



## Long-term analysis of carbonaceous fractions of particulate at a Central Mediterranean site in Italy

Eva Merico<sup>a,\*</sup>, Daniela Cesari<sup>a</sup>, Adelaide Dinoi<sup>a</sup>, Serena Potì<sup>a,b</sup>, Antonio Pennetta<sup>a</sup>, Ermelinda Bloise<sup>a</sup>, Daniele Contini<sup>a</sup>

<sup>a</sup> Institute of Atmospheric Sciences and Climate, ISAC-CNR, Str. Prv. Lecce-Monteroni km 1.2, 73100, Lecce, Italy

<sup>b</sup> Department of Engineering for Innovation, University of Salento, Str. Prv. Lecce-Monteroni 6, 73100, Lecce, Italy

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### ABSTRACT

Several research efforts were devoted to investigate long-term trends of carbonaceous aerosols in atmospheric particulate matter. Carbon is known to affect climate, human health, and cultural heritage being an important component also for planning future policies. This work investigates the longest dataset of carbon content (EC and OC) in PM<sub>2.5</sub> and PM<sub>10</sub> fractions available in south Italy (2015–2022). Equivalent black carbon (eBC) was determined hourly in PM<sub>10</sub>, using the multi-angle absorption photometer (MAAP), and it was used to evaluate the trend of the mass absorption coefficient (MAC) determined in-situ. PM<sub>2.5</sub> and its OC and EC content showed a statistically significant decreasing trend throughout years, since 2017–2018, approximately  $-5\% \text{ yr}^{-1}$ , falling in the ranges reported for other urban background sites in Europe. The decrease of PM<sub>10</sub> and its carbon content was more limited, approximately  $-2\% \text{ yr}^{-1}$  but it was not statistically significant ( $p > 0.05$ ). Temporal analysis revealed that seasonality played the most significant role in concentration pattern of PM and carbonaceous species, including secondary organic aerosol (SOC) and EC/OC ratio, compared to weekly or daily variabilities, with the highest daily values in winter/autumn. This was likely due to the contributions of biomass burning (for domestic heating and agricultural practices) and of road traffic both larger at this site during the autumn/winter. The value of MAC in-situ was  $12.4 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$  (as geometric mean  $\pm$  standard deviation) or  $12.6 \pm 2.5 \text{ m}^2 \text{ g}^{-1}$  (as arithmetic mean  $\pm$  standard deviation), showing a slight increase during summer and early autumn compared to the other season. MAC showed an increasing long-term trend of  $2.5\% \text{ yr}^{-1}$ .

### 1. Introduction

Research interest in characterising carbonaceous aerosol and related atmospheric processes has grown in the last years because of their influence on climate, human health, and conservation of cultural heritage (Bond et al., 2013; Fuzzi et al., 2015). The carbonaceous fraction is an abundant component of PM, often representing between 20 % and 50 % of PM (Bond and Bergstrom, 2006; Kanakidou et al., 2005; Putaud et al., 2010). Typically, the highest concentration values are found in urban areas densely populated (Ji et al., 2019; Qi et al., 2018; Sharma et al., 2018), compared to suburban and rural sites (Sandrini et al., 2014).

Carbon content of PM is typically divided in organic carbon (OC), elemental carbon (EC), and inorganic carbon (IC), whose major fraction is carbonate carbon (CC). While EC is mainly a primary component released during combustions (Chow et al., 2001), OC can have both pri-

mary and secondary origin (Gentner et al., 2012; Jimenez et al., 2009). According to the terminology, EC is a refractory light-absorbing component, minor in term of TC mass, containing only carbon, and it is determined by thermal or thermo-optical methods. It is defined eBC (equivalent black carbon) when measured by optical techniques (Petzold et al., 2013). CC is not often included in aerosol studies and it is not discussed further in this work being, on average, negligible at this site (Contini et al., 2014).

The European Directive 2008/50/EC requires measurements of EC and OC in PM<sub>2.5</sub> fraction in background areas. Furthermore, OC and EC (or eBC) are considered pollutants of emerging concern and possible components to be in atmospheric pollutants subjected to concentration limits, even if a standard common methodology to determine them is not yet available.

Speciation of carbonaceous aerosol in these two components OC and EC is operational, with the location of split point that is dependent on:

\* Corresponding author.

E-mail address: [eva.merico@cnr.it](mailto:eva.merico@cnr.it) (E. Merico).

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thermal protocol, chemical composition of PM, presence of water-soluble organic compounds (WSOC) (Piazzalunga et al., 2011), contribution of Brown carbon (BrC) (Corbin and Gysel-Beer, 2019; Pileci et al., 2021; Popovicheva et al., 2025; Sharma et al., 2017). Optical transmittance or reflectance is used to distinguish EC from the charring part of organic carbon (Birch and Cary, 1996; Chow et al., 1993). Therefore, in research networks such as the ACTRIS (Aerosol, Clouds and Trace Gases Research Infrastructure), harmonization protocols and inter-comparison of EC and TC measurements at different stations are used to maximize the comparability measurements at different sites.

The absorption coefficient (used for estimating eBC data) is usually measured on filter tapes using instruments like the Multi Angle Absorption Photometer (MAAP), the aethalometer, and the Particle Soot Absorption Photometer (PSAP). Measurements on filters are affected by multiple scattering and by loading effects that can be corrected with different approaches (Bernardoni et al., 2020; Petzold et al., 2002). Instruments are generally sensitive to chemical and optical properties of sampled carbon-containing particles, to relative humidity and filter-particle interactions (Bond et al., 1999; Lack et al., 2009; Weingartner et al., 2003). Among the various instruments, the MAAP is considered a reference for absorption measurements (Ammerlaan et al., 2017; Müller et al., 2011; Petzold et al., 2005).

Comparisons among different methods and different laboratories showed that comparability of eBC and thermo-optically determined EC could vary by up to a factor 4 (Watson et al., 2005). The factors mainly influencing comparability were: the thermal protocol used; the particle size distribution; the mixing state, and the chemical composition (Hitzenberger et al., 2006; Ram et al., 2010; Zhao et al., 2021).

The Mass Absorption Cross-section (MAC) is an important parameter, firstly introduced in Bond et al. (2006), commonly used for the conversion of the light absorption to eBC concentration. MAC is influenced by EC emission sources (Bond et al., 2004; Chen et al., 2020), by the chemical composition (Cheng et al., 2006), by ageing processes of particles (Knox et al., 2009; Peng et al., 2016; Wu et al., 2025), and by the mixing state of particles (Bond et al., 2006; Lan et al., 2013; Zhao et al., 2021). Commercial instruments often use a fixed MAC value, not related to in situ aerosol properties, leading to discrepancies between eBC and EC measurements (Wu et al., 2025). Recent studies discussed the need of harmonization in extensive inter-comparison exercises, in order to get a more realistic picture in determining MAC or eBC (Savadkoohi et al., 2023).

Recent studies investigate the trends of carbonaceous aerosol in different sites in Europe, as well as the trends of the MAC observed in different typologies of sites (Savadkoohi et al., 2023, 2024; Singh et al., 2024), finding a decrease in eBC mass concentrations related to traffic at urban/suburban sites, especially moving from southern to northern Europe. Temporal variation was larger than spatial one, with higher values in winter mostly due local anthropogenic sources in all European sites studied. However, long-term analysis of carbonaceous aerosols in south of Italy, that is a central Mediterranean hot spot, are still missing.

This study tries to address the mentioned gaps with the following objectives: (i) to present and to discuss the longest data series available in south Italy, and one of the longest available in Europe with simultaneous measurements on two size fractions, for OC, EC, in PM<sub>2.5</sub> and PM<sub>10</sub> and eBC (in PM<sub>10</sub>); (ii) to investigate the inter-annual trends and the seasonal trends of carbonaceous aerosol; to estimate the value of MAC in-situ comparing it with those obtained in other European ACTRIS sites. Results will be helpful in investigating the role of different sources of carbonaceous aerosols acting at local or regional level furnishing information for future mitigation policies.

## 2. Materials and methods

### 2.1. Sampling site and study approach

A long-term measurement dataset (January 01, 2015–December 31, 2022) was obtained at the Environmental Climate Observatory (thereafter ECO) of ISAC-CNR in Lecce (SE Italy, 40°20'8" N - 18°07'28" E, 37 m a.s.l.). The station is located inside the University campus (Fig. S1) and it is classified as an urban background site (Cristofanelli et al., 2017). ECO is a regional station of the Global Atmosphere Watch (GAW) Programme of World Meteorological Organisation (WMO), and a national facility of the European research network ACTRIS. The observatory is located in a strategic area, subjected to long-range transport phenomena from East Europe and coarse dust advection events (roughly 10 % of the days, average duration of 2.6 days, with a typical seasonality) from Africa (Conte et al., 2020), as well as sea spray contribution and secondary aerosol formation due to photo-chemical transformation of air pollutants, thus representing a mix of local emissions and regional background conditions in the heart of the Mediterranean basin.

Sampling was conducted by using an automatic low-volume sampler (SWAM 5A Dual Channel Monitor, FAI Instruments srl operating at 2.3 m<sup>3</sup>h<sup>-1</sup>) based on the  $\beta$ -attenuation method (Dinoi et al., 2017). Samples were collected on quartz fibre filters (Whatmann, 47 mm in diameter) thermally pre-treated (for 2h at 700 °C). The typical final uncertainties on PM concentrations were between 2 % and 3 % for both size fractions. A subset of 1578 simultaneous PM<sub>10</sub> and PM<sub>2.5</sub> samples were selected (roughly one sample every three days) for this study.

In addition, hourly eBC<sub>raw</sub> concentration, only for the PM<sub>10</sub> fraction, was obtained by a MAAP (Thermoscientific, mod. 5012), operating at a wavelength of  $\lambda = 670$  nm at 1 m<sup>3</sup>h<sup>-1</sup>. (Petzold et al., 2002). The instrument's algorithm converts the light absorption of aerosol to mass concentration of eBC<sub>raw</sub>, using a pre-defined value of MAC, equal to 6.6 m<sup>2</sup>g<sup>-1</sup> (Donateo et al., 2018; Kanaya et al., 2008). A schematic picture of the methodology approach is shown in Fig. S1.

### 2.2. Evaluation of EC and OC

Daily samples, stored at 4 °C between sampling and analysis, were analysed with a Sunset laboratory analyser (Sunset Laboratory Inc., OR, USA) using the thermo-optical transmittance (TOT) approach. Punches of 1.0 cm<sup>2</sup> obtained from quartz fibre samples, in both size fractions, were analysed using the EUSAAR2 thermal protocol (Cavalli et al., 2010). The instrument was calibrated (multi-point) using a standard a sucrose solution (2.198 g/l in water, CPAchem Ltd) only for OC because a reference material for EC is not available (Baumgardner et al., 2012). The calibration was linear with a slope of 0.97 and a negligible intercept (R<sup>2</sup>~1). In addition, the analyser undergone period intercomparison within other laboratories of the ACTRIS network. Blank filters, approximately one every thirty samples, were used to correct measured concentrations. The average OC in blanks was 2.46 ± 0.95 µg cm<sup>-2</sup> while the EC was negligible (<0.1 µg cm<sup>-2</sup>). The uncertainties on OC and EC concentrations were 5 % (OC) and 10 % (EC) (Conte et al., 2020).

### 2.3. Determination of SOC and POC

Determination of SOC and POC (i.e. the primary organic carbon) can be done following Turpin and Huntzicker (1995) using the EC tracer method. This is the most widely used approach, and it is based on the assumption of co-emission of POC and EC by primary combustion sources having a (OC/EC)<sub>pri</sub> ratio constant in time. The equations are:

$$POC = (OC/EC)_{pri} \times EC \quad (1)$$

$$SOC = OC - POC \quad (2)$$

where  $(OC/EC)_{pri}$  is the ratio in PM emitted by primary sources (not influenced by SOC formation) and OC and EC are the measured values. A number of approaches have been proposed for the estimation of  $(OC/EC)_{pri}$ : selecting the lowest OC/EC ratio (Castro et al., 1999; Pio et al., 2011); taking into account only selected samples with low photochemical activity (Cabada et al., 2004); choosing the EC/OC values above the average values of two (or more) standard deviations (high EC edge method) (Day et al., 2015). In addition, Wu and Yu (2016) introduced the minimum R-squared (MRS) method, that assumes source-independent EC and SOC and a hypothetical  $(OC/EC)_{pri}$  ratio corresponding to the minimum correlation between the estimated SOC and measured EC. All methodologies have limitations and advantages, in this work, the empirical EC-tracer approach was used, estimating the  $(OC/EC)_{pri}$  based on the MRS approach that may be more robust, in several sites, than the minimum OC/EC and percentile OC/EC method (Wu and Yu, 2016). In MRS method, the squared-correlation ( $R^2$ ) between measured EC and estimated SOC is examined as a function of a series of hypothetical  $(OC/EC)_{pri,h}$  ratios varying between 1 and 8. If variations of EC and SOC are independent, the  $(OC/EC)_{pri,h}$  corresponding to the minimum  $R^2$  represents the actual  $(OC/EC)_{pri}$  ratio. The evaluation of  $(OC/EC)_{pri}$  was done separately for the cold and warm periods and separately for  $PM_{2.5}$  and  $PM_{10}$ .

#### 2.4. Determination of MAC

The MAC used by MAAP is a predefined value equal to  $6.6 \text{ m}^2\text{g}^{-1}$  and it is used to infer  $eBC_{raw}$  mass concentration (in  $\mu\text{g m}^{-3}$ ) from light absorption coefficient measurements ( $\sigma_{abs, 670 \text{ nm}}$ ) (Eq. (3)). In this study, the actual MAC for the specific site was calculated (Baumgardner et al., 2012; Bond et al., 2013; Zanatta et al., 2016), re-computing all eBC concentration data:

$$eBC_{raw} = \sigma_{abs, 670 \text{ nm}} / 6.6 \quad (3)$$

$$MAC = \sigma_{abs, 670 \text{ nm}} * (1/CF) / (EC * 10^6) = \sigma_{abs, 637 \text{ nm}} / (EC * 10^6) \quad (4)$$

$$eBC = \sigma_{abs, 637 \text{ nm}} / MAC * 10^6 \quad (5)$$

Where  $CF = 0.952$  is a conversion factor applied to the output of the MAAP, to estimate the absorption coefficient at 637 nm (Müller et al., 2011); MAC and eBC are the final values calculated for the ECO site. This approach of evaluation is consistent with the analysis done on MAC on other sites including some stations of the ACTRIS network (Savadkoohi et al., 2024; Zanatta et al., 2016). In the rest of this work  $eBC_{raw}$  indicates the values directly calculated by the MAAP and eBC those calculated using the MAC in situ.

#### 2.5. Temporal variability analysis

A trend analysis was performed, similarly to other studies (Li et al., 2014; Luoma et al., 2021; Matos et al., 2025a, 2025b; Yttri et al., 2021; Zhao et al., 2017), including all 8 years of data available at the ECO

monitoring station, by applying different statistical tests and using ad-hoc RStudio packages (Carslaw and Ropkins, 2012), as described in the following.

1. Test for homogeneity and breakpoints with the Standard Normal Homogeneity test (SNHT) (Alexandersson, 1986), the Buishand's U and Range tests (Buishand, 1982), and the Pettitt's test (Pettitt, 1979; Wijngaard et al., 2003);
2. Trend analysis using the Modified Mann-Kendall test and the Theil-Sen's estimator.

Trends in time series are commonly investigated by parametric and non-parametric tests (suitable also for missing data points), and not necessarily normally distributed data. Specifically, temporal variations in data are identified and investigated by means the Modified Mann-Kendall non-parametric test, designed to detect statistically significant tendency in the data having significant increase or decrease over time, applying a variance correction approach for accounting serially correlated data (Yue and Wang, 2004). The non-parametric Theil-Sen method (Sen, 1968; Theil, 1950), on daily averages de-seasonalised (with *stl* function for seasonal trend decomposition using loess), estimates regression parameters of trends (i.e., slope, *p* value, slope uncertainty) in univariate time series, through bootstrap resampling. This estimator, using the median, is more robust to outliers and less influenced by missing values (Hollander and Wolfe, 1999), thus being preferred over OLS (ordinary least squares) estimator and is useful to quantify magnitude of variation (i.e., change per unit time).

### 3. Results and discussion

#### 3.1. Concentration and variabilities of PM and carbon

The long-term mean concentration of PM obtained at ECO were  $24.2 \pm 17.8 \mu\text{g m}^{-3}$  for  $PM_{10}$  and  $13.5 \pm 8.5 \mu\text{g m}^{-3}$  for  $PM_{2.5}$ . Considering the whole measurement period, OC and EC represented, on average, 17.4 % and 3.5 % of  $PM_{10}$  in mass, and 24.4 % and 5.3 % of  $PM_{2.5}$  (Table 1). An overview of the time series is reported in Fig. S2 that shows a similar periodicity of EC and OC, in both size fractions, with larger values during the cold seasons (autumn and winter) compared to the warm ones (spring and summer), likely as a consequence of biomass burning and traffic emissions that are larger at this site during the cold seasons (Cesari et al., 2018a). OC and EC are highly correlated, Pearson coefficient 0.84 ( $PM_{2.5}$ ) and 0.85 ( $PM_{10}$ ), suggesting the dominant role of similar combustion sources and the role of local meteorology including the boundary-layer height.

Table 2 compares the dataset of this work with those of other studies that include OC and EC measured with EUSAAR2 protocol simultaneously in  $PM_{2.5}$  and  $PM_{10}$  and at least for one year in order to include all seasons. There is variability associated to site typology leading to significant differences in concentration of PM and carbon fractions, i.e. high-altitude < suburban < urban background < urban sites. The values of this study fall in the ranges, for urban background sites (Perez et al., 2008; Samara et al., 2014), for  $OC_{PM_{2.5}}$  and for

**Table 1**

Average and inter-quartile range, between 25th and 75th percentiles (in parenthesis), of measured concentrations (in  $\mu\text{g m}^{-3}$ ) for the two fractions. The OC/EC and EC/TC ratios are also reported. Results are reported for all period and separating cold (winter-autumn) and warm (spring-summer) season.

	PM <sub>10</sub>						PM <sub>2.5</sub>					
	PM <sub>10</sub>	OC	EC	OC/EC	EC/TC	eBC	PM <sub>2.5</sub>	OC	EC	OC/EC	EC/TC	
All data	24.2 (15.9–27.7)	4.2 (2.1–4.9)	0.8 (0.4–1.1)	5.6 (4.1–6.6)	0.17 (0.13–0.19)	0.8 (0.4–1.0)	13.5 (7.7–17.0)	3.3 (1.5–4.0)	0.7 (0.3–0.9)	4.8 (3.6–5.7)	0.19 (0.15–0.22)	
Cold period	25.1 (15.5–29.6)	5.4 (2.3–6.9)	1.0 (0.4–1.4)	5.6 (4.2–6.7)	0.17 (0.13–0.19)	1.0 (0.4–1.3)	15.0 (7.7–19.2)	4.3 (1.7–5.7)	0.9 (0.4–1.2)	5.0 (3.6–6.2)	0.18 (0.14–0.21)	
Warm period	23.5 (16.2–26.6)	3.3 (1.9–4.1)	0.7 (0.3–0.9)	5.6 (4.1–6.4)	0.17 (0.14–0.19)	0.6 (0.3–0.9)	12.2 (7.8–15.3)	2.4 (1.3–3.0)	0.6 (0.3–0.8)	4.6 (3.5–5.4)	0.19 (0.16–0.22)	

**Table 2**

Average concentration ( $\pm$  std. Dev., in  $\mu\text{g m}^{-3}$ ) of  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$ , their carbonaceous fractions (OC, EC), in different sites (IT – Italy, SP – Spain, GR – Greece, JO – Jordan, NW – Norway), with, at least, one year-long study period and determination of carbonaceous fractions performed by EUSAAR2 protocol on both size fractions.

Site (period)	Site typology	$\text{PM}_{10}$	$\text{PM}_{2.5}$	$\text{OC}_{\text{PM}_{10}}$	$\text{EC}_{\text{PM}_{10}}$	$\text{OC}_{\text{PM}_{2.5}}$	$\text{EC}_{\text{PM}_{2.5}}$	Reference
Lecce ECO - IT (2015–2022)	Suburban	$24.2 \pm 17.8$	$13.5 \pm 8.5$	$4.2 \pm 3.5$	$0.8 \pm 0.7$	$3.3 \pm 3.2$	$0.7 \pm 0.6$	This study
Lecce ECO - IT (2015–2016)	Suburban	26.4	15.2	4.44	0.86	3.68	0.77	Merico et al. (2019)
Monte Curcio - IT (2016–2017)	High-altitude	$16.7 \pm 19.0$	$8.8 \pm 6.5$	$1.7 \pm 1.9$	$0.2 \pm 0.1$	$1.2 \pm 1.6$	$0.14 \pm 0.12$	Bencardino et al. (2019)
Monte Curcio - IT (2016)	High-altitude	$12.4 \pm 8.1$	$7.0 \pm 3.1$	1.43	0.12	1.09	0.12	Moretti et al. (2021)
Amman – JO (2018–2019)	Urban background	$64 \pm 39$	$47 \pm 32$	$6.5 \pm 3.0$	$1.9 \pm 1.1$	$5.9 \pm 2.8$	$1.7 \pm 1.1$	Hussein et al. (2022)
Barcelona - SP (2005–2006)	Urban background	45	29	5.6	2.2	5.5	2.3	Perez et al. (2008)
Thessaloniki - GR (2011–2012)	Urban traffic	$51.1 \pm 14$	$31.2 \pm 11.4$	$11.3 \pm 5.03$	$6.56 \pm 2.14$	$8.44 \pm 4.08$	$5.29 \pm 1.54$	Samara et al. (2014)
	Urban background	$33.7 \pm 15.2$	$23.5 \pm 12.3$	$6.62 \pm 4.59$	$0.93 \pm 0.61$	$5.72 \pm 4.36$	$0.69 \pm 0.39$	
Birkenes Observatory - NW (2001–2018)	Regional supersite	6.1	3.5	0.85	0.10	0.65	0.07	Yttri et al. (2021)

$\text{EC}_{\text{PM}_{2.5}}$  (Samara et al., 2014). Also, OC values both in  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  were comparable with previous evidence at urban background sites.

Fig. 1 reports average yearly and monthly values of EC and OC in both  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ . Results showed a decreasing trend of concentrations during the three-years period 2017–2020, especially for  $\text{PM}_{2.5}$ , however, this reduction was only limited from 2018 to 2019 for  $\text{PM}_{10}$ . The trend is relatively small if compared with the interquartile ranges. The monthly variability of OC and EC in  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , together with PM concentrations, demonstrated the highest concentrations from October to March, following an already observed seasonal trend at this site interpreted as an increased contributions of biomass burning and traffic sources during the cold seasons (Cesari et al., 2018b). The seasonal trend of the contributions of different natural and anthropogenic sources to  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  in this area was investigated by using the Positive matrix Factorization (PMF) receptor model in the recent work

of Potì et al. (2025) for the year 2022. Results showed that combustion sources (i.e. biomass burning and road traffic) had the mentioned seasonal trend with biomass burning mainly impacting on water-soluble organic carbon and road traffic mainly impacting on water-insoluble organic carbon. The same seasonal trend was observed for the period 2013–2014 (Cesari et al., 2018a) and for the period 2015–2016 (Giannossa et al., 2022). Thereby, this is a trend spreading over several years of the dataset analysed here. A first little relative increase of carbonaceous concentrations (OC and EC in both fractions) was also observed from July to September, probably as a consequence of the frequent wildfires and biomass burning (that typically are used as agricultural practice in these areas) during the summer periods (Contini et al., 2010). Finally, the minimum EC and OC concentrations appeared in spring (i.e., in May) in conditions of no domestic heating and with limited photochemistry activity compared to summer.

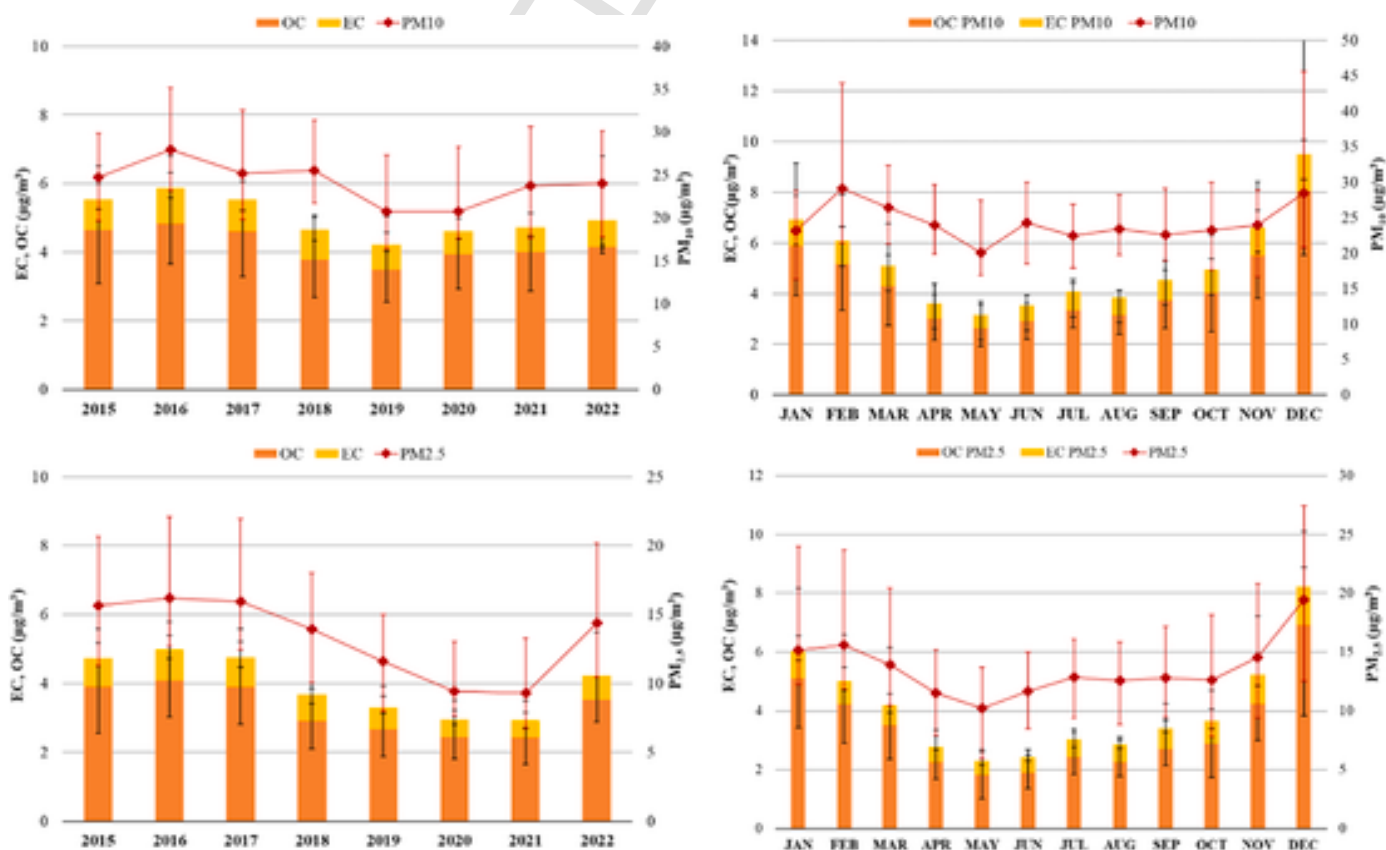


Fig. 1. Yearly (left) and monthly (right) trend of OC and EC both in  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  fractions. Top panel refers to  $\text{PM}_{10}$  and bottom one to  $\text{PM}_{2.5}$ . Error bars represent interquartile range.

It is known that the EC-OC correlation can furnish information regarding potential common carbonaceous aerosol sources (Khan et al., 2012; Na et al., 2004; Yang et al., 2011). The time series of this study showed a very good correlation was observed between EC and OC with  $R^2$  values of about 0.7, for both size fractions, corresponding to Pearson correlation coefficients between 0.84 ( $PM_{2.5}$ ) and 0.85 ( $PM_{10}$ ). In general, the mean OC/EC values (5.6 and 4.8, in  $PM_{10}$  and  $PM_{2.5}$ ) (Table 1) fall in the range of Italian urban background sites (Sandrini et al., 2014), and elsewhere in Europe (Luoma et al., 2021; Mbengue et al., 2018, 2021; Paraskevopoulou et al., 2014; Plaza et al., 2011). The monthly variability of the ratios  $PM_{2.5}/PM_{10}$ ,  $OC(PM_{2.5})/OC(PM_{10})$ ,  $EC(PM_{2.5})/EC(PM_{10})$ , and  $OC/EC$  for the two size fractions are reported in Fig. 2. All these ratios have a marked seasonality with larger values found during autumn/winter months. This suggest that the trend may be due to larger contributions of combustion sources during the cold months due to both traffic and biomass burning, as previously mentioned. It is interesting to observe that the ratio  $PM_{2.5}/PM_{10} < OC(PM_{2.5})/OC(PM_{10}) < EC(PM_{2.5})/EC(PM_{10})$  suggesting that the coarse fraction of aerosol (i.e.  $PM_{10-2.5}$ ) is rich of components originating non-combustion sources like sea salt or crustal/resuspended material. The OC has also relevant component in the coarse fraction likely originating from both biogenic emissions and secondary organic aerosol. Instead, EC has the lowest contribution in the coarse fraction suggesting that primary combustion sources are mainly contributing to fine aerosol. The OC/EC ratio presented, for both  $PM_{2.5}$  and  $PM_{10}$ , a marked seasonal variability with lower values during the warm seasons. Moreover, this ratio was larger in the  $PM_{10}$  fraction mainly because of the non-negligible content of OC of the coarse fraction.

SOC was obtained with the methodology described in Section 2.3, differentiating cold and warm period, because of different season-related carbonaceous sources acting in the area (Cesari et al., 2018b). The  $(OC/EC)_{pri}$  was 4.6 and 4.5 for  $PM_{10}$  and  $PM_{2.5}$ , respectively, during

the cold period, and 3.5 ( $PM_{10}$ ) and 3.6 ( $PM_{2.5}$ ) in the warm period. The average SOC concentration was  $1.1 \pm 0.1 \mu\text{g m}^{-3}$  (10.9 % of  $PM_{10}$ ) and  $0.8 \pm 0.1 \mu\text{g m}^{-3}$  (14.3 % of  $PM_{2.5}$ ) in  $PM_{10}$  and  $PM_{2.5}$  fraction during the cold period. While it ranged from  $1.1 \mu\text{g m}^{-3}$  (9.0 % of  $PM_{2.5}$ ) and  $1.4 \mu\text{g m}^{-3}$  (6.3 % of  $PM_{10}$ ) in warmer months, indicating that the SOC was mainly segregated in fine fraction. Monthly SOC and POC patterns (Fig. 3) showed that SOC increased during spring and summer, especially in  $PM_{10}$ . On average, SOC represents 24.3 % of OC in  $PM_{10}$  and 16.8 % in  $PM_{2.5}$ . The ratio SOC/OC showed a clear seasonal trend with higher values during the warm seasons (31 % in  $PM_{10}$  and 19.5 % in  $PM_{2.5}$ ) compared to the cold seasons (17.7 % in  $PM_{10}$  and 14.1 % in  $PM_{2.5}$ ). This suggests that both photochemistry during the warm period and favourable conditions to the condensation or adsorption of volatile organic compounds play a role in SOC formation (Yttri et al., 2007). The ratio OC/PM (%) also showed a relevant seasonal variability, with average winter values of  $20.5 \pm 1.0$  % ( $PM_{10}$ ) and  $26.7 \pm 1.0$  % ( $PM_{2.5}$ ) and average summer values of  $14.5 \pm 0.6$  % ( $PM_{10}$ ) and  $19.4 \pm 0.6$  % ( $PM_{2.5}$ ). Similarly, the ratio EC/PM (%) showed higher winter values ( $4.1 \pm 0.3$  % for  $PM_{10}$  and  $6.0 \pm 0.3$  % for  $PM_{2.5}$ ) compared to summer months ( $3.0 \pm 0.2$  % for  $PM_{10}$  and  $4.6 \pm 0.2$  % for  $PM_{2.5}$ ). The weight of carbonaceous aerosol species was most significant in colder months and mainly segregated in fine particulate matter.

### 3.2. EC vs. eBC

The comparison of daily EC and eBC concentrations was reported in Fig. 4 for the entire period and separating cold and warm seasons. Considering the whole sampling period, eBC average concentrations were strongly correlated with EC (Pearson's coefficient 0.94,  $p < 0.05$ ), with comparable slopes (1.02 and 1.11) and high determination coefficient ( $R^2$ ) of 0.86 and 0.92, for cold and warm period, respectively. Definitely, differences between eBC and EC could be due to various reasons:

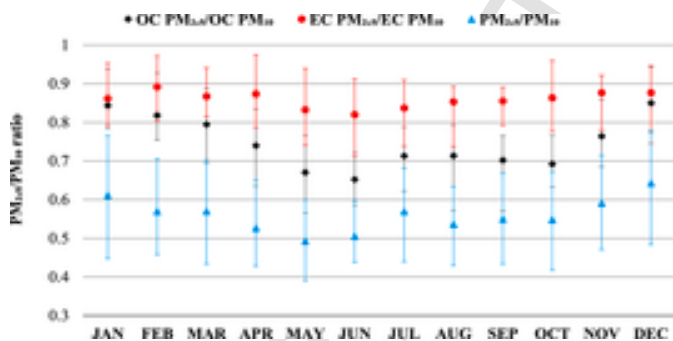


Fig. 2. Monthly OC/EC ratios, separating  $PM_{10}$  and  $PM_{2.5}$ . Error bars represent interquartile range.

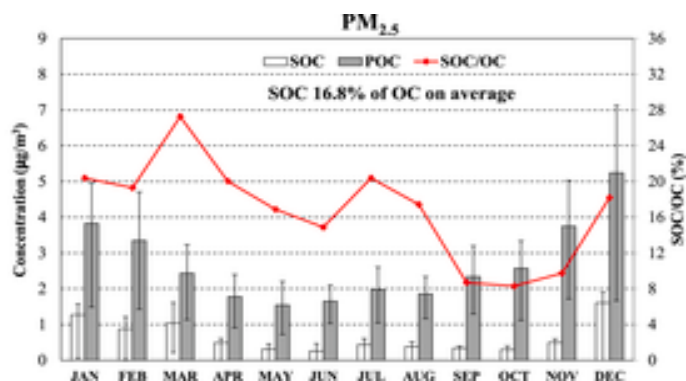
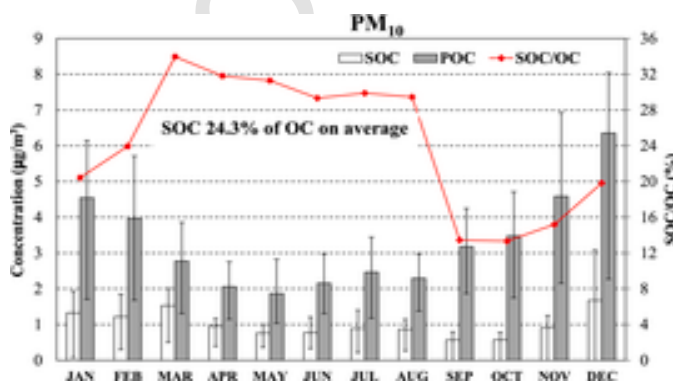
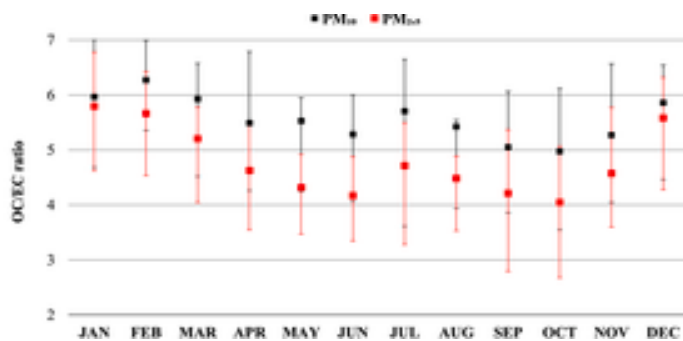


Fig. 3. Monthly SOC and POC in  $PM_{10}$  (left) and  $PM_{2.5}$  (right), with indication of SOC contribution to OC for both fractions. Error bars represent interquartile range.

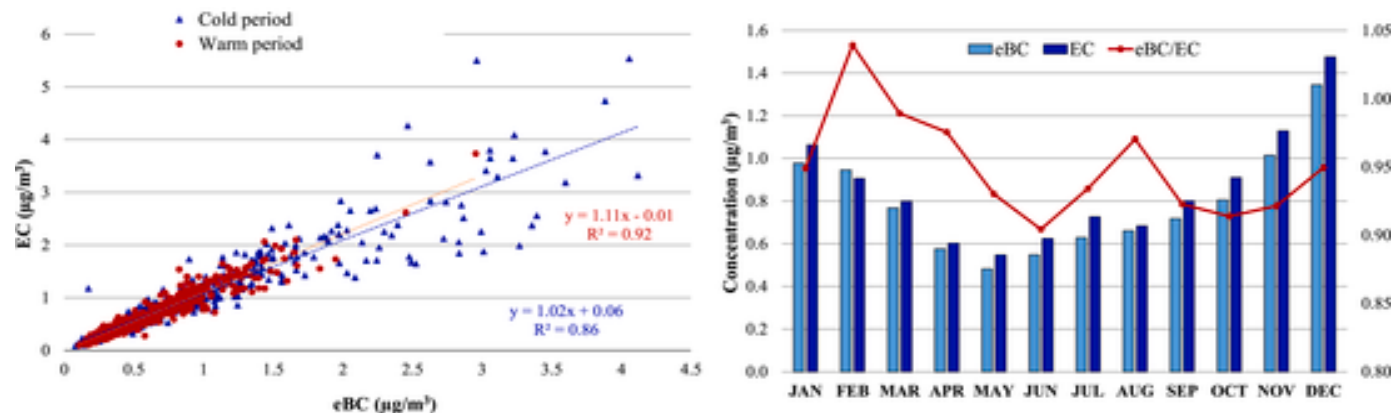


Fig. 4. Scatterplot of EC and eBC concentration (left), with linear fit for cold (blue) and warm (red) period. (right). Monthly trends of eBC and EC concentrations and their ratio.

charring of OC in thermo-optical analysis (Chow et al., 2004; Subramanian et al., 2006), that could be reduced by the removal WSOC by water/methanol (Cheng et al., 2017; Piazzalunga et al., 2011); thermal protocol used, that determines different split points (Giannoni et al., 2016; Merico et al., 2019); presence of BrC, related to biomass burning activities, that could lead to higher eBC measurements by the MAAP (Andreae and Gelencsér, 2006); assumed constant MAC value, actually depending on chemical composition and particle size, location and season (Zhang et al., 2017). The calculated ratio eBC/EC was, on average  $0.95 \pm 0.62$  with limited seasonal variability (Fig. 4).

Fig. S3 shows the weekly trend of eBC as well as the daily trends. There is not an evident weekly pattern on warm seasons and a very limited variability (on average) on the cold seasons with an exclusion of a slightly lower eBC concentration during Sunday likely due to a lower anthropogenic activity in the Campus area. The daily patterns show three interesting aspects. The first is that, in all seasons, the lowest concentrations are observed around midday and the first hours of the afternoon and there is a clear modulation of atmospheric eBC concentrations due the daily evolution of boundary-layer height (BLH). The second aspect is that in early morning it is evident a peak, likely due to traffic emissions that is at 6 a.m. (i.e. the hour between 6 a.m. and 7 a.m.) during the warm season and between 7 a.m. and 8 a.m. during the cold season. The difference in time is related to the daylight-saving time. Instead at night there is an increase of concentration likely due to a mixed effect between the BLH dynamics and evening/night traffic emissions. The third aspect is that the night peak reaches almost the same concentration of the morning one during the warm season but it starts before and it becomes much higher during evening/nocturnal hours during the cold season. This is likely due to a biomass burning (domestic heating) during the cold seasons especially during evening/night.

### 3.3. Analysis of trends

Table 3 and Fig. S4 reported the results of the analysis done for characterisation of long-term trends. Corrected Z values, that measure the deviation of the observed Kendall's Tau from expected values, indicated  $\text{PM}_{2.5}$  as the variable with the largest discrepancy and with the highest negative (Tau =  $-0.16$ ) and positive (Tau =  $0.22$ ), respectively, trend over the period. The trend strength is expressed by the Theil-Sen's estimator obtaining a statistically significant decreasing trend (approximately  $5\% \text{ yr}^{-1}$ ) for  $\text{PM}_{2.5}$  and its OC and EC content. The  $\text{PM}_{10}$  concentration and OC content had a relatively small change per year (approximately  $2\% \text{ yr}^{-1}$ ) that was not statistically significant. EC in  $\text{PM}_{10}$  had a stronger decreasing trend (approx-

Table 3

Values of test statistics for trend analysis (Modified Mann-Kendall test and Theil-Sen's estimator). Statistical significance levels \*\*\* for  $p < 0.001$ , \*\* for  $p < 0.01$ , \* for  $p < 0.1$ .

	Modified Mann-Kendall test			Theil-Sen's estimator		
	Tau	p-value	Corrected Z	Trend [ $\mu\text{g}\text{m}^{-3}\text{yr}^{-1}$ ]	Slope range (min; max) [ $\mu\text{g}\text{m}^{-3}\text{yr}^{-1}$ ]	Change [ $\%\text{yr}^{-1}$ ]
$\text{PM}_{10}$	-0.05	8.30E-03	-2.64	-0.35	-0.82; 0.15	-1.45
$\text{PM}_{2.5}$	-0.16	1.20E-06	-4.85	-0.7***	-1.06; -0.36	-5.18
$\text{OC}_{\text{PM}_{10}}$	-0.05	2.00E-02	-3.04	-0.09	-0.21; 0.05	-2.14
$\text{OC}_{\text{PM}_{2.5}}$	-0.12	8.97E-06	-4.44	-0.17**	-0.28; -0.04	-5.15
$\text{EC}_{\text{PM}_{10}}$	-0.08	7.93E-04	-3.35	-0.03*	-0.05; 0	-3.75
$\text{EC}_{\text{PM}_{2.5}}$	-0.12	6.20E-05	-4.00	-0.03***	-0.06; -0.01	-4.29
$\text{TC}_{\text{PM}_{10}}$	-0.06	9.87E-04	-3.29	-0.12	-0.26; 0.04	-2.35
$\text{TC}_{\text{PM}_{2.5}}$	-0.12	8.84E-06	-4.44	-0.2***	-0.33; -0.06	-5.00
eBC	-0.03	3.80E-02	-2.07	-0.01	-0.03; 0.01	-1.28
MAC	0.22	8.20E-07	4.93	0.32***	0.18; 0.48	2.53
				( $\text{m}^2\text{g}^{-1}\text{yr}^{-1}$ )	( $\text{m}^2\text{g}^{-1}\text{yr}^{-1}$ )	

mately  $3.8\% \text{ yr}^{-1}$ ) compared to OC but with a  $0.05 < p < 0.1$ . The eBC mass concentration at ECO site decreased at an annual rate of about 1.3 %, a lower decrease compared to EC provoking an increasing trend of MAC (Section 3.4). Other traffic and urban background sites were found to exhibit relatively comparable decreasing trends (Matos et al., 2025a, 2025b), especially those directly influenced by primary emissions (i.e., from road traffic) subjected to both European mitigation strategies and local scale actions in urban areas (Savadkoobi et al., 2023).

### 3.4. Determination and temporal variability of MAC in-situ

The complete dataset of absorption coefficient and EC concentration was used to estimate the MAC in-situ, as described in Section 2.4. The value found was  $12.4 \pm 1.2 \text{ m}^2\text{g}^{-1}$  (as geometric mean  $\pm$  standard deviation) and  $12.6 \pm 2.5 \text{ m}^2\text{g}^{-1}$  (as arithmetic mean  $\pm$  standard deviation). Monthly average values of MAC are reported in Fig. 5 together with the inter-annual variability in the different seasons. Monthly pattern of MAC indicated limited variability with slightly larger values during summer and early autumn compared to the other periods. The inter-annual trend of the MAC could be due to a trend of the relative contributions of the dominant sources to  $\text{PM}_{10}$  that could influence the optical properties of aerosol more than the concentrations of EC or to a trend of the mixing state and/or a change of the relative abundance of aged aerosol that could influence optical properties. Ageing processes in the atmosphere by such as condensation, cloud processing or coagu-

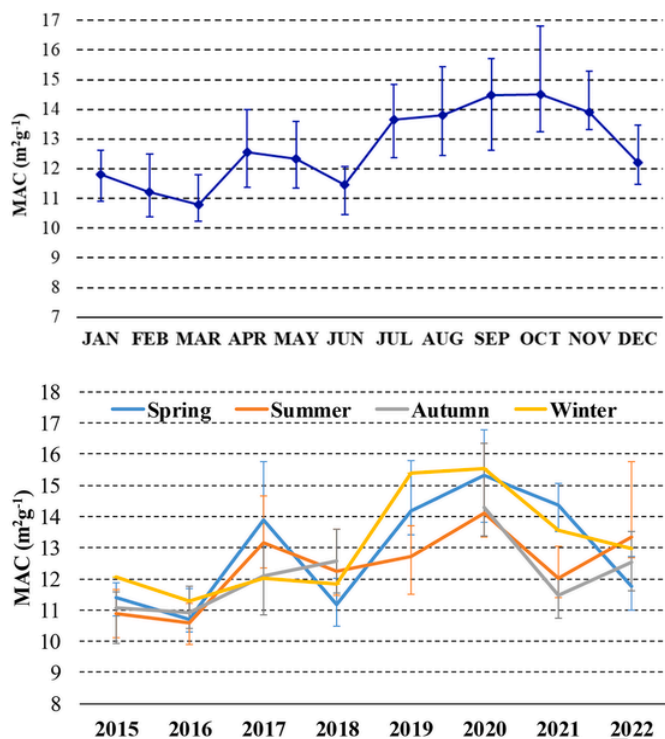


Fig. 5. Monthly trend of MAC for the period 2015–2022 (top) and inter-annual variability of MAC during the different seasons (bottom). Error bars represent interquartile range.

lation may transform externally mixed BC to internally mixed BC (lensing effect). In Zanatta et al. (2016), the mass ratio between EC and non-light-absorbing matter was used as a proxy for the thickness of coatings around the BC cores, finding that MAC increased with increasing values of the coating thickness proxy. Unfortunately, the available dataset for this study does not allow to clearly identify the main responsible process driving the trend of the MAC and further studies would be useful. Potential source of errors in the determination of MAC could be the interference of different light-absorbing organic matter, especially for aerosol having high contribution of biomass burning (Sandradewi et

al., 2008a) or negative interference (up to 15 %) on MAC estimation during desert dust episodes (Karanasiou et al., 2015).

The work of Zanatta et al. (2016) investigated the MAC obtained in several European sites finding a geometric mean value of  $10 \pm 1.3 \text{ m}^2\text{g}^{-1}$ , with a range between  $8.92 \pm 1.65 \text{ m}^2\text{g}^{-1}$  and  $17.3 \pm 1.71 \text{ m}^2\text{g}^{-1}$  (Mediterranean and continental Europe background). The results of Zanatta et al. (2016) for Mediterranean and European continental sites are compared to the values found here in Fig. 6. There is a very good agreement with the values found in Finokalia site (Greece), that is likely more similar to our site being located in the South-Eastern Mediterranean and mostly influenced by long-range transport pollution (sea salt and mineral dust).

The work of Savadkoobi et al. (2024) investigated the MAC at different typologies of sites (urban background, rural, and traffic) in Europe. A comparison of the MAC found here with the results of urban background sites is reported in Fig. 6 showing the comparability of the results at ECO with the range (10–14 m<sup>2</sup>g<sup>-1</sup>) observed by Savadkoobi et al. (2024). Other studies conducted at the wavelength of 637 nm with MAAP instrument, estimated comparable MAC values of  $10.4 \pm 0.2$ ,  $10.9 \pm 3.5$  and  $10.2 \pm 3.2 \text{ m}^2\text{g}^{-1}$ , for regional background (Pandolfi et al., 2011), remote mountaintop (Pandolfi et al., 2014), rural high alpine sites (Liu et al., 2010), respectively. This confirmed the limited spatial variability of MAC, oppositely with chemical and physical transformations of BC (i.e., aging) in atmosphere, that could influence BC light absorption (Singh et al., 2024; Sun et al., 2020; Yuan et al., 2021; Zanatta et al., 2016).

In general terms, differences between values reported in literature may be ascribed to various factors like sources, mixing state of aerosols, atmospheric transformations during transport, aging processes, experimental systematic biases (Bond et al., 2013; Chen et al., 2020; Cozic et al., 2008; Genberg et al., 2013; Laborde et al., 2013; Liousse et al., 1993; Pandolfi et al., 2014; Reche et al., 2011; Yuan et al., 2021).

#### 4. Conclusions

An 8-year-long dataset, from January 2015 to December 2022, was built collecting and analysing measurements of PM<sub>2.5</sub>, PM<sub>10</sub>, and optical and thermo-optical determined carbonaceous fractions at the Environmental Climate Observatory (ECO) in Lecce (Italy). This is an urban background site in south Italy representative of the central Mediterranean area. Relevant results were summarised in the following points.

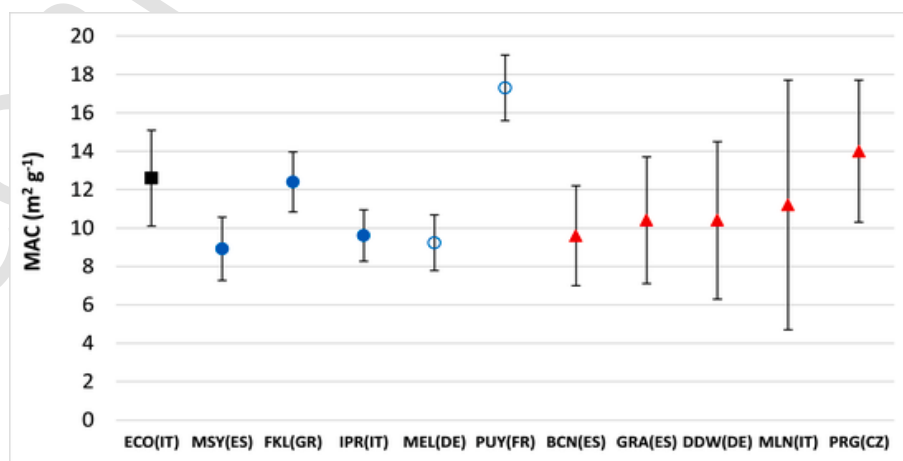


Fig. 6. Comparison of MAC found in this work (black square) with the results found in Zanatta et al. (2016) for Mediterranean sites (blue filled circles) and continental Europe sites (blue empty circles) and in Savadkoobi et al. (2024) for urban background sites. x-axis includes the acronyms of the sites used in the mentioned works and the country in parenthesis.

- PM<sub>2.5</sub> concentrations showed a statistically significant decreasing trend throughout years approximately of 5 % yr<sup>-1</sup>. PM<sub>10</sub> was also slightly decreasing (approximately 2.5 % yr<sup>-1</sup>) but this trend was not statistically significant.
- The OC and EC showed a trend similar to that of PM<sub>2.5</sub> and PM<sub>10</sub>. A statistically significant decreasing trend for carbon content of PM<sub>2.5</sub> (approximately 5 %yr<sup>-1</sup>) and a slightly decreasing trend (approximately 2 %yr<sup>-1</sup> not statistically significant) for carbon content of PM<sub>10</sub> and eBC.
- Regarding the carbon content, a similar seasonal trend was observed for OC and EC with larger concentrations during the cold seasons suggesting a potential contribution of combustion sources. The seasonal variability of PM and carbonaceous species concentration is larger than the long-term variability. The OC/EC ratio values for PM<sub>2.5</sub> and PM<sub>10</sub>, typical for urban background sites, confirmed the presence of a potential contribution from biomass burning (for domestic heating and agricultural practices) even larger than that of fossil fuel (i.e., traffic) at this site.
- SOC was present in both the cold and the warm periods. The fraction of SOC in total OC was slightly larger during the warm period suggesting that photochemistry has a relevant role on SOC formation, however, meteorological conditions during autumn and winter may be favourable to the condensation or adsorption of volatile organic compounds.
- eBC<sub>raw</sub> and EC concentrations were well correlated (Pearson's coefficient 0.94, p < 0.05). The calculated ratio eBC<sub>raw</sub>/EC was, on average, 1.82 ± 0.36, and the same ratio computed with eBC (re-calculated by using MAC in-situ) was 0.95 ± 0.62. Weekly trend of eBC showed a little increase on Wednesday and Thursday, with the minimum recorded on Sunday, due to almost absent road traffic at the site.
- The value of MAC in-situ was estimated as 12.4 ± 1.2 m<sup>2</sup>g<sup>-1</sup> (as geometric mean) and 12.6 ± 2.5 m<sup>2</sup>g<sup>-1</sup> (as arithmetic mean), in agreement with values found for other similar measuring stations of the ACTRIS network. MAC showed a limited seasonal trend with larger values during summer and early autumn and a statistically significant increasing long-term trend of approximately 2.5 %yr<sup>-1</sup>.

Finally, the use of harmonized procedures for data collection and processing is highly recommended to facilitate comparability among different sites thus providing a solid research base to use for definition of future mitigation policies.

#### CRedit authorship contribution statement

**Eva Merico:** Writing – original draft, Formal analysis, Conceptualization. **Daniela Cesari:** Writing – review & editing, Investigation, Formal analysis. **Adelaide Dinoi:** Writing – review & editing, Investigation, Formal analysis. **Serena Poti:** Writing – review & editing, Investigation, Formal analysis. **Antonio Pennetta:** Writing – review & editing, Investigation, Formal analysis. **Ermelinda Bloise:** Writing – review & editing, Investigation, Formal analysis. **Daniele Contini:** Writing – review & editing, Formal analysis, Conceptualization.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apr.2025.102668>.

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