

ESSD-2026-211

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*.

Review of: “Characterization and application of multi-decadal black carbon observations at a Mediterranean mountain site” by Zanatta and coauthors. This manuscript presents 18 years of eBC mass concentration data obtained from multiangle absorption photometer (MAAP). eBC data were integrated with ERA5 reanalysis and FLEXPART/CAMS regional and global transport models. Measurements were performed at CMN WMO/GAW global station (Italy). The manuscript is well written and present a rather unique dataset of eBC data. The sources of uncertainties are properly discussed and the manuscript can be published in ESSD after some minor correction.

We thank the reviewer for raising several interesting points for discussion. Hereafter, the authors provide a point-by-point response to the reviewer's comments.

The authors should mention in the Introduction section the inclusion of eBC in the new Air Quality Directive to further reinforce the usefulness of this work.

Thanks for the nice suggestion. Text was modified in:

- Beginning of introduction:
... “Although particulate matter remains the main aerosol metric used for air-quality assessment in Europe (e.g. Pisoni et al., 2025), the recent European Union Ambient Air Quality Directive (Directive (EU) 2024/2881) identifies black carbon as a pollutant of emerging concern and includes its measurement at monitoring supersites.” ...*
- End of Introduction:
... “It also provides an operational example of long-term eBC monitoring that may support environmental agencies implementing the monitoring requirements of the European Union Ambient Air Quality Directive (2024/2881).” ...*

The results of the trend analysis could be compared with the trend analyses reported for eBC by Savadkoobi et al. (2023) for Europe (<https://doi.org/10.1016/j.envint.2023.108081>). Moreover, given that the trend analysis reported here is for the absorption coefficient divided by a constant, the reported trends could also be compared (in %/year) with those for absorption reported by Rovira et al. (2025), at least in a qualitative sense, as the total absorption in Rovira et al., 2025 is that at 370 nm.

The discussion on trends was expanded and the level of details increased. To be more comparable with previous literature we introduced absolute and relative decrease of eBC mass concentration per year. The text now reads:

*... “Normalized to the median eBC concentration, this corresponds to a decrease of approximately 5 % yr⁻¹ over the 2007–2024 period at CMN. This value is within the range of decreasing eBC trends reported for European urban and traffic sites (–1.6 to –8.4 % yr⁻¹) until the early 2020s (Savadkoobi et al., 2023), as well as those reported for aerosol absorption at European mountain sites (–1.67 to –11.79 % yr⁻¹) until 2018 (Collaud Coen et al., 2020b). It is important to note, however, that the contribution of BrC to aerosol absorption at short wavelengths (370 nm) has been reported to increase across Europe, with trends ranging from +1.06 to +2.69 % yr⁻¹ across different environments (Rovira et al., 2025b). Although this does not directly imply a corresponding increase in BrC absorption at CMN, it highlights the potential for changes in aerosol composition to influence long-term absorption trends and the interpretation of eBC records. Previous trend analyses have often focused on annual or monthly aggregations, with limited consideration of seasonal or diel variability. This aspect is important in stations like CMN, where seasonal and diel cycles are remarkably evident, with increasing concentration in the summer months compared to winter and a marked increase during the central-day hours (**Error! Reference source not***

found.). Error! Reference source not found.b shows the eBC time series and corresponding trend lines calculated separately for each season (winter = DJF, spring = MAM, summer = JJA, autumn = SON). The results reveal marked seasonal differences in the magnitude of the long-term decline. The strongest eBC decrease is observed in spring ($-7.9 \text{ ng m}^{-3} \text{ yr}^{-1}$), whereas the winter trend is considerably weaker ($-0.94 \text{ ng m}^{-3} \text{ yr}^{-1}$), with summer ($-3.8 \text{ ng m}^{-3} \text{ yr}^{-1}$) and autumn ($-4.7 \text{ ng m}^{-3} \text{ yr}^{-1}$) showing intermediate values. When expressed relative to the median seasonal concentration, however, winter exhibits a stronger decrease ($-3.5 \% \text{ yr}^{-1}$), compared with $-1.8 \% \text{ yr}^{-1}$ in summer while intermediate seasons showed highest values (spring = $-6.4 \% \text{ yr}^{-1}$; autumn = $7.4 \% \text{ yr}^{-1}$). The high-resolution measurements presented in this work offer the additional advantage of covering the full diel cycle. The dataset can therefore be further aggregated into daytime and nighttime periods. To investigate how concentration changed between day and night in the 2007-2024 period, **Error! Reference source not found.c** shows the absolute increase in concentration from nighttime to daytime for summer and winter. This metric represents the increase in eBC concentration from the morning minimum to the central-day maximum observed at CMN ($\Delta \text{eBC}_{\text{day-night}}$). This metric provided an interesting insight on the long-term diel cycle, with day and night difference reducing over almost two decades in the summer months ($-3.8 \text{ ng m}^{-3} \text{ yr}^{-1}$), driven by a steep reduction in daily eBC concentration ($-2.7 \% \text{ yr}^{-1}$) compared to nighttime ($-1.1 \% \text{ yr}^{-1}$). Interestingly, winter $\Delta \text{eBC}_{\text{day-night}}$ remained almost constant, showing a negligible increase ($+0.38 \text{ ng m}^{-3} \text{ yr}^{-1}$), during the considered period, results of a similar decreasing trend in night ($-2.4 \% \text{ yr}^{-1}$) and day ($-2.2 \% \text{ yr}^{-1}$) eBC concentration.” ...

In relation to the comment above, it should be noted that eBC cannot be directly measured; therefore, the trends reported in the manuscript are based on absorption, since eBC is calculated using a constant MAC value of $10 \text{ m}^2/\text{g}$ over the entire period. Any change in MAC would therefore directly affect the derived eBC values. Unfortunately, EC data are not available at CMN, and the local MAC cannot be determined. While it is acknowledged that trends based on absorption normalized by a constant (i.e. eBC) are commonly reported in the literature, this limitation should be explicitly discussed in the manuscript.

We agree with the reviewer that eBC is not directly measured but calculated from the absorption coefficient, firstly derived from measured attenuation. The uncertainty of eBC calculation, implying the assumption of a mass absorption cross-section, is already discussed in Sect. 4.3.1. Since the same MAC is applied throughout the entire dataset, the reported eBC trends are directly proportional to the measured absorption trends. We believe that the existing discussion in Sect. 4.3.1 adequately describes this limitation and have clarified this point in our response. We acknowledge that potential temporal changes in MAC cannot be assessed in the absence of collocated elemental carbon or refractory black carbon measurements at CMN and could affect the absolute magnitude of the derived eBC trend. We therefore interpret the reported trends as trends in absorption expressed in eBC units based on a constant MAC. A specific sentence was added at the end of the trend section:

... “Because eBC is derived from absorption using a constant MAC, the reported eBC trends are directly proportional to the trends in the measured absorption coefficient; potential temporal changes in MAC cannot be assessed in the absence of collocated elemental carbon measurements at CMN.” ...

Line 97: I would use the term “pseudo-reference” for the MAAP instrument, as it cannot be a reference method.

From the metrology point of view the reviewer is right, there is no official reference for measuring aerosol absorption coefficient. However, we prefer avoiding the term “pseudo-reference”. We thus introduced a statement underlining the absence of standardized reference method:

... “Despite no official reference is defined to measure the absorption coefficient of atmospheric aerosol particles, thanks to its design” ...

I do not understand what use is made of the nephelometer data in this manuscript. In Table 1, “sensor” is associated with the different nephelometer models used at CMN. Does this mean that the P, T, and RH data are taken from the nephelometer? If this is the case, only the pressure measured by the nephelometer corresponds to ambient conditions. The temperature and RH do not correspond to ambient conditions. The authors should clarify this point more clearly, as later in the manuscript it is stated that meteorological data were recorded in the sampling line.

From the nephelometer we extracted temperature, pressure and relative humidity. These parameters do not refer to ambient or meteorological conditions but to the conditions inside the sampling line. Temperature and pressure were used to convert the eBC concentration from volumetric to STP conditions, while RH was used to flag humid measurement point (RH>40%). Following a comment of referee#1 the notation was changed and should improve the understanding. We tried to clarify the use of nephelometer data by improving:

- Caption of Table 1:
... *“Table 1 Temporal coverage of MAAP measurements, evolution of inlet type and nephelometer models (monitoring of temperature, relative humidity and pressure in the sampling line) at CMN from 2007 to 2024.”*
...
- Text in Section 3.2:
... *“These variables were used in data treatment line to flag uncertain observations and adjust the MAAP measurement to standard pressure and temperature (Section 4.2).”* ...
- Text in section 4.1:
... *“We thus addressed the variability of the conditions inside the sampling line (measure with nephelometer instruments) as a function of the various setups and compared it with outdoor ambient atmospheric conditions (measured with a meteorological station).”* ...

Lines 178–186: Rolling hourly negative values were flagged as invalid, which I agree with, even though the absorption measured at CMN is very low and negative values can therefore be expected. What is the percentage of invalid negative hourly values? How was the 1 mg/m³ threshold (defined as the difference between the 95th and 5th percentiles) selected? In my view, Level 0 data should include invalid data and calibration periods. However, I understand that these data were removed from Level 0.

Following the ACTRIS guidelines, the absorption photometer data L0 has to undergo a first cleaning procedure flagging the invalid and/or calibration periods. On native time resolution (minutes), the “negative” flag represents between 6% to 40% of all invalid observation points per year (Figure 1). This type of flagging contributes substantially to contain the negative values at Level 2 (hourly average). Although we were unable to remove all the negative hourly average values, these “survived” in a negligible number representing 0.007% of valid points. Considering the “noise” flagging, the threshold of 1 µg m⁻³ was chosen after several sensitivity tests. The limit was varied in the range of 0.1 to 5 µg m⁻³. Visual inspection helped to further limit the threshold range, while statistical analysis of the occurrence frequency of positive maxima and negative minima extremes narrowed down the choice to 1 µg m⁻³. The latter must not be assumed to be representative of other MAAPs deployed at other observatories. Although we tried to automatize the full procedure, a last visual inspection of the data by the operator is still necessary, especially for periods where noise is skewed towards very low values.

Line 186 was complemented with:

... *“The relative contribution of each “error” to invalid data points varied year by year without any tendency over the considered period (**Error! Reference source not found.**), indicating the absence of clear or univocal deterioration of the instrumental performances. It is important to note that a final visual inspection of the data remained necessary to complement the automatic data flagging, as the automated procedure may occasionally flag valid observations or fail to identify anomalous data points.”* ...

Table 2: It is indicated a MAC for dust of 16 m²/g. Please, correct.

A factor 10⁻³ was missing. Now corrected. Thanks.

Line 200: raw eBC with a MAC of 6.6 m²/g is classified as Level 0, whereas eBC calculated using a MAC of 10 m²/g is classified as Level 3. This point should be clarified more clearly.

In this work we decided to follow the full data reduction recommended by ACTRIS. The final product (L2) available on the EBAS database is the aerosol absorption coefficient and not eBC mass concentration. Considering the recent EU air quality directive, we intend to present eBC concentration, which is not an ACTRIS-compliant variable. Hence, L3-eBC was calculated from L2-absorption using the MAC value of 10 m² g⁻¹. The full process is described in Section 4, however, it results slightly fragmented due to the detailed description of the flagging

(Section 4.1) preceding the final product calculation (Section 4.2). Besides adjusting some details in the conclusive part of Section 4.2, we introduced a new figure (Figure 2) showing the data reduction scheme.

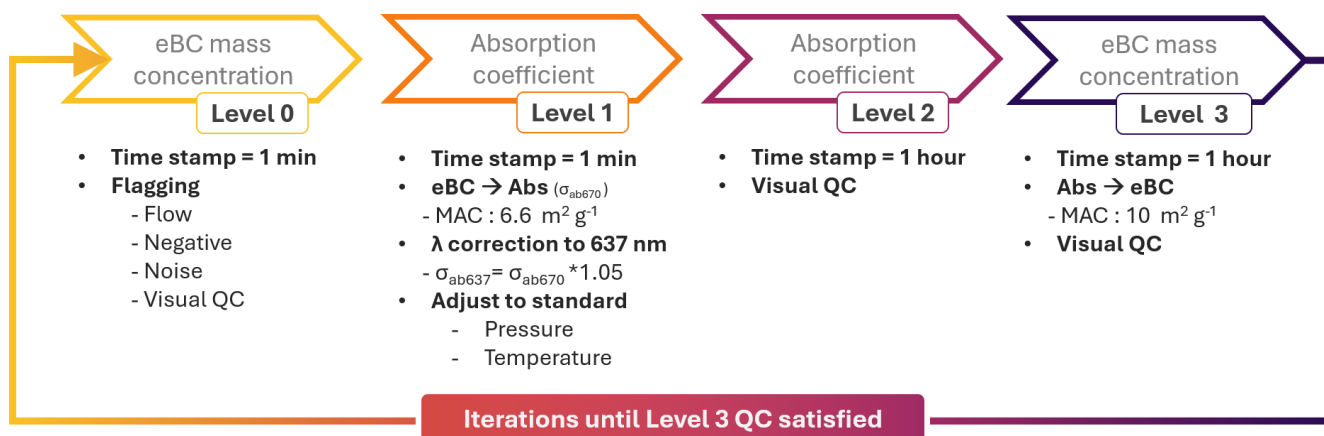


Figure 2 Schematics of data reduction applied to MAAP data acquired at CMN from 2007 to 2025. Level 0: raw eBC data validation; Level 1: conversion to absorption coefficient and data correction; Level 2: 1-hour averaging of absorption coefficient; Level 3: conversion of absorption coefficient to eBC with ambient relevant MAC and 1 hour averaging. Levels 0, 1, 2 are generated following the ACTRIS-RI guidelines.

For the period 2020-2024 AE33 data were also available. Using the simple AE source apportionment method (as e.g. Sandradewi), the authors could estimate the fossil and non-fossil contributions to BC and compare the trends calculated for this short period with the trend of eBC reported in the manuscript.

In view of the rising concern of wild-fires, an eBC source apportionment would be clearly interesting. Despite the AE33 data were fully analysed (including source apportionment) to estimate BrC contribution to absorption, we decided to exclude it from the current work. First, the data coverage of the AE33 was too short to provide a meaningful trend analysis. Second, we want to preserve the narrative and the scope of the manuscript and of the journal, which should not exceed in scientific or climatic dissertation. Nonetheless we gladly provide some short details here. We observed a clear seasonality with non-fossil fuel contribution up to 50% in winter period. This may confirm the higher influence of biomass burning in rural mountain area in winter and the traffic influence from the boundary layer in summer. However, it remains to be fully understood if this apportionment is fully driven by an actual shift of sources or also ageing processes connected with boundary layer and free troposphere.

Line 250: The pc1 of AAE calculated from fits 370-950 nm with $R^2 > 0.99$ has been suggested as good indicator of AAE_{BC} . Which is the pc1 in your dataset?

Thanks for pointing out this gap. We applied the pc1 method to linear-log fits in the 370-950, excluded the R^2 lower than 0.99 and dust periods. The pc 1 sits at 1.02, centred in the chosen window of AAE for BC (0.8-1.2). See figure below. The text was modified as:

... "To provide a range of estimations we used two different AAE_{BC} (0.8 and 1.2), representing a $\pm 20\%$ range around the AAE_{BC} (1.02) identified following the method proposed by Savadkoobi et al. (2025)." ...

Line 252: Is the contribution of BrC to absorption in winter due to long-range transport? I assume that in winter CMN is mostly well above the PBL. Or it is just due to less BrC emissions in summer?

The reviewer is right, CMN in winter is influenced mostly by FT conditions, while in summer the contribution of freshly emitted BC from the Po Valley dominates concentrations and absorption properties during daytime. Although we did not come to a clear conclusion yet, in winter the shift towards higher AAE values may be driven by both an increase of wood burning activity in the surrounding mountain areas and photochemical ageing. Considering the scope of the manuscript and the journal we did not speculate further on this aerosol processes.

Line 267: How have you estimated PMdust?, as PM10-PM1?

The full dust estimation is based on OPC measurements as described in (Vogel et al., 2025). More details were added as:

... "For dust days, the dust particle mass concentration (PMDust) was derived from the aerosol optical size distribution considering only particles larger than $1 \mu\text{m}$ and assuming a particle size dependent density. The full methodology is described in (Vogel et al., 2025c)." ...

Line 293: How were the different cloud types distinguished?

The method was introduced in Section 2 and fully described in Section 1 of the supplementary material. To improve the paper “navigation” we added a note:

... *“Here we focus on the drying efficacy under different cloud conditions (the cloud identification method is fully described in Section 1 of the supplementary material). ”* ...

Line 327: The winter eBC trend shows no significant variation. What could be the reason for this? Could this be due to free-tropospheric conditions in winter, or to high noise levels in the eBC measurements in winter?

Most likely, it is a combination of the two effects, we do not observe a decreasing gradient due to eBC signal already close to the limit of detection and to a dominance of free tropospheric conditions, which appears to be, for the moment, still representative of pristine conditions. In true honesty, we hypothesized a potential increase of eBC concentration in winter driven by higher temperature which may favour vertical extend of boundary layer and increase the coupling with CMN. Although we are working on this hypothesis by combining in-situ observations with FLEXPART simulations and ERA5 analysis and considering the scope of the manuscript and the journal, we did not speculate further on this topic.

Line 393: Please also quantify the offsets between FLEXPART and the measurements in relative terms (%), since eBC values at CMN are very low.

The median relative offsets were added to the text: Winter = +228%; Summer = - 9% .