

# The role of size in the multiple scattering correction C for dual-spot aethalometer: a field and laboratory investigation

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**Abstract.** The dual-spot aethalometer AE33 is a widely used instrument for measuring the aerosol absorption coefficient, but the accuracy of its measurements is heavily dependent on the multiple scattering correction factor (C), which compensates for multiple scattering effects in the filter matrix. Despite its widespread use, several aspects influencing the variability of C, particularly those related to aerosol microphysical properties, are still not fully constrained.

In this work, we explore the variability of C for the AE33 in a wide range of conditions and aerosol properties by combining chamber experiments with freshly emitted laboratory-generated soot and ambient data from a mountaintop site in Italy (Monte Cimone, CMN). The C factor is derived by comparison with independent filter-based instruments such as the MAAP (Multi-Angle Absorption Photometer) and MWAA (Multi-Wavelength Absorption Analyzer) at CMN and the extinction-minus-scattering (EMS) approach in chamber experiments.

The mean C value at a wavelength of 637 nm derived at CMN is 2.35 with a standard deviation of 0.58, while the average values obtained in chamber experiments in different conditions range from  $2.89 \pm 0.03$  to  $3.9 \pm 0.06$ . The variability of C at CMN appears to be primarily influenced by the signal-to-noise ratio of the instruments, especially during the colder months when absorption coefficient values fall below  $1 \text{ Mm}^{-1}$ . In contrast, in the chamber experiments, the variability is mainly driven by particle properties. The C value at 637 nm, derived from measurements at CMN, increases with increasing single scattering albedo (SSA), particularly for SSA values above 0.94, while showing no statistically significant spectral variability. 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. 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.

42 Understanding these dependencies is essential for refining correction approaches and reducing inter-site discrepancies in  
43 network datasets.

## 44 **1 Introduction**

45 Absorbing aerosols have a unique and important role in the Earth's climate system (Bond et al., 2013; Liu et al., 2020a).  
46 By absorbing solar and terrestrial infrared radiation, aerosols exert an influence on low tropospheric temperature, liquid  
47 and ice cloud distribution and properties, atmospheric dynamics, as well as surface reflectivity, in particular over snow  
48 and ice surfaces (Ramanathan and Carmichael, 2008; Liu et al., 2020a; Li et al., 2022; Kok et al., 2023). This impact is  
49 especially significant for strongly absorbing species such as black carbon (BC), brown carbon (BrC), and mineral dust.  
50 These components account for the majority of aerosol light absorption and contribute substantially to the direct radiative  
51 effect at both regional and global scales (e.g. Chung et al., 2012; Kok et al., 2023; Sand et al., 2021; Zhu et al., 2021).

52 The strength of this radiative effect is typically characterized by the aerosol absorption coefficient, expressed in inverse  
53 mega meters ( $b_{\text{abs}}$ ,  $\text{Mm}^{-1}$ ) that represents the amount of light absorbed by aerosol per unit volume. The  $b_{\text{abs}}$  is commonly  
54 measured using filter-based techniques, differential extinction-minus-scattering (EMS) methods, or photoacoustic  
55 instruments. Despite its importance, aerosol absorption remains poorly constrained. This is primarily due to the lack of  
56 standardized measurement approaches and the presence of instrument-specific biases (Petzold et al., 2013), which  
57 introduce substantial uncertainties in  $b_{\text{abs}}$  data. In consequence, a major difficulty lies in capturing the magnitude and  
58 spatio-temporal variability of the spectral mass absorption cross section (MAC,  $\text{m}^2\text{g}^{-1}$ ), i.e. the absorption coefficient per  
59 unit of absorbing specie mass concentration ( $\mu\text{g m}^{-3}$ ), or the single scattering albedo (SSA), representing the fraction of  
60 scattered radiation with respect to extinction. The MAC and SSA depend on a wide variety of variables such as the  
61 composition (via the complex refractive index, CRI), size, morphology and mixing state of the particles (Liu et al., 2020b;  
62 Moteki, 2023). Understanding these dependencies is required for a better representation of absorbing aerosols in climate  
63 models and remote sensing algorithms (e.g., Samset et al., 2018). In this regard, improving the accuracy of in situ  
64 measurements of the spectral aerosol absorption coefficient  $b_{\text{abs}}$  is fundamental for contributing to robust CRI, MAC and  
65 SSA evaluation.

66 The Aethalometer (Hansen et al., 1984; Drinovec et al., 2015) is the most used instrument for routine measurements of  
67 the aerosol absorption coefficient in the GAW (Global Atmospheric Watch) and ACTRIS-RI (Aerosol, Clouds, and Trace  
68 gases Research Infrastructure; <https://www.actris.eu/>) observational networks (e.g., Laj et al., 2020; Savadkoochi et al.,  
69 2023; Rovira et al., 2025), with a long-record of applications in ground-based and airborne intensive field campaigns (e.g.  
70 Fialho et al., 2005; Sandradewi et al., 2008; Formenti et al., 2011; Di Biagio et al., 2016; Favez et al., 2021; Ohata et al.,  
71 2021) and laboratory experiments (e.g. Weingartner et al., 2003; Baldo et al., 2023; Di Biagio et al., 2019; Kalbermatter  
72 et al., 2022).

73 The aethalometer is a filter-based photometer. It measures the light attenuation (ATN) through a filter on which the  
74 aerosol is continuously collected at a constant flow rate. The volume absorption coefficient is estimated via the Beer-  
75 Bouguer-Lambert law based on the ATN rate of change over the time interval  $\Delta t$  (typically 1–2 minutes). This rate is  
76 proportional to the absorbing aerosol concentration and it is also used to calculate an optically-equivalent BC mass  
77 concentration (eBC,  $\mu\text{g m}^{-3}$ ) using prescribed instrumental MAC spectral values (Petzold et al., 2013). The aethalometer  
78 measurement technique has the advantage of being sensitive to low aerosol signals even for relatively low integration  
79 times (e.g., Hansen et al., 1984) and so to be adapted to measurements in environments with variable aerosol loadings.

80 The aethalometer is also easily deployable and provides  $b_{\text{abs}}$  at seven wavelengths covering the broad range from 370 to  
81 950 nm.

82 Despite its operational advantages, the aethalometer measurement is affected by environmental and aerosol-related  
83 artefacts that can seriously bias the retrieved absorption coefficient (Arnott et al., 2005; Schmid et al., 2006; Virkkula et  
84 al., 2015; Weingartner et al., 2003, hereafter referred as W2003; Collaud Coen et al., 2010, hereafter referred as C2010).  
85 These are linked to three causes: 1. the accumulation of particles on the filter that reduces the linearity in the detected  
86 attenuation signal, therefore causing an underestimate of the absorption coefficient for increasing aerosol deposition (the  
87 loading effect); 2. the scattering from particles deposited on the filter that contributes to detected attenuation and that  
88 causes an overestimation of the absorption (the scattering effect); 3. the multiple scattering by the filter fibers causing an  
89 increased optical path and absorption overestimation (the multiple scattering effect).

90 Aethalometer models commercialized by Magee Scientific have evolved over time to address the known measurement  
91 artefacts. Earlier models, such as the AE31, did not include any automatic correction for these effects. Instead, post-  
92 processing algorithms and empirical correction factors—extensively studied and developed in the literature—were  
93 required to compensate for biases such as loading, scattering, and multiple scattering (Weingartner et al., 2003; Arnott et  
94 al., 2006; Bond and Bergstrom, 2006; Collaud Coen et al., 2010; Segura et al., 2014; Backman et al., 2017; Di Biagio et  
95 al., 2017; Saturno et al., 2017).

96 The more recent AE33 model, includes a dual-spot system that automatically compensates for the loading effect in real  
97 time (Drinovec et al., 2015). However, even in newer models, the correction for scattering and multiple scattering still  
98 relies on the application of a scaling factor, typically referred to as  $C$  or  $C_{\text{ref}}$ . As of today, estimations of the  $C$  from various  
99 field and laboratory studies have been performed, investigating aerosols of different types and properties and varying  
100 ambient conditions (Corbin et al., 2018; Kim et al., 2019; Laing et al., 2020; Valentini et al., 2020; Moschos et al., 2021;  
101 Yus-Díez et al., 2021; Bernardoni et al., 2021; Kalbermatter et al., 2022). These studies have reported a wide range of  $C$   
102 values, from about 2.26 to 8.26, which translates into significant uncertainty, as  $b_{\text{abs}}$  is inversely proportional to this factor.  
103 This variability remains one of the major challenges in standardizing and comparing absorption measurements across  
104 different studies and networks.

105 As of today, there is still no consensus on the appropriate value of  $C$  or its dependence on specific aerosol properties or  
106 experimental conditions. The influence of parameters such as particle size, chemical composition, and spectral wavelength  
107 on  $C$  remains debated. Particle size can lead to bias in the measurement of the absorption coefficient carried out with  
108 filter-based instruments (Lack et al., 2009; Nakayama et al., 2010). This dependence is related to the different depth of  
109 penetration of particles of different sizes into the filter, as well as to the characteristics of the filter (Hinds, 1999; Huang  
110 et al., 2013; Berry et al., 2023). For the aethalometer, particle size has been suggested as a factor that could contribute to  
111 the variability of  $C$  values across different locations and seasons (Bernardoni et al., 2021, Luoma et al., 2021), although  
112 direct observational evidence for this dependence in the aethalometer AE33 remains limited (Drinovec et al., 2022;  
113 Romshoo et al., 2022; Yus-Díez et al., 2025). Regarding the composition, previous studies have demonstrated that the  
114 optical properties of sampled particles can directly influence aethalometer measurements by altering the amount of  
115 scattered light, which the instrument may incorrectly interpret as absorption. An increase in the SSA has been linked in  
116 these studies to a corresponding rise in the  $C$  values (Schmid et al., 2006; Backman et al., 2017; Yus-Díez et al., 2021).  
117 Moreover, very few studies have focused on  $C$  estimations for aerosol populations with high fractions of black and brown  
118 carbon (BC and BrC), such as those found near combustion sources or in laboratory studies on combustion aerosols.

119 This uncertainty has practical implications, particularly for international observation networks like ACTRIS RI, which  
120 currently recommend the use of a single, wavelength-independent  $C$  value. The objective of this study is to improve the  
121 characterization of the  $C$  parameter for the AE33 aethalometer model, currently the most widely used.

122 Specifically, we examine its dependence on particle size, while also considering the effects of other properties such as  
123 SSA and wavelength to support and complement existing evidence from the literature. The chamber experiments were  
124 designed to explore a broader particle size range than in previous studies (Drinovec et al., 2022), taking advantage of  
125 controlled conditions to better isolate the role of size and evaluate its relative importance compared to other aerosol  
126 properties. These experiments were conducted using freshly generated soot and non-filter-based reference techniques,  
127 ensuring independence from filter-related artifacts. Ambient observations, although subject to more variable and less  
128 controlled conditions, were used to assess whether the dependence on particle size can also be observed under real  
129 atmospheric conditions. This contributes additional context to previous work, where ambient investigations remain  
130 limited (e.g., Yus-Díez et al., 2025).

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

## 137 **2 Methods**

138 The  $C$  dependencies were investigated through the synergic use of both chamber experiments and observational long term  
139 data series. While ambient measurements are valuable for investigating the spatial and temporal variability of  $C$  in the  
140 real atmosphere, chamber experiments provide a more controlled setting, where variability of multiple variables can be  
141 minimized, enabling a clearer understanding of how the  $C$ -factor depends on specific particle properties.

142 This section provides description of the experimental setup for field and chamber experiments and description of  $C$  factor  
143 calculations. A summary of the instruments considered in this study, including deployment details and availability, and  
144 measurement uncertainties is provided in Table 1 and Table 2. All volumetric quantities used in the analysis were  
145 converted to standard temperature and pressure conditions (STP), assuming 1013.25 mbar and 273.15 K.

### 146 **2.1 Experimental set-up**

#### 147 **2.1.1 Field measurements at Mt Cimone**

148 Field measurements were carried out at the mountain site of Monte Cimone (CMN, Italy, 44°11'N-10°42'E, 2165 m  
149 a.s.l.), located in northern Italy near the Po Valley. The CMN observatory is situated on the summit of the highest  
150 mountain in the northern Apennines, and its orographic features allow air masses to arrive from all directions, making it  
151 representative of a relatively pristine atmosphere. However, particularly during summer and midday hours, it may be  
152 subject to the uplift of pollutants due to planetary boundary layer (PBL) rise and valley breezes (Marinoni et al., 2008;  
153 Cristofanelli et al., 2009; Andrews et al., 2011; Cristofanelli et al., 2018). The site is also subject to Saharan dust intrusions  
154 (Vogel et al., 2025) and the long-range transport of wildfire emissions throughout the year (Cristofanelli et al., 2009,  
155 2024). These diverse conditions affect aerosol properties, including particle size (e.g., following dust transport or new

156 particle formation events; Vogel et al. 2025) as well as their optical and chemical characteristics, influenced by sources  
157 such as the Po Valley, wildfires, and dust.

158 The CMN station is part of the World Meteorological Organization's (WMO) Global Atmosphere Watch (GAW) program  
159 and high-quality routine measurements of major aerosol properties are conducted in accordance with the ACTRIS-RI  
160 guidelines. This study presents measurements collected between July 2020 and November 2022. Aerosols were sampled  
161 through a TSP (Total Suspended Particles) inlet, specifically designed for high mountain stations.

162 The spectral absorption coefficient was measured using a dual-spot AE33 aethalometer (RH < 40% ensured by a Nafion®  
163 dryer), and complemented by reference absorption techniques: the Multi-Angle Absorption Photometer (MAAP, online,  
164 637 nm, Thermo Sci.; Petzold and Schönlinner, 2004) and the Multi-Wavelength Absorption Analyzer (MWAA, offline,  
165 5 wavelengths, 375-405-532-635-870 nm, Massabò et al., 2015). The scattering coefficient was measured with an  
166 integrating nephelometer (TSI Inc., model 3563, Anderson et al., 1996; Massoli et al., 2009).

### 167 **2.1.2 Simulation experiments in the CESAM chamber**

168 Chamber experiments were performed in controlled conditions in the 4.2 m<sup>3</sup> stainless-steel multi-instrumented CESAM  
169 chamber (French acronym for Experimental Multiphase Atmospheric Simulation Chamber, <https://cesam.cnrs.fr/>; Wang  
170 et al., 2011), which has been extensively used in recent years to simulate the formation and aging of different types of  
171 aerosols and investigate their physico-chemical and spectral optical properties (Denjean et al., 2015; Di Biagio et al.,  
172 2017, 2019; De Haan et al., 2020; Baldo et al., 2023).

173 The experiments were performed during three campaigns occurred in May 2021, October 2021 and December 2022 and  
174 involved primary combustion BC- and BrC-containing soot aerosols. Full detailed description of the setup and data  
175 analysis for those experiments is provided in Heuser et al. (2025).

176 Soot aerosols with varying BC and BrC content were generated by a computer-controlled miniCAST burner (model  
177 6204C, Jing Ltd., Switzerland). The miniCAST is a co-flow diffusion flame generator. It produces soot aerosols (hereafter  
178 identified as cast soot, CS) by burning a mixture of propane (C<sub>3</sub>H<sub>8</sub>), N<sub>2</sub> and oxidation air. Varying the proportions of these  
179 gases allows to vary the size, composition and morphology of the generated CS particles (e.g. Moore et al., 2014; Bescond  
180 et al., 2016). In this study five different miniCAST operation points were investigated, corresponding to four fuel-lean  
181 burning conditions (CS1–CS4) and one fuel-rich condition (CS5) (Table S1). The CS aerosol particles were injected in  
182 CESAM via a charcoal denuder to remove volatile organic compounds possibly present in the miniCAST exhaust. After  
183 injection in the chamber, the CS was left in suspension to age under different controlled conditions. In this work we only  
184 consider experiments in which CS is aged under dry conditions for aging times between 2 and 6 hours.

185 Between experiments, the chamber was mechanically evacuated and pumped to 3·10<sup>-4</sup> mbar for at least a few hours and  
186 usually over a full nighttime period. In addition to overnight evacuation, the chamber was manually cleaned at the  
187 beginning of each campaign. Background concentrations of aerosols in the chamber were usually less than 0.05 µg m<sup>-3</sup>  
188 (that is nearly 10<sup>4</sup> times less than the injected CS mass concentration in the chamber reaching up to 60 to 95 µg m<sup>-3</sup>). The  
189 different experimental conditions for chamber experiments are summarized in Table S1 and S2. For the majority of  
190 experiments several repetitions are performed to test the reproducibility of results.

191 The spectral absorption coefficient was measured in CESAM by means of a dual spot aethalometer AE33, complemented  
192 by the extinction-minus-scattering (EMS) approach (Onasch et al., 2015; Modini et al., 2021) as reference absorption  
193 technique. The extinction coefficient was measured by means of a CAPS PMex instrument (Aerodyne, Massoli et al.,

194 2010) and the scattering coefficient by an integrating nephelometer (TSI Inc., model 3563, Anderson et al., 1996; Massoli  
195 et al., 2009).

## 196 2.2 Quantification of C factor for the dual spot aethalometer

197 The aethalometer measures the attenuation coefficient  $b_{atn}(\lambda)$  which is proportional to the light attenuation (ATN) rate of  
198 change over time. The multiple scattering correction factor C is defined as:

$$199 \quad C(\lambda) = \frac{b_{atn,LC}(\lambda)}{b_{abs,ref}(\lambda)} \quad (1)$$

200 where  $b_{atn,LC}$  is the attenuation coefficient measured by the aethalometer and corrected for the loading effect based on the  
201 AE33 automatic internal algorithm, described in the next section, and  $b_{abs,ref}$  is the reference absorption coefficient.

### 202 2.2.1 Dual-spot aethalometer loading correction

203 The model AE33 automatically corrects for the loading effect, through an internal algorithm, by measuring the change in  
204 light attenuation in two filter spots, crossed by different air flows, therefore with different aerosol loads. The ATN  
205 measurements from the two spots are combined through a set of equations detailed in Drinovec et al. (2015) to provide  
206 as output the  $eBC_{AE33}$  based on the following formulation:

$$207 \quad eBC_{AE33}(\lambda) = \frac{S \cdot \left( \frac{\Delta ATN_1(\lambda)}{100} \right)}{F_1(1 - \zeta) \cdot MAC_{AE33}(\lambda) \cdot C_{instr} \cdot (1 - k(\lambda) \cdot ATN_1(\lambda)) \cdot \Delta t} \quad (2)$$

208 where S is the spot surface area ( $m^2$ ),  $\Delta ATN_1$  is the variation of attenuation through the filter spot1 (that is the one with  
209 the highest flow rate) in the time interval  $\Delta t$ ,  $F_1$  is the flow rate through spot1 ( $m^3 \text{min}^{-1}$ ),  $\zeta$  is the flow leakage (taking into  
210 account lateral flow in the filter matrix),  $MAC_{AE33}(\lambda)$  is the instrumental mass absorption cross section (18.47, 14.54,  
211 13.14, 11.58, 10.35, 7.77, 7.19  $m^2 \text{g}^{-1}$  at 370, 470, 520, 590, 660, 880, 950 nm, respectively), and  $k(\lambda)$  is a corrective  
212 parameter derived by the instrument algorithm to automatically account for the loading effect. As indicated in Eq. (2) the  
213 algorithm also accounts for an instrumental multi scattering correction  $C_{instr,AE33}$  (wavelength-independent) for  
214 harmonization among aethalometers using different tape materials (quartz, TFE-coated glass; Drinovec et al., 2015). The  
215  $C_{instr,AE33}$  value depends on the filter material and is 1.39 for the for the M8060 filter tape used in this study. Starting from  
216 Eq. (2) the spectral loading-corrected attenuation coefficient  $b_{atn,LC}(\lambda)$  is derived from  $eBC_{AE33}$  data by multiplying for the  
217 default instrumental  $C_{instr,AE33}$  and the  $MAC_{AE33}(\lambda)$  as:

$$218 \quad b_{atn,LC,AE33}(\lambda) = eBC_{AE33}(\lambda) \cdot MAC_{AE33}(\lambda) \cdot C_{instr,AE33} \quad (3)$$

219 As previously noted by Cuesta-Mosquera et al. (2021), the automatic AE33 loading correction method has limitations,  
220 particularly around filter changes, potentially introducing biases. These limitations are particularly critical when  
221 absorbing aerosol concentration is high, filter changes frequent (tens of minutes-few hours), and experiment durations  
222 relatively short. At CMN, where the concentration of absorbing species is typically low ( $0.2 \mu\text{g m}^{-3}$ ), the filter changes  
223 occur after several hours or days. Conversely, for chamber experiments, the higher soot concentration leads to frequent  
224 filter changes (tens of minutes-few hours). In the present analysis, both for field measurements and chamber experiments,  
225 the internal correction automatically applied by the instrument was utilized. For field measurements, the  $b_{atn,LC}(\lambda)$  values  
226 obtained at 1-minute resolution from the  $eBC(\lambda)$  values provided by the instrument were averaged over one hour.  
227 Conversely chamber data, for which experiment duration is limited, were analysed at 1-min resolution.

228 A detailed discussion on the effect of high soot concentrations on the loading correction for the AE33  $b_{\text{atn,LC}}(\lambda)$  calculation  
229 and C evaluation for chamber experiments is provided in the Supplementary Information (Text S1 and S2).

### 230 **2.2.2 Reference absorption coefficient at CMN: filter-based MAAP and MWAA**

231 The MAAP and the MWAA are the filter-based reference instruments used at CMN. The MAAP is based on the  
232 simultaneous measure of both the reflected and the transmitted light of aerosols embedded in a filter. An algorithm based  
233 on the Mie theory and the two stream approximation is then used to convert these signals in values of  $SSA_{PF}$  and optical  
234 thickness ( $\tau_{PF} = \ln(T_{PF})$ ) of the particle–filter (PF) system. These parameters allow to retrieve the aerosol absorption  
235 coefficient using the equation:

$$236 \quad b_{\text{abs,ref}} = \frac{S}{V} ABS = -\frac{S}{V} (1 - SSA_{PF}) \ln(T_{PF}) \quad (4)$$

237 where S is the spot surface area ( $\text{m}^2$ ) of collection and V the sampled volume of air ( $\text{m}^3$ ). The basic principle of the  
238 MWAA technique is based on the MAAP concept. However, while the MAAP is a field–deployable instrument working  
239 at a single wavelength (637 nm), the MWAA is a laboratory–based setup that extends the principle of the MAAP to 5  
240 wavelengths and analyze filter aerosol samples. Because of their different nature, the MAAP is a high resolution  
241 instrument (with observations typically every minute) while the MWAA integrates observations across an aerosol  
242 sampling period that can be variable depending on the concentrations (minutes to hours).

243 The MAAP is routinely measuring at the CMN site collecting data of  $b_{\text{abs,ref}}$  at 637 nm at 1-min time resolution. Data  
244 measured in the period from July 2020 to November 2022 are considered in this study. The MWAA technique was instead  
245 used to measure the  $b_{\text{abs,ref}}$  at 5 different wavelengths on 43 samples collected every 24 hours by the MAAP at CMN in  
246 the period September–October 2022.

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

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

### 255 **2.2.3 Reference absorption coefficient at CESAM: extinction-minus-scattering approach**

256 The EMS approach is considered as reference absorption technique for chamber experiments. This method involves the  
257 simultaneous measurements of both the extinction ( $b_{\text{ext}}$ ) and scattering coefficient ( $b_{\text{sca}}$ ) and the absorption coefficient is  
258 obtained as the difference of the two. When applied in the field to predominantly scattering samples, this technique is  
259 affected by high level of uncertainty as discussed in Modini et al. (2021). However, it can be successfully used as a good  
260 standard in laboratory experiments with high absorption signals. Validation of the methodology is provided in Heuser et  
261 al. (2025). In this study the extinction coefficient at 630 nm was measured by means of a CAPS PMex instrument  
262 (Aerodyne, Massoli et al., 2010). Extinction coefficient measured from the CAPS PMex does not require further

263 adjustments. The scattering coefficient at 450, 550, and 700 nm was measured by an integrating nephelometer (TSI Inc.,  
264 model 3563, Anderson et al., 1996). The nephelometer  $b_{sca}$  needs to be corrected for truncation, in order to compensate  
265 for the limited field of view of the instrument. The truncation correction was calculated using the Massoli et al. (2009)  
266 approach adapted to highly absorbing aerosols as those probed in chamber experiments, as discussed in Heuser et al.  
267 (2025). Assumptions on the real refractive index are required in the Massoli et al. (2009) calculation. For the different  
268 experiments considered in this study the real index was set at  $n=1.9$ , based on the proposed CRI for fresh BC by Bond  
269 and Bergstrom (2006). A sensitivity test was performed to investigate the potential error caused by this assumption.  
270 Changes in the corrected scattering signal for a range of likely real parts was found to be 4%.

271 The truncation-corrected  $b_{sca}$  values were interpolated at 630 nm based on the Scattering Ångström Exponent (SAE)  
272 calculated as the power-law fit of  $b_{sca}$  vs the wavelength ( $b_{sca} \sim \lambda^{-SAE}$ ). The  $b_{abs}$  at 630 nm was calculated at 1-min resolution  
273 as the difference of the CAPS-PMex  $b_{ext}$  and the nephelometer  $b_{sca}$ . The C values at 630 nm were derived at 1-minute  
274 resolution and then averaged over each aethalometer filter spot.

## 275 **2.3 Ancillary measurements**

### 276 **2.3.1 Single scattering albedo (SSA)**

277 The aerosol SSA was calculated at 637 nm and 1-hour resolution at CMN and at 630 nm at 1-minute resolution for  
278 chamber experiments, as:

$$279 \quad SSA = \frac{b_{sca}}{b_{ext}} \quad (5)$$

280 The  $b_{sca}$  was measured at 450 nm, 550 nm, and 700 nm by the TSI nephelometer both in field and in the chamber. At  
281 CMN the nephelometer data were corrected for truncation effects following the method of Anderson and Ogren (1998)  
282 and extrapolated at 637 nm as ( $b_{sca} \sim \lambda^{-SAE}$ ). Only measurements taken at relative humidity levels below 40% were  
283 considered. The  $b_{ext}$  in Eq. (5) was calculated as ( $b_{sca} + b_{abs,ref}$ ) at CMN, while retrieved from CAPS PMex data in the  
284 chamber.

### 285 **2.3.2 Particle size distribution**

286 The particle number size distribution ( $dN/d\log D_m$ ;  $D_m$ , mobility diameter), in the range of diameters from 10 to 800 nm,  
287 was measured at CMN by using a scanning particle mobility sizer (SMPS-TROPOS), operating at 5-minutes resolution  
288 and at 5/1  $l \text{ min}^{-1}$  sheath/aerosol flow rates. In chamber experiments the particle number size distribution, was instead  
289 measured in the range of diameters from 19.5 to 881.7 nm, by using a scanning particle mobility sizer (TSI SMPS)  
290 consisting in an  $^{85}\text{Kr}$  neutralizer, a Differential Mobility Analyzer (DMA, model 3080, TSI Inc.), and a Condensation  
291 Particle Counter (CPC, model 3772, TSI Inc.), operated at 2.0/0.2  $l \text{ min}^{-1}$  sheath/aerosol flow rates, and at a time resolution  
292 of 3 minutes. Measurements were corrected for diffusion losses and multiple charge effects with the instrument software.

293  
294 The size distribution measured by the SMPS was used to derive the volume mean mobility diameter ( $D_{m,vol}$ ). This  
295 parameter was derived from hourly averaged size distributions at CMN and to both 3-minutes size distributions and size  
296 distribution averaged on each filter spot of the aethalometer in chamber experiments.

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

299

300 **Table 1:** Datasets and optical instruments deployed for field observations at Cimone (CMN) and CESAM chamber experiments.

<b>Site</b>	<b>Period</b>	<b>Optical instrumentation</b>
<b>CMN</b>	July 2020 - November 2022	AE33, MAAP, nephelometer
	September - October 2022	AE33, MAAP, MWAA, nephelometer
<b>CESAM</b>	May 2021	
	October 2021	AE33, CAPS Pmex, nephelometer
	December 2022	

301

302

303

304 **Table 2:** Technical details of the instruments used in the present analysis.

Instrument	Wavelengths (nm)	Percent uncertainty	Reference	Flow rate (l min <sup>-1</sup> )		Temporal resolution		Size range (nm)	
				CMN	CESAM	CMN	CESAM	CMN	CESAM
<b>Aethalometer (model AE33, Magee Sci.)</b>	370, 470, 520,	± 24 %	Drinovec et al. (2015)	5	2	1 min	1 min	-	-
	590, 660, 880, 950		Mosquera et al. (2021)						
<b>Multi-Angle Absorption Photometer MAAP (model 5012, Thermo Sci.)</b>	637	±12%	Petzold and Schönlinner (2004) Petzold et al. (2005)	17	-	1 min	-	-	-
<b>Multi-Wavelength Absorption Analyzer (MWAA, custom-made)</b>	375, 405, 532, 635, 850	± 3-20 %	(Massabo et al., 2013)	-	-	24 hours	-	-	-
<b>Nephelometer (model 3563, TSI Inc.)</b>	450, 550, 700	± 8 %	Anderson (1996) Anderson and Ogreen (1998) Massoli et al. (2009)	35	2	1 min	1 sec	-	-
<b>Cavity Attenuated Phase Shift Extinction (CAPS PMex, Aerodyne)</b>	450, 630	± 6%	Massoli et al. (2010)	0.85	-	-	1 sec	-	-
<b>SMPS</b>	-	±10%	(Wiedensohler et al., 2012)	5	2	5 min	3 min	10-800	19.5-881.7

305

## 306 3 Results

### 307 3.1 Investigating C dependence on particle properties with chamber experiments

308 The experiments in the CESAM simulation chamber, presented in this study, were designed to investigate the specific  
 309 dependence of the C-factor on the chemical and physical properties of highly absorbing BC-containing soot particles. The  
 310 five different cast soot (CS) studied in the present work correspond to four fuel–lean burning conditions (CS1–CS4) and  
 311 one fuel–rich condition (CS5) leading to particles with a decreasing EC/TC ratio ( $0.79 \pm 0.11$  for CS1 to  $0.00 \pm 0.22$  for  
 312 CS5) and decreasing number median diameter (from an average of  $145 \pm 12$  nm for CS1 to  $79 \pm 2$  nm for CS5, expressed  
 313 as mobility diameter (Heuser et al., 2025)).

#### 314 3.1.1 Average chamber C value and the role of loading correction

315 The average values of C and their corresponding uncertainties for each experimental condition in the chamber are  
 316 summarized in Table 3. These values range from  $2.89 \pm 0.03$  to  $3.9 \pm 0.06$ , with the highest values corresponding to CS5,  
 317 which is characterized by a higher organic content and smaller particle sizes. The lowest values correspond to CS1, which  
 318 has a higher content of elemental carbon and larger particle sizes. The C values obtained for soot in the chamber  
 319 experiments are similar, considering the uncertainties, to the values reported in other studies from the urban site of Milan  
 320 (Bernardoni et al., 2021; Ferrero et al., 2021) and laboratory experiments using soot generated with a miniCAST burner  
 321 (Kalbermatter et al., 2022).

322 Due to the high concentration of absorbing soot, the possible dependence of the retrieved C on the loading correction  
 323 scheme was also evaluated. Indeed, in case of high absorbing aerosols concentration and frequent filter changes, the  
 324 internal AE33 correction can provide uncertain results. The time variability of the  $b_{\text{atm,LC}}$  may in some cases not to follow  
 325 the same decreasing trend over time as the reference measurement. However, it showed a steeper decline before and after  
 326 the filter change, with a more rapid decrease in these points, making the trend appear more abrupt and also discontinuous  
 327 around the filter transition. In these cases, the AE33 data can be re-corrected for loading effect. With this aim, the AE33  
 328 can be treated like a single-spot aethalometer and the loading correction schemes from W2003 and C2010, or the Dual  
 329 Spot aethalometer method itself with a constant compensation parameter, be applied. In the present analysis we tested the  
 330 impact of different loading correction schemes on C retrieval and found that the method applied to correct loading can  
 331 introduce differences of maximum 30% in the average values. Despite not impacting the conclusion of the present study,  
 332 the role of loading effect and correction scheme on retrieved C and  $b_{\text{abs}}$  should be taken into consideration. A detailed  
 333 discussion of this specific analysis is provided in Supplementary Information.

335 **Table 3:** Average C values for each soot type (CS1-CS5) and associated particles properties. In particular, the average volume mean  
 336 diameter ( $D_{\text{m,vol}}$ ), absorption coefficient  $b_{\text{abs}}$  and single scattering albedo (SSA). These values were derived averaging the results  
 337 obtained for all aethalometer filter spots corresponding to the same CS type. Values obtained in this study are shown together with the  
 338 ratio between elemental and total carbon mass concentration (EC/TC) and the Absorption Ångstrom Exponent (AAE) derived in Heuser  
 339 et al. (2025).

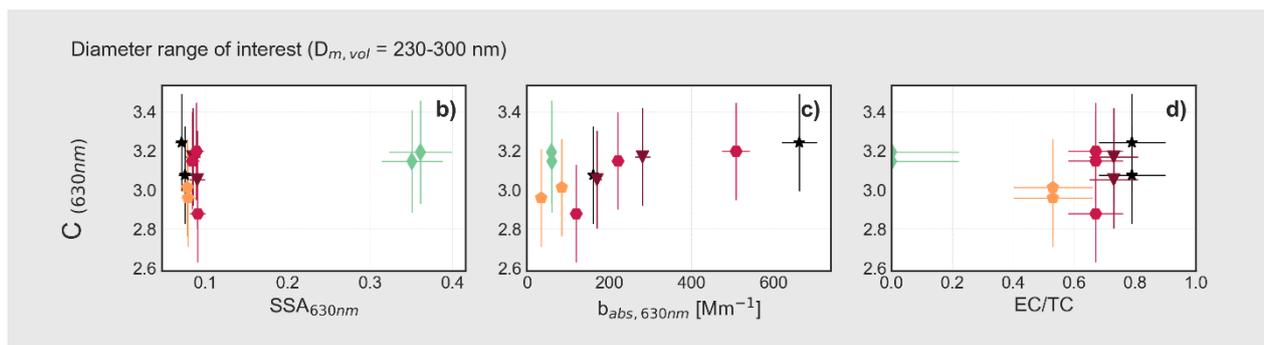
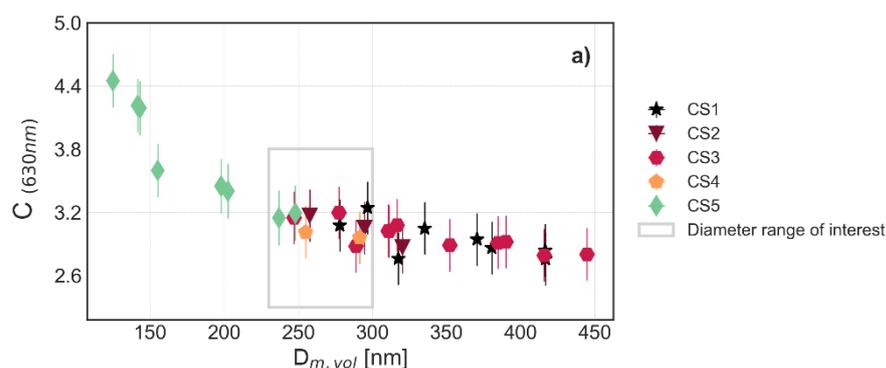
	C (630 nm) (this study)	$D_{\text{m,vol}}$ [nm] (this study)	$b_{\text{abs}}$ [ $\text{Mm}^{-1}$ ] (630 nm) (this study)	SSA (630 nm) (this study)	AAE (Heuser et al., 2025)	EC/TC (Heuser et al., 2025)
CS1	$2.89 \pm 0.03$	$351.5 \pm 15.9$	$450 \pm 30$	$0.08 \pm 0.01$	$1.27 \pm 0.13$	$0.79 \pm 0.11$
CS2	$3.00 \pm 0.03$	$291.0 \pm 18.2$	$179 \pm 11$	$0.09 \pm 0.01$	$1.36 \pm 0.21$	$0.73 \pm 0.08$
CS3	$2.96 \pm 0.03$	$339.9 \pm 18.8$	$400 \pm 30$	$0.09 \pm 0.01$	$1.59 \pm 0.22$	$0.67 \pm 0.09$
CS4	$2.99 \pm 0.03$	$273.2 \pm 18.3$	$59 \pm 4$	$0.08 \pm 0.01$	$1.88 \pm 0.31$	$0.53 \pm 0.13$
CS5	$3.90 \pm 0.06$	$181.2 \pm 16.5$	$63 \pm 6$	$0.29 \pm 0.03$	$3.79 \pm 0.33$	$0.00 \pm 0.22$

340

### 341 **3.1.2 C dependence on soot microphysical properties: the dominating role of size**

342 The C values at 630 nm averaged over each aethalometer filter spot interval were analyzed to identify the dependence on  
343 average properties of the particles collected on the filter. These properties include particle size, absorption coefficient,  
344 SSA, and chemical composition (measured as the EC/TC ratio). The results are summarized in Figure 1. The analysis  
345 shows that the C factor increases from 2.75 to 4.45 as the volume mean particle diameter of BC generated particles  
346 decreases from 444.8 nm to 124.9 nm. This is particularly evident for particles smaller than 200 nm, while at larger sizes  
347 the trend is not significant when considering the associated uncertainties. The points with the highest C values and smallest  
348 sizes correspond to experiments involving CS5 particles. These particles are not only smaller but also differ in  
349 composition, with a negligible fraction of EC in spite of dominating OC composition. This compositional difference  
350 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  
351 the AAE from around 1.27-1.88 to 3.79 (Heuser et al., 2025). However, focusing on a subset of points with limited size  
352 variability (230-300 nm, highlighted in the grey box), it becomes evident that at similar diameters, C values are  
353 comparable for CS5 particles and CS1–CS4 particles. Moreover, for CS5 particles, C increases significantly from 3.15 to  
354 4.45 as the volume mean diameter decreases from 247.9 nm to 124.9 nm. This suggest that the size is the dominant factor  
355 impacting the C variability in these experiments. This result is likely attributable to the greater penetration of smaller  
356 particles into the filter matrix (Huang et al., 2013; Hinds, 1999), that affect the multiple scattering effects between particles  
357 and filter, leading to a larger overestimation of true absorption by the aethalometer. This interpretation agrees with  
358 modelling results indicating that particle stratification increases the ratio of filter-based and real absorption as particle  
359 size decreases (Moteki et al., 2010). Our results are consistent with previous laboratory studies on other filter-based  
360 instruments such as COSMOS and PSAP (Lack et al., 2009; Nakayama et al., 2010), and two previous studies on the  
361 aethalometer (Drinovec et al., 2022; Yus-Díez et al., 2025). These studies show that for the AE33, C is larger for soot  
362 particles below ~250 nm than for particles above ~400 nm, while dust samples generally exhibit lower C values, in line  
363 with reduced artefact for larger particles.

364 The variability in SSA, which was observed to influence the C factor at high values in previous works (Yus-Díez et al.,  
365 2021), appears to have a limited impact in this low-SSA regime. The C-size dependence was observed for any loading  
366 correction scheme applied as shown in Supplementary Information.



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**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).

372

### 3.2 Ambient variability of the C factor

373

In this section, the results obtained from field measurements on the environmental variability of the correction factor C are presented. These results provide insights into how C values fluctuate across different seasons and aerosol properties.

374

375

#### 3.2.1 Average ambient C value and seasonal variability

376

The variability of the C factor at a wavelength of 637 nm was analysed on hourly base at the CMN site. The average C value over the period July 2020 - November 2022 is 2.35 with a standard deviation of 0.58. This value is comparable, within the combined uncertainties of MAAP and AE33 (27%), with the 2.45 value suggested by the ACTRIS guidelines for M8060 filter tape (obtained as the product of a  $C_{instr}$  factor (1.39) and the harmonization factor  $H^*$  (1.76); <https://www.actris-ecac.eu/particle-light-absorption.html>). This value is also in line, even if slightly lower, to mean values reported in other studies conducted at various sites (urban, background and mountain sites) using the same reference techniques (MAAP, MWA) and filter tape, which range between 2.44 and 2.66 (Moschos et al., 2021; Valentini et al., 2020; Yus-Díez et al., 2021). However, when considering the range of values reported in the literature, which consider different reference techniques for measuring absorption (e.g., two-wavelength Photothermal Aerosol Absorption Monitor PTAAM-2 $\lambda$ , and the Cavity Attenuated Phase Shift CAPS SSA), various AE33 filter types, different wavelengths, and diverse environmental or experimental conditions (such as chamber studies with artificially generated soot or emissions from heavy fuel oil), the reported values can vary significantly from 2.26 to 8.26 (Corbin et al., 2018; Laing et al., 2020; Ferrero et al., 2021; Bernardoni et al., 2021; Kalbermatter et al., 2022). The averaged ambient value was also 23-66% lower compared to the average values obtained for freshly emitted soot in the chamber experiments. 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

391

392 particles (diameter <200 nm) and for aged aerosol (Yus-Díez et al., 2025; Kalbermatter et al., 2022; Romshoo et al.,  
393 2022). Such bias would lead to higher MAAP absorption values and, consequently, to lower C values in ambient  
394 measurements at CMN compared to those obtained in CESAM. In addition, the aethalometers operated at different flow  
395 rates in the ambient and chamber setups (5 and 2 L min<sup>-1</sup>, respectively). As shown by Moteki et al. (2010) and Huang et  
396 al. (2013), variations in flow rate and face velocity can affect particle penetration and stratification by size within the  
397 filter. These effects may modify both the absolute value of C (which is expected to decrease with increasing flow rate)  
398 and its dependence on particle size. A detailed quantification of this effect is beyond the scope of the present study, but it  
399 may represent one of the factors contributing to the lower absolute C values observed in ambient measurements.

400

401 Figure 2 and Table 4 summarize the seasonal variability of C values at the CMN site. Throughout the year, C mean ranged  
402 from a minimum of 2.30 in spring to a maximum of 2.41 in winter, indicating a slight increase in the colder months. At  
403 the same time, the coldest seasons (winter and autumn), also showed the greatest variability, resulting in the highest  
404 standard deviations (0.7-0.8). The difference between the seasons, although significant based on the Kolmogorov-  
405 Smirnov test, was small compared to the combined instrumental uncertainty of the MAAP and the AE33. Nonetheless,  
406 the seasonal variability observed at CMN was smaller in magnitude and opposite in trend compared to the biogenic-  
407 dominated site in the Finnish boreal forest (Luoma et al., 2021) and to the mountain site of Montsec d'Ares (Spain, 1570  
408 m a.s.l.; Yus-Díez et al., 2021). In these previous studies, seasonal variability was explained as a result of dependence on  
409 optical (SSA; Yus-Díez et al., 2021) and microphysical (diameter; Luoma et al., 2021) properties, as well as aerosol  
410 composition (presence of dust; Yus-Díez et al., 2021). Hence, we addressed the dependency of C at CMN on particles  
411 properties, concentration and wavelength.

### 412 3.2.2 Lower limit of $b_{\text{abs}}$ for C determination

413 One of the most critical challenges in determining C at a remote site like CMN, where aerosol concentrations are low, is  
414 that the absorption coefficient ( $b_{\text{abs}}$ ) often approaches the detection limits of the instruments used. At very low  
415 concentrations, measurements are increasingly affected by instrumental noise, compromising the reliability of the  
416 resulting C values. To set a reasonable threshold for investigating the dependence of C on particle properties—while  
417 avoiding the exclusion of a substantial portion of the dataset—we first examined how C varies with the absorption  
418 coefficient ( $b_{\text{abs}}$ ) measured with the MAAP, and assessed the influence of low  $b_{\text{abs}}$  values on the C estimates.

419 Figure 3 shows the dependence of C on  $b_{\text{abs}}$ , with higher C values observed at decreasing  $b_{\text{abs}}$ . In particular, C more  
420 frequently reaches values of 10 or higher when  $b_{\text{abs}}$  falls below 0.06 Mm<sup>-1</sup>, although this condition accounts for less than  
421 3% of the dataset. These extreme values can significantly affect the median C (Figure 3), and are more commonly  
422 observed during winter and autumn (Table 4) seasons less influenced by anthropogenic emissions from the Po Valley  
423 (Marinoni et al., 2008). At such low  $b_{\text{abs}}$  levels—close to or below the detection limits of both the MAAP and AE33—  
424 the signal becomes increasingly dominated by instrumental noise, undermining the reliability of the computed C values  
425 (Cuesta-Mosquera et al., 2021). This likely explains both the higher standard deviation observed in winter and the  
426 unusually high C values recorded during that season. For this reason, data points where  $b_{\text{abs}}$  is below 0.06 Mm<sup>-1</sup> were  
427 excluded from subsequent analyses.

428

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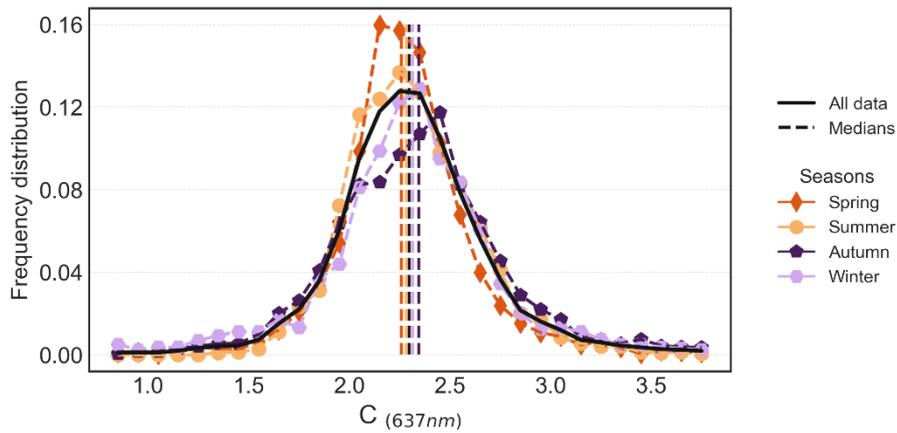
430

431 **Table 4:** The mean and standard deviation of C and  $b_{abs}$  for each season.

	Winter	Spring	Summer	Autumn	Whole
<b>C</b>					
<b>Mean (SD)</b>	2.40 (0.80)	2.30 (0.45)	2.32 (0.35)	2.41 (0.70)	2.35 (0.58)
<b><math>b_{abs}</math> [<math>Mm^{-1}</math>]</b>					
<b>Mean (SD)</b>	0.77 (1.06)	1.49 (1.26)	1.89 (1.08)	1.08 (1.19)	1.36 (1.23)

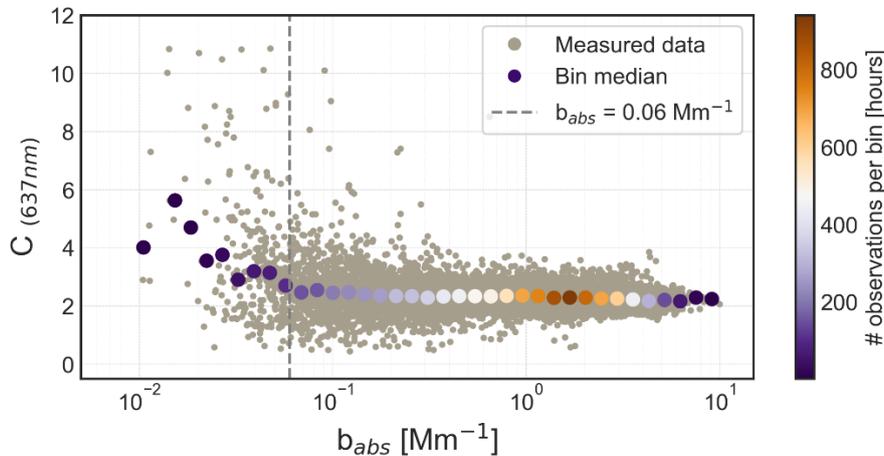
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433



434

435 **Figure 2:** Histograms representing hourly averaged C values frequency distribution at 637 nm for the period July 2020 – November  
 436 2022 at CMN. The black line represents the entire period, the coloured histograms correspond to each season in the analysed period.  
 437 The median values corresponding to each season are also represented as dashed vertical lines. The MAAP was used as reference  
 438 technique for measuring the absorption coefficient.



439

440 **Figure 3:** Scatterplot of hourly averaged C values at 637 nm as a function of the corresponding  $b_{abs}$  values measured by the MAAP at  
 441 CMN. The reference technique used to measure the absorption coefficient is the MAAP. Bigger dots are used to represent the median  
 442 C values in bins of  $b_{abs}$ .

### 443 3.2.3 C factor increase at high SSA

444 Previous studies have demonstrated that the optical properties of sampled particles can directly influence aethalometer  
 445 measurements by altering the amount of scattered light, which the instrument may incorrectly interpret as absorption. An  
 446 increase in the SSA has been linked in these studies to a corresponding rise in the C values (Schmid et al., 2006; Backman  
 447 et al., 2017; Yus-Díez et al., 2021).

448

449 The C values measured during the period considered in this study, are binned to the SSA values (Figure 4). The number  
450 of bins was chosen using the ‘auto’ mode of the numpy histogram\_bin\_edge function (python). This function selects as  
451 best number of bins, the highest value between those obtained with the Freedman and Diaconis, (1981) or the Sturges,  
452 (1926) criteria. For each bin, the median C and the corresponding 25<sup>th</sup> and 75<sup>th</sup> percentiles were derived.

453

454 At CMN, C remained relatively constant for SSA values below 0.90. It increased slowly for values between 0.90 and  
455 0.94, while a more significant variability was observed for SSA higher than 0.94 (Figure 4). This trend aligns with  
456 observations in other studies, where C becomes higher at higher SSA values due to an increase in the multiple scattering  
457 between particles and filter fibers and a reduced signal-to-noise ratio in low-absorption regimes (Schmid et al., 2006;  
458 Yus-Díez et al., 2021). The SSA-C relationship was fitted using the cross-sensitivity to scattering law proposed by Schmid  
459 et al. (2006) and applied in Yus-Díez et al. (2021) as:

460

$$461 \quad C = C_f + m_s \left( \frac{SSA}{1 - SSA} \right) \quad (7)$$

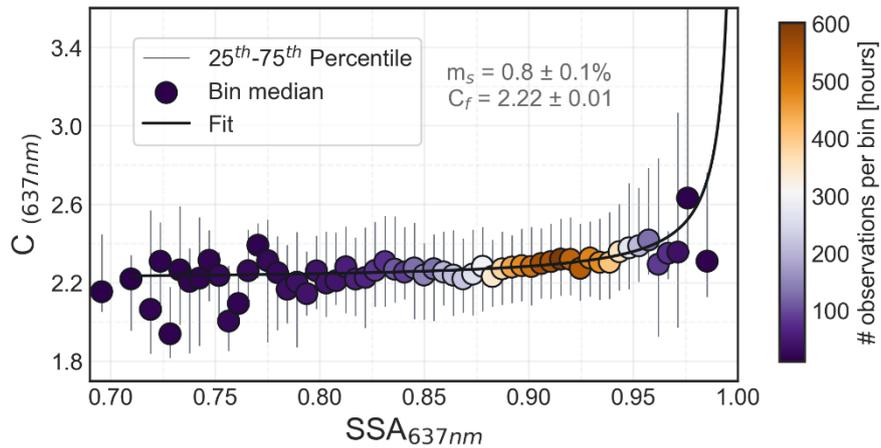
462

463 Compared to the results presented in Yus-Díez et al. (2021) for another mountaintop site, this curve shows a pronounced  
464 increase only at higher SSA values. The fitted curve closely matches the data up to a SSA of 0.97, becoming less  
465 representative at higher SSA. The deviation from the expected curve appears to be linked to the occurrence of strong dust  
466 events during the two-year period. In fact, over 80% of the hours with SSA values above 0.97 and C values below the  
467 mean of 2.3 correspond to days identified as dust-affected, characterized by coarse PM mass concentrations exceeding  
468 37  $\mu\text{g}/\text{m}^3$ , according to Vogel et al. (2025). The median C during these days is 2.0, suggesting a decrease in C during dust  
469 events, likely related to the increased presence of coarse particles collected on the filter, even at high SSA values. This  
470 decrease is consistent with the findings of Di Biagio et al. (2017), who reported lower C values for the AE31 when  
471 comparing measurements of ambient air and mineral dust particles.

472

473 These results highlight the importance of carefully selecting appropriate correction factors. Under conditions of high SSA,  
474 absorption is typically minimal and may approach the detection limits of the instruments, increasing uncertainty in C  
475 estimation. Moreover, we should consider that this dependence, estimated using the MAAP as a reference, may have been  
476 underestimated. As shown by Yus-Díez et al. (2025), this instrument is also affected by cross-sensitivity to scattering  
477 when compared with a non-filter-based technique.

478



479

480 **Figure 4:** C (637 nm) dependence on the SSA (derived at 637 nm combining MAAP and nephelometer). Reference absorption  
 481 instrument: MAAP. Dots represent the mean values in each SSA bin, the grey lines the 25<sup>th</sup> and 75<sup>th</sup> percentile. The dots colours  
 482 represent the number of observations used in each bin for calculating the mean and the curve obtained using Yus-Díez et al. (2021)  
 483 relation.

#### 484 3.2.4 C factor increase with decreasing particles size

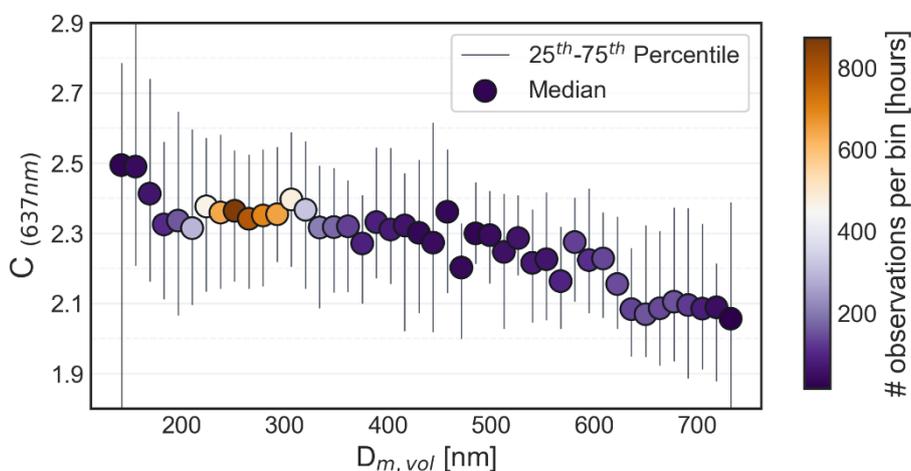
485 In this section, we examine the dependence of the C factor on particle size under ambient atmospheric conditions, where  
 486 isolating the influence of individual aerosol properties is not straightforward. The objective is to evaluate whether particle  
 487 size remains a significant driver of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

490 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  
 491 resulting C–size relationship shows an increase of approximately 25% in C as  $D_{m,vol}$  decreases from 732.1 nm to 141.4  
 492 nm (Figure 5). This trend is consistently observed across the median as well as the interquartile range, indicating a clear  
 493 dependence of C on particle size. To ensure that the observed variability in C was not primarily driven by other  
 494 parameters—such as the SSA or exceptionally low absorption values—rather than by particle size itself, we examined  
 495 the mean SSA and absorption coefficient within each size bin. The results show that neither of these parameters alone  
 496 can account for the whole observed variability in C with size in Figure 5 (see Supplementary Information, Figure S3 and  
 497 Figure S4).

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

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

509 particle size on  $C$  under ambient conditions could be partially masked by the measurement bias inherent to the reference  
 510 technique used. Differences in sampling flow of the aethalometer between the ambient and chamber measurements may  
 511 also have contributed to this weaker dependence, since different flow rates are expected to affect the penetration of the  
 512 particles as well as the resulting  $C$  dependence on size (Moteki et al., 2010; Huang et al., 2013).  
 513



514  
 515 **Figure 5:**  $C$  (637 nm) dependence on the geometric mean diameter  $D_{m,vol}$ . Reference absorption instrument: MAAP. Dots represent  
 516 the mean values in each  $D_{m,vol}$  bin, the grey lines the 25<sup>th</sup> and 75<sup>th</sup> percentile. The dots colours represent the number of observations  
 517 used in each bin for calculating the median.

### 518 3.2.5 Absence of spectral dependence

519 Figure 6 shows the spectral dependence of  $C$  at CMN for observations between September and October 2022, with 43  
 520 filters collected. The mean  $C$  values range from  $2.38 \pm 0.62$  at 470 nm to  $2.13 \pm 0.60$  at 880 nm (Table 5). A statistical  
 521 Kolmogorov-Smirnov test was applied to quantify the significance of the spectral dependence, and a potentially  
 522 significant distinction was observed between the 470 nm and 880 nm mean  $C$  values ( $p$ -value = 0.042). However, for all  
 523 other wavelength pairs, the test yielded  $p > 0.05$ , indicating no statistically significant differences, and that despite the  
 524 high variability in the data, with standard deviations reaching 0.63. Since the overall precision of the AE33 is not  
 525 wavelength-dependent (Cuesta-Mosquera et al., 2021), all wavelength-dependent  $C$  values reported in this study fall  
 526 within the relative error of  $\pm 31\%$ , obtained combining the AE33 and MWAA uncertainties.

527  
 528 Limited studies have investigated the multi-wavelength absorption reference methods for both models of the  
 529 aethalometer. For the AE31 model, Bernardoni et al. (2021) and Ferrero et al. (2021) did not observe a notable wavelength  
 530 dependence of the  $C$  factor, although they found that applying a wavelength-dependent  $C$  correction improved the  
 531 agreement in the measured absorption coefficient for both AE31 and AE33. In contrast, for the AE33 model, neither  
 532 Bernardoni et al. (2021) nor Ferrero et al. (2021) detected significant wavelength dependence after accounting for  
 533 uncertainties. However, Moschos et al. (2021) reported a decreasing trend in the  $C$  factor from 2.5 at 370 nm to 2.3 at 880  
 534 nm in Swiss background conditions. Moreover, Yus-Díez et al. (2021) found no significant wavelength dependence,  
 535 except for aged aerosols, where they observed an increasing  $C$  value with wavelength, ranging from 3.47 to 4.03.

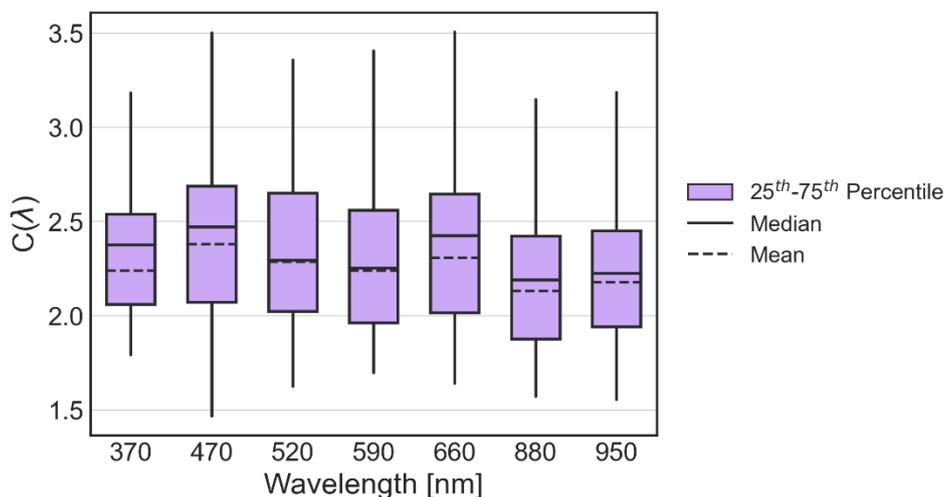
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**Table 5:** Mean and standard deviation (SD) of C values for each aethalometer wavelength at CMN. The MWAA was used as reference instrument for the absorption coefficient.

	370 nm	470 nm	520 nm	590 nm	660 nm	880 nm	950 nm
<b>Mean</b>	2.24	2.38	2.29	2.24	2.31	2.13	2.18
<b>(SD)</b>	(0.58)	(0.62)	(0.58)	(0.58)	(0.63)	(0.60)	(0.63)

540  
541



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544

**Figure 6:** Boxplots describing the statistics of C values derived at each aethalometer wavelength, the 25<sup>th</sup> – 75<sup>th</sup> percentile range, the mean and median are reported. The MWAA was used as the reference technique for measuring the absorption coefficient.

#### 545 4 Conclusions

546 In this work we studied the role of aerosol properties on the variability of the multiple scattering correction factor C for  
547 the dual-spot aethalometer AE33. For this, we combined data from a long term series in a mountaintop station in northern  
548 Italy at Monte Cimone, CMN, and from multiple experiments on laboratory-generated soot at the CESAM simulation  
549 chamber. We calculated the C factor comparing the attenuation coefficient measured by the AE33 with the absorption  
550 coefficient measured with independent techniques: the MAAP and MWAA at CMN, and the extinction-minus-scattering  
551 in chamber experiments.

552

553 The C factor obtained by chamber experiments ranged from 2.89 to 3.9 at 630 nm, the highest values corresponding to  
554 higher organic content and smaller particle sizes, while the lowest values corresponding to a higher content of elemental  
555 carbon and larger particle sizes. The possible dependence of the retrieved C on the loading correction scheme applied was  
556 also evaluated and quantified in max 30% of difference in the averaged values.

557

558 We found an averaged C factor at 637 nm of  $2.35 \pm 0.58$  at CMN. This value shows a slight seasonal variability with  
559 higher values in winter than in summer, difficult to associate to specific particle properties and mainly driven by the  
560 impact on C of the very low signal-to-noise ratio of the absorption coefficient, causing very high C values at low  $b_{\text{abs}}$   
561 ( $<0.06 \text{ Mm}^{-1}$ ). This was significant particularly at very high SSA values ( $>0.94$ ), where an impact of the low signal-to-  
562 noise ratio of the absorption coefficient cannot be excluded. Moreover, part of the variability at high SSA values was  
563 influenced by changes in aerosol properties, such as the increased presence of coarse particles during dust events. We  
564 studied also the C spectral dependence finding no evidence of a variability of C within the 370-950 nm spectral range.

565

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

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

## 578 5 Appendix

579 **Table A1.** List of digital object identifier (DOI) for the CESAM chamber experimental data supporting the findings of this study.  
 580 Those data are available through the Database of Atmospheric Simulation Chamber Studies (DASCS) of the EUROCHAMP Data  
 581 Centre (<https://data.eurochamp.org/data-access/chamber-experiments/>).

582

Cast Soot Type (Date of experiment)	Dataset DOI reference in the EUROCHAMP Data Centre
CS1 (23/02/2021)	<a href="https://doi.org/10.25326/8KVR-AA70">https://doi.org/10.25326/8KVR-AA70</a>
CS1 (24/02/2021)	<a href="https://doi.org/10.25326/M24W-V933">https://doi.org/10.25326/M24W-V933</a>
CS1 (19/10/2021)	<a href="https://doi.org/10.25326/5144-DY86">https://doi.org/10.25326/5144-DY86</a>
CS1 (12/12/2022)	<a href="https://doi.org/10.25326/2BD3-Q151">https://doi.org/10.25326/2BD3-Q151</a>
CS1 (14/12/2022)	<a href="https://doi.org/10.25326/9G66-MG61">https://doi.org/10.25326/9G66-MG61</a>
CS1 (15/12/2022)	<a href="https://doi.org/10.25326/XJKP-JD08">https://doi.org/10.25326/XJKP-JD08</a>
CS1 (16/12/2022)	<a href="https://doi.org/10.25326/6030-BD74">https://doi.org/10.25326/6030-BD74</a>
CS2 (20/10/2021)	<a href="https://doi.org/10.25326/ET9M-7H20">https://doi.org/10.25326/ET9M-7H20</a>
CS3 (26/10/2021)	<a href="https://doi.org/10.25326/0HMW-HM76">https://doi.org/10.25326/0HMW-HM76</a>
CS3 (06/12/2022)	<a href="https://doi.org/10.25326/T145-0S52">https://doi.org/10.25326/T145-0S52</a>
CS3 (07/12/2022)	<a href="https://doi.org/10.25326/9TR7-BB18">https://doi.org/10.25326/9TR7-BB18</a>
CS4 (22/10/2021)	<a href="https://doi.org/10.25326/VPM3-3W86">https://doi.org/10.25326/VPM3-3W86</a>
CS5 (28/05/2021)	<a href="https://doi.org/10.25326/BGCT-4J45">https://doi.org/10.25326/BGCT-4J45</a>
CS5 (21/10/2021)	<a href="https://doi.org/10.25326/34DX-YJ98">https://doi.org/10.25326/34DX-YJ98</a>
CS5 (08/12/2022)	<a href="https://doi.org/10.25326/FWN3-A342">https://doi.org/10.25326/FWN3-A342</a>
CS5 (09/12/2022)	<a href="https://doi.org/10.25326/6M6N-8P33">https://doi.org/10.25326/6M6N-8P33</a>

583

## 584 6 Open Research / Data availability

585 The CESAM data used in this study are available through the Database of Atmospheric Simulation Chamber Studies  
 586 (DASCS) of the EUROCHAMP Data Centre (<https://data.eurochamp.org/data-access/chamber-experiments/>) with the  
 587 identifiers listed in Appendix Table A1. The CMN database is available through ACTRIS DC (EBAS/NILU) and dust  
 588 event identification at CMN is available through the ITINERIS HUB (<https://doi.org/10.71763/XDZA-FA77>).

## 589 **7 Competing interests**

590 The authors declare that they have no competing interests.

## 591 **8 Author contributions**

592 LR, AM and CDB conceived the study and discussed the results. LR, AM, MM took care of field measurements. JH and  
593 LR conducted the CESAM experiments with contributions by CDB, MC, AB, EP, AM, JY, CY, MZ, PF, BPV, and JFD.  
594 DM, TI and VV performed the MWAA measurements. LR performed the full data analysis under the supervision of MZ,  
595 AM and CDB and with contributions from JH, MM and FV. CDB contributed to funding acquisition and project  
596 administration for CESAM experiments. LR, MZ, AM and CDB wrote the manuscript. All authors reviewed and  
597 commented on the paper.

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613 2014 – 2020.

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