on the multiple scattering correction factor (C factor) is valuable. However, as mentioned in the conclusions, the study explores the broader role of aerosol properties on C variability, and not only particle size. If the intention were to limit the scope exclusively to particle size, the manuscript would require significant restructuring to address this aspect in more detail.

We thank the reviewer for their thoughtful comment and for recognizing the relevance of our work. As correctly pointed out, the primary focus of this study is the impact of particle size on the multiple scattering correction factor (C factor). However, the analysis is not limited to this variable. Hereafter, we provide a general author's consideration to this major reviewer comment.

The focus on diameter was firstly motivated by a generalized scarcity of works addressing the C factor relationship with particle diameter. Most of these works (e.g., Yus-Díez et al., 2025; Romsho et al., 2022 and Drinovec et al. 2022) have addressed this aspect mainly under controlled laboratory conditions. Although, the influence of variables such as SSA and wavelength on the C factor has been extensively investigated in previous studies, the present work explores these properties to quantify their influence on C factor relative to the size effect. This covariance of C factor with multiple properties results important and non-negligible in ambient uncontrolled observations.

The objective of the manuscript remains to identify the impact of size on C factor in controlled conditions and explore its importance in ambient and well mixed conditions given the natural variability of other properties. We have clarified this rationale and scope more explicitly in the revised version of the manuscript, together with more specific changes as described below.

The text in **Introduction** was modified to better explain our aim

... "Specifically, we examine its dependence on particle size, while also considering the effects of other properties such as SSA and wavelength to support and complement existing evidence from the literature. The chamber experiments were designed to explore a broader particle size range than in previous studies (Drinovec et al., 2022), taking advantage of controlled conditions to better isolate the role of size and evaluate its relative importance compared to other aerosol properties. These experiments were conducted using freshly generated soot and non-filter-based reference techniques, ensuring independence from filter-related artefacts.

Ambient observations, although subject to more variable and less controlled conditions, were used to assess whether the dependence on particle size can also be observed under real atmospheric conditions. This contributes additional context to previous work, where ambient investigations remain limited (e.g., Yus-Díez et al., 2025). "...

We have slightly extended the analysis to the specific case of coarse particle presence during dust events, in order to better illustrate its impact on C at the CMN site and at other locations potentially affected by Saharan dust transport. In particular, we added a figure in the Supplementary Information showing how C decreases during dust events of different intensities when coarse particles are present, and included a specific comment on this aspect in Section 3.2.4.

- Figure S7 was added to Supplementary Information to better show one of the implications of C dependence on size dependence, causing biases in our station: the significant decrease in C for increasingly strong dust events transporting coarse particles to the station.

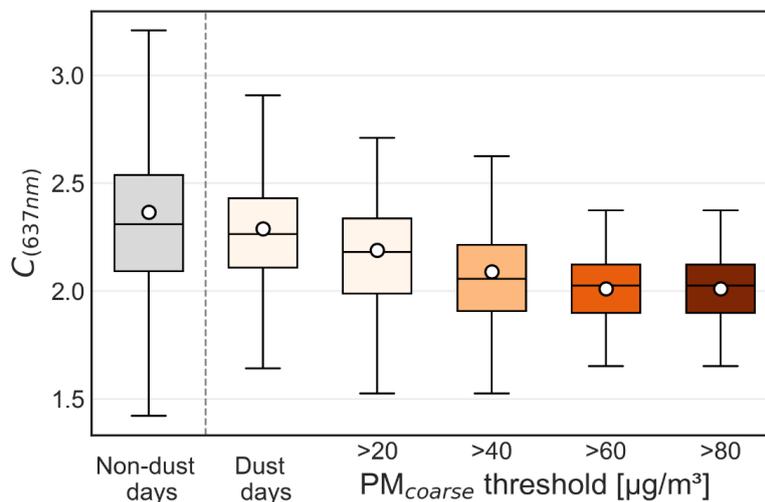


Figure S7: Boxplot comparing C values statistics during non-dust days and dust days of increasing intensity. The different intensities are represented by increasing threshold in the PM coarse fraction measured by an OPC during transport events. The identification of dust events at CMN is done according to the method detailed in Text S4 (Vogel et al., 2025) as well as the quantification of PM coarse.

- Text S4 was added to Supplementary Information to detail the method for dust days identification and the quantification of PM coarse.

...” Text S4 Identification of dust transport days

Dust transport days were identified following the method described in detail in Duchi et al. (2016), which combines in-situ measurements of the coarse particle concentration (particles with a diameter larger than 1 μm) and backward trajectories. The coarse particle concentration is obtained from measurements of an optical particle counter (Grimm® model 1.108). The data are averaged over 24h. On the time series of daily data, a multi-step smoothing is applied, and subtracted from the original time series to obtain the high frequency component. If a data point of the high frequency component exceeds the threshold value given as the 95th confidence interval, it is flagged as a potential dust transport day. For these potential dust transport days 7-day backward trajectories are taken and if they passed over the Saharan desert, the according days is finally flagged as a dust transport day.

The particulate matter concentration of the coarse particles (PM_{coarse}) is calculated from the coarse particle size distribution measured by the optical particle counter and a particle size dependent density (Wittmaack 2002). “...

Added references:

Wittmaack, K.: Advanced evaluation of size-differential distributions of aerosol particles, *Journal of Aerosol Science*, 33, 1009–1025, 2002.

Duchi, R., Cristofanelli, P., Landi, T. C., Arduini, J., Bonafe', U., Bourcier, L., Busetto, M., Calzolari, F., Marinoni, A., Putero, D., and Bonasoni, P.: Long-term (2002–2012) investigation of Saharan dust transport events at Mt. Cimone GAW global station, Italy (2165 m 360 a.s.l.), 4, 000 085, <https://doi.org/10.12952/journal.elementa.000085>, 2016.

- the text in **Section 3.2.4** was extended

...” To ensure that the observed variability in *C* was not primarily driven by other parameters—such as the SSA or exceptionally low absorption values—rather than by particle size itself, we examined the mean SSA and absorption coefficient within each size bin. The results show that neither of these parameters alone can account for the whole observed variability in *C* with size in Figure 5 (see Supplementary Information, Figure S3 and Figure S4).

Although its influence is moderate compared to the instrumental uncertainty, particle size appears to affect *C* even under ambient atmospheric conditions. This suggests that variations in particle size could contribute to the differences in *C* observed across sites and seasons, as proposed in previous studies (Bernardoni et al., 2021; Luoma et al., 2021), particularly when there are significant changes in the mean volume size distribution or during specific events. At CMN, this effect can be especially significant during dust transport events, which occur on about 20% of days throughout the year (Vogel et al., 2025). As shown in Supplementary Information Figure S7, *C* at 637 nm decreases with increasing dust event intensity, consistently with the findings of Yus-Díez et al. (2025).”...

Specific comments:

Abstract: The statement that *C* characterization is “poorly understood” is not entirely accurate. As the authors themselves note, there is a growing body of recent literature on this topic. The recent study by Yus-Díez et al. (2025), which provides a comprehensive analysis of *C* factor, should be considered for the interpretation of the present results.

We agree that several factors influence the multiple scattering correction factor (*C*) — such as the effects of single scattering albedo, filter loading, and wavelength — and have been extensively investigated in recent years, leading to a much better understanding of the *C* behaviour. Our intention in the abstract was to emphasize that, despite this progress, certain aspects of *C* variability, particularly those related to aerosol microphysical properties and their combined effects under ambient conditions, remain less well constrained.

To make this clearer, we have revised the abstract by rewording the original sentence to better reflect the current state of knowledge.

- The text in Abstract line 20:

...” Despite its widespread use, several aspects influencing the variability of *C*, particularly those related to aerosol microphysical properties, are still not fully constrained.” ...

We also expanded the discussion in Section 3.1.2, to better discuss the comparability of our results with recent studies such as Yus-Díez et al. (2025) and Drinovec et al. (2022).

- the text in **Section 3.1.2**

...“ This result is likely attributable to the greater penetration of smaller particles into the filter matrix (Huang et al., 2013; Hinds, 1999), that affect the multiple scattering effects between particles and filter, leading to a larger overestimation of true absorption by the aethalometer. This interpretation agrees with modelling results indicating that particle stratification increases the ratio of filter-based and real absorption as particle size decreases (Moteki et al., 2010). Our results are consistent with previous laboratory studies on other filter-based instruments such as COSMOS and PSAP (Lack et al., 2009; Nakayama et al., 2010), and two previous studies on the aethalometer (Drinovec et al., 2022; Yus-Díez et al., 2025). These studies show that for the AE33, *C* is larger for soot particles below ~250 nm than for particles above ~400 nm, while dust samples generally exhibit lower *C* values, in line with reduced artefact for larger particles.” ...

Use of MAAP as a reference instrument: The reliance on MAAP as a reference instrument is questionable. Yus-Díez et al. (2025) demonstrated that MAAP can overestimate absorption coefficients by ~47%, which directly affects the estimation of eBC and MAC. The authors should explicitly discuss this limitation and justify their approach, considering these known biases.

Both reviewers emphasized the importance of this aspect, which is critical for interpreting results obtained for atmospheric observations at CMN where the MAAP and the MWAA were used as reference instruments. To fully address this common point, we provide a more complete literature study and implications in the main text.

As noted, the reliability of filter-based instruments for measuring the absorption coefficient is debated in the literature. A recent study by Yus-Díez et al. (2025) shows that MAAP can overestimate absorption by up to 47% when compared to PTAAM-2 λ , particularly for aerosols with high single scattering albedo (SSA > 0.9). On the other hand, Heuser et al. (2025) reported an excellent comparison (less than 10% differences) when comparing filter-based techniques (MAAP, MWAA, PP-UniMi) with EMS for the highly absorbing CS particles from laboratory experiments in this study with $D_{m,vol}$ between 130 and 450 nm. Earlier work by Romshoo et al. (2022) reported good agreement between MAAP and the EMS technique for particles with a volume mean diameter $D_{m,vol} > 100$ nm, but MAAP slightly overestimated absorption (~20%) for smaller particles ($D_{m,vol} < 100$ nm). Similarly, Kalbermatter et al. (2022) demonstrated that MAAP tends to overestimate absorption measured with other non filter-based techniques by at least 20%, especially for particles with a high total mass-to-BC mass ratio. Finally, Moteki et al. (2010) highlighted that the two-stream model on which the MAAP is based may underestimate its size dependent bias, when stratification occurs in the filter, in particular for particles smaller than 200 nm.

Together, these studies indicate that filter-based reference instruments such as MAAP and MWAA may be subject to systematic biases, but the magnitude of these biases depends on particle properties and the reference technique used, being larger for: high SSA (> 0.9), small particle diameters (< 200 nm) and BC particles mixed with secondary organic materials.

Although further investigation is needed, using these references in our study could lead to a possible general underestimation of the C factor in ambient measurements. If PTAAM-2 λ or photoacoustic techniques are considered as more accurate references, the underestimation could reach up to 47%, particularly for high SSA and mixed particles. This affects not only the absolute C values but also the observed dependencies of C on SSA and particle size. At CMN, the C dependencies we report may therefore be underestimated for SSA > 0.9 and diameters < 200 nm, while estimates for SSA < 0.9 and diameters > 200 nm are expected to be more reliable.

To better include this discussion, we extended the text in **Section 3.2.1, 3.2.3 and 3.2.4**. In particular, we added these paragraphs:

- the text in **Section 3.2.1**

... "The lower ambient values may be partly due to the different reference techniques used. The MAAP can exhibit a positive bias in absorption when compared with non-filter-based reference techniques as the EMS, particularly at relatively high SSA (>0.9), small particles (diameter <200 nm) and for aged aerosol (Yus-Díez et al., 2025; Kalbermatter et al., 2022; Romshoo et al., 2022). Such bias would lead to higher MAAP absorption values and, consequently, to lower C values in ambient measurements at CMN compared to those obtained in CESAM."...

- the text in **Section 3.2.3**

... "Moreover, we should consider that this dependence, estimated using the MAAP as a reference, may have been underestimated. As shown by Yus-Díez et al. (2025), this instrument is also affected by cross-sensitivity to scattering when compared with a non-filter-based technique."...

- the text in **Section 3.2.4**

... "When comparing the size dependence of C with that observed in the chamber experiments and in other laboratory studies on soot (Drinovec et al., 2022; Yus-Díez et al., 2025), the ambient relationship appears more linear and overall weaker. This attenuation may be partly related to the use of the MAAP. Its positive bias compared to non filter-based techniques could be more relevant for smaller particles (Moteki et al., 2010; Romshoo et al., 2022). As a result, the influence of particle size on C under ambient conditions could be partially masked by the measurement bias inherent to the reference technique used."...

Added reference:

Romshoo, B., Pöhlker, M., Wiedensohler, A., Pfeifer, S., Saturno, J., Nowak, A., Ciupek, K., Quincey, P., Vasilatou, K., Ess, M. N., Gini, M., Eleftheriadis, K., Robins, C., Gaie-Levrel, F., and Müller, T.: Importance of size representation and

morphology in modelling optical properties of black carbon: comparison between laboratory measurements and model simulations, Atmospheric Measurement Techniques, 15, 6965–6989, <https://doi.org/10.5194/amt-15-6965-2022>, 2022.

Lines 231–236: This section is currently confusing for the reader. It should be rewritten with improved clarity to better explain the intended message.

We thank the reviewer for pointing out that the description of this section was unclear. The text has been rewritten to improve readability and to clarify the steps used to derive C both for the MAAP-based and MWAA-based analysis.

- The text in Section 2.2.2 Reference absorption coefficient at CMN: filter-based MAAP and MWAA was revised in lines 231–239 to improve clarity:

... "The absorption coefficient measured by the MAAP at 637 nm ($b_{abs,ref}$) was averaged over the same 1-h intervals as the AE33 data and used to derive the C factor at 637 nm according to Eq. (1). For consistency in wavelength, before calculating the C factor, the AE33 attenuation coefficient ($b_{am,LC}$) measured at 660 nm was interpolated to 637 nm using the Absorption Ångström Exponent (AAE) obtained from the power-law fit of $b_{am,LC}(\lambda)$ versus wavelength.

The spectral dependence of C was analysed using MWAA measurements. In this case, the reference absorption coefficients $b_{abs,ref}(\lambda)$ were interpolated to the AE33 wavelengths. C values were then calculated at the seven AE33 wavelengths by averaging the AE33 $b_{am,LC}(\lambda)$ data over the 24 h corresponding to each MWAA filter sample." ...

The study presents a detailed analysis of C factor characterization, but the conclusions could be clearer in showing why these findings matter. In particular, the authors should expand on the broader relevance of their work:

Since Aethalometers are widely used in monitoring networks, what do these results mean for improving data quality and ensuring comparability across sites?

Do the results provide recommendations to narrow the variability in C factor estimates, and how generalizable are they across instruments and locations?

The discussion could better balance the value of controlled chamber experiments with the ambient measurements, which include complex, mixed-source aerosols. Since monitoring is done under ambient conditions, it would be important to highlight how representative and applicable the results are in these real-world settings.

We thank the reviewer for these constructive comments. We fully agree that the discussion and conclusions should better emphasize the broader implications of our findings, the link between controlled and ambient measurements, and the representativeness of the results for real-world conditions.

Our results confirm that the dependence of the Aethalometer C factor on particle size—previously indicated by a few chamber studies—is also observable under ambient conditions. The chamber experiments clearly demonstrate this dependence under controlled conditions. Importantly, the ambient observations, also show a detectable size dependence that cannot be explained solely by other known dependencies. This indicates that the effect identified in chamber studies can indeed influence ambient Aethalometer measurements and may contribute to the variability observed in this parameter across monitoring sites and seasons.

While we recognize the potential implications of these findings for improving data comparability, we consider that a single-site study is not sufficient to define quantitative correction recommendations or generalized parameterizations. Instead, our aim is to draw attention to the relevance of this size effect in real-world conditions and to demonstrate that it can have a measurable and significant impact—particularly in environments dominated by small particles (e.g., nucleation or Aitken mode) or influenced by dust transport that increases the coarse fraction. The studied site, a mountain location characterized by particles mainly in the fine mode, and frequent dust intrusions, provides conditions potentially representative of other elevated or dust-affected environments.

Finally, the revised discussion clarifies the complementary role of chamber and ambient experiments: the chamber setup provides ideal conditions to isolate the size effect and establish its magnitude, whereas the ambient observations show that this dependence persists under complex, mixed-source aerosol conditions relevant to monitoring networks. Further coordinated studies across different sites and aerosol regimes, ideally including non-filter-based reference instruments,

will be essential to quantify the robustness and generality of this dependence and to ultimately support the harmonization of Aethalometer data quality across networks.

These aspects have been integrated throughout the revised manuscript, in the Abstract, Introduction, and Conclusions, to clarify the relevance, applicability, and implications of our findings.

- Text in **Abstract**

...” These results are relevant for improving the accuracy and comparability of aerosol absorption measurements performed by aethalometers, particularly within monitoring networks. The observed dependence of C on particle size suggests that differences between urban and remote sites, or between periods dominated by nucleation versus coarse-mode particles (e.g., dust), may contribute to the variability reported across locations. Understanding these dependencies is essential for refining correction approaches and reducing inter-site discrepancies in network datasets.”...

- Text in **Introduction**

...” Given the widespread use of aethalometers in long-term observational networks, constraining the variability of the C factor is crucial for ensuring the quality and comparability of absorption data across sites. Differences in aerosol size distributions and mixing states—such as between freshly emitted soot in urban areas and aged or coarse particles at remote stations—can introduce systematic biases if a single, constant C is applied. A more robust understanding of these dependencies is therefore required to guide the harmonization of network measurements and to support the development of improved parameterizations for data correction.”...

- Text in **Conclusions**

...” The present analysis evidences a dependence of C on particle size. This dependence was moderate (within 25%) under ambient conditions dominated by mostly scattering aerosols, while it became much more pronounced (>60%) in chamber experiments using soot-only particles and a non-filter-based absorption reference. This behaviour likely contributes to the variability of C values reported in the literature and highlights the need for further investigation to improve the accuracy of absorption coefficient measurements within international research infrastructures.

The consistency between chamber and ambient results indicates that, although the chamber setup better isolates the size dependence, this effect is also detectable under complex real-world aerosol conditions. These findings suggest that C may vary systematically across monitoring sites depending on the dominant particle size modes—for example, smaller C values at sites dominated by larger, aged particles, and higher C values at locations influenced by fresh combustion or nucleation events. Further studies, combining targeted chamber experiments and modelling under a wider range of conditions, are needed to better quantify these dependencies and support the development of more generalizable parameterizations for harmonized absorption measurements and improved data quality in monitoring networks.”...