



# Relationships between air particulate matter capture efficiency and leaf traits in twelve tree species from an Italian urban-industrial environment

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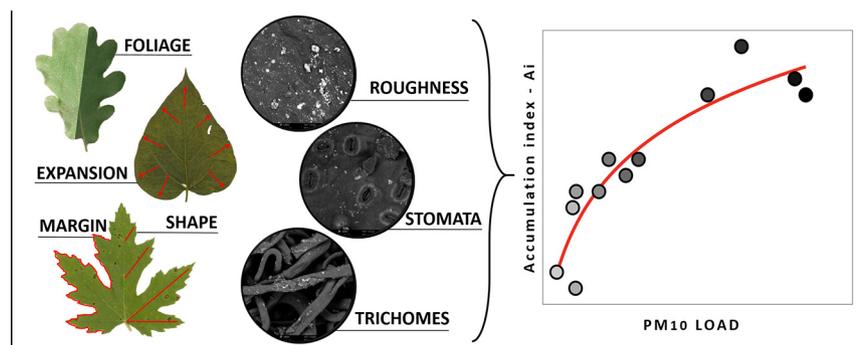
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## HIGHLIGHTS

- Nature-based Solutions efficiency in air quality mitigation depends on plant species.
- PM load on twelve tree species is quantified by SEM/EDX technique and V/F method.
- PM loads are related to leaf micro and macro morphological characteristics.
- A species-specific morphological-based index for PM removal efficiency is presented.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Air pollution in the urban environment is widely recognized as one of the most harmful threats for human health. International organizations such as the United Nations and the European Commission are highlighting the potential role of nature in mitigating air pollution and are now funding the implementation of Nature-Based Solutions, especially at the city level. Over the past few decades, the attention of the scientific community has grown around the role of urban forest in air pollution mitigation. Nevertheless, the understanding on Particulate Matter (PM) retention mechanisms by tree leaves is still limited. In this study, twelve tree species were sampled within an urban park of an industrial city. Two techniques were used for leaf analysis: Vacuum/Filtration and Scanning Electron Microscopy coupled with Energy Dispersive X-ray spectroscopy, in order to obtain a quali-quantitative analysis of the different PM size fractions. Results showed that deposited PM loads vary significantly among species. Different leaf traits, including micro and macromorphological characteristics, were observed, measured and ranked, with the final aim to relate them with PM load. Even if no significant correlation between each single leaf characteristic and PM deposition was observed ( $p > 0.05$ ), multivariate analysis revealed relationships between clusters of leaf traits and deposited PM. Thus, by assigning a score to each trait, an Accumulation index (Ai) was calculated, which was significantly related to the leaf deposited PM load ( $p \leq 0.05$ ).

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## 1. Introduction

Many epidemiological studies evidenced the highly detrimental effects of Particulate Matter (PM) at different dimensional classes (Englert, 2004). In Europe, as well in Asia or North America, there is a growing attention to the associated high-mortality factor across PM size, of which is showing no sign of decline in the near future (Goldberg et al., 2013; Lu et al., 2015; Maté et al., 2010; Yorifuji et al., 2016). Since 2015, the United Nations (UN) have been giving clear indications to enhance the environmental and human conditions, by drawing up the 17 Sustainable Development Goals (SDGs) to be adopted before 2030 (UN General Assembly, 2015). Unsurprisingly, a number of these ambitious goals involves or is relevant to urban environments and air pollution. To face this challenge, the European Commission (EC) has been adopting strategies in the last decade, focused on sustainable actions able to mitigate the risks deriving from the exposure of different kinds of pollutants to human populations. A concrete approach is represented by the implementation of Nature-based Solutions (NBS) in urban environments (Raymond et al., 2017; Calfapietra and Cherubini, 2019; <http://ec.europa.eu/environment>). The direction given by the EC is increasing the value and attention given to urban forests and urban trees. The multiple ecosystem services provided by urban trees have been widely described (Escobedo et al., 2011; Livesley et al., 2016). In particular, urban green areas provide important mitigation systems of PM and other air pollutants (Freiman et al., 2006; Liu et al., 2017), and urban parks represent a significant tool, retaining huge amounts of particles upon leaves (Cohen et al., 2014; Fantozzi et al., 2013; Yin et al., 2011). However, improvements in research, application and monitoring of these effective and low-cost solutions are still needed (Baró et al., 2015).

In particular, deposition mechanism of PM on leaves is the object of increasing interest to the international scientific community. In the last few years, scientists have been investigating the differential PM deposition on leaves both in controlled environments (Freer-Smith et al., 2005; Lin et al., 2012; Rouspard et al., 2013; Schaubroeck et al., 2014; Xie et al., 2018), and in field conditions (Baldacchini et al., 2019; Chen et al., 2016; Hofman et al., 2013; Mo et al., 2015). Eddy covariance towers started to measure PM fluxes in urban parks, providing evidence for the relationship between PM deposition and resuspension (Fares et al., 2016; Guidolotti et al., 2017). Models have also been developed to estimate PM deposition on tree leaves and have so far been producing reliable outputs through their implementation in a number of urban case studies (Hirabayashi et al., 2015; Manes et al., 2016; Nowak et al., 2008). Results obtained by this large number of typology of studies vary considerably. Different environmental conditions, species and experimental assumptions strongly influence the quantitative analysis. In order to provide better comparison, and therefore understanding, of the experimental results, it is thus mandatory to investigate, and possibly control, the factors affecting the PM capturing capability of different tree species. To this aim, several recent studies have been focusing on the complex relationship between PM deposition and leaves morphological variability at micro and macro scale, such as shape, margin and surface structures (Chen et al., 2016; L. Chen et al., 2017; Chiam et al., 2019; Tomasevic and Anicic, 2010; Weerakkody et al., 2018a).

In this research, leaves from different tree species were sampled in an urban park. Quali-quantitative analysis of PM was performed through two techniques (vacuum filtration: V/F; and scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy: SEM/EDX). V/F is a widely used technique for quantitative analysis of PM deposition on leaves in urban environments, while SEM/EDX is primarily used for PM density measurements on the leaf surface and/or PM chemical characterization (Dzierzanowski et al., 2011; Popek et al., 2013; Wilkinson et al., 2013; Przybysz et al., 2014; Sgrigna et al., 2015; Terzaghi et al., 2013; Shi et al., 2017; Zhang et al., 2017a). Recent researches have been applying SEM/EDX technique also for quantitative analysis of PM on leaves surface (Baldacchini et al., 2017, 2019).

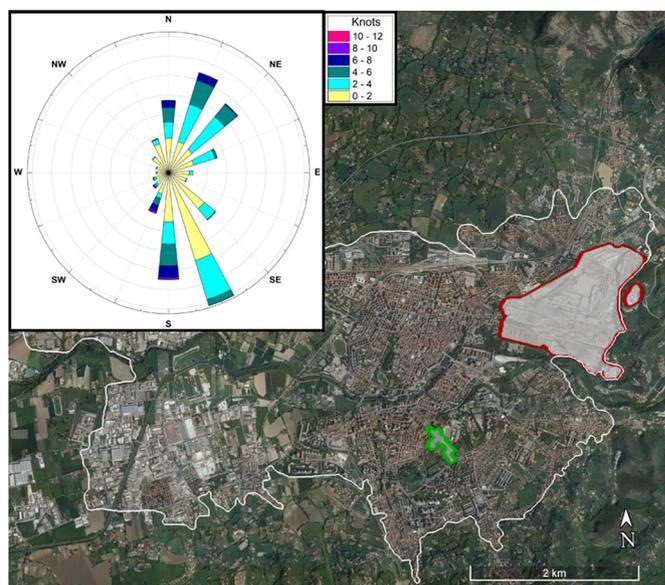
Strengths and weaknesses of the two approaches have been discussed, and the accuracy of SEM/EDX technique to study PM is confirmed (Baldacchini et al., 2017, 2019; Shao et al., 2019; Weerakkody et al., 2018b). Thereafter, leaf morphologies were assessed through both direct macro-observations and SEM imaging magnifications, and classified according to the literature (J. Chen et al., 2017; Kardel et al., 2013; Mo et al., 2015; Wang et al., 2013; Weerakkody et al., 2018b). A multivariate approach (by Principal Component Analysis: PCA) evidenced the relationship among leaf traits and PM amounts. Finally, leaf traits were ranked, depending on their influence on PM deposition, and combined in the newly introduced Accumulation index (Ai), which is species-specific and highly correlated with the PM deposition load.

The leaf PM load has been previously discussed in connection with single leaf characteristic (Weerakkody et al., 2018a; Yang et al., 2014). Nevertheless, as suggested by Leonard et al. (2016), the combination of different leaf traits is a key factor to furthering the investigation into PM deposition. Describing the Ai, or using similar approaches, could be then considered a key tool for future assessment of PM mitigation strategies; providing stakeholders, such as policy makers and urban planners with a valuable instrument in the evaluation, management and/or design of NBS and Green Infrastructure.

## 2. Materials and methods

### 2.1. Study area and sampling

An urban park within the industrial city of Terni (approx. 112,000 inhabitants), located in the Umbria region in central Italy, has been chosen as study area (Fig. 1). This site was selected since the city is the most industrialized in the region, characterized by high concentration levels of PM with diameter lower than 10  $\mu\text{m}$  and 2.5  $\mu\text{m}$  (PM10 and PM2.5, respectively) in the air (Sgrigna et al., 2015). In particular, based on the data reported by the local environmental agency Arpaumbria ([www.arpaumbria.it](http://www.arpaumbria.it)), the 2017 annual average concentrations of PM10 and PM2.5 recorded by the environmental control station closest to the study area (named "Le Grazie") were 34  $\mu\text{g}/\text{m}^3$  and 24  $\mu\text{g}/\text{m}^3$ , respectively, while the EU reference values for annual concentration are 40  $\mu\text{g}/\text{m}^3$  for PM10 and 25  $\mu\text{g}/\text{m}^3$  for PM2.5. Moreover, for 48 days in the whole year, the daily average concentration of PM10 exceeded the limit of 50  $\mu\text{g}/\text{m}^3$ , which is higher than the EU reference value of



**Fig. 1.** Le Grazie park (green patch) and industrial area of the steel factory (red patch) in the city of Terni (Italy). Inset: wind rose by mean wind speed values calculated over the 6 months prior to sampling. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

maximum 35 days per year (<https://ec.europa.eu/environment/air/quality/standards.htm>).

The city is set in a valley, surrounded by two main mountain chains along with a smaller one (Cattuto et al., 2002). It is hypothesized that the presence of industrial hubs and urban activities within the valley has been contributing to a large number of recorded exceedances of air quality standards in PM concentration throughout the year, in particular over stable atmospheric conditions (i.e., during winter and summer times [[www.arpa.umbria.it](http://www.arpa.umbria.it)]).

Among the industrial hubs included in the Terni urban area (Capelli et al., 2011), we focused on the “Thyssen Krupp AST” (Acciai Speciali Terni – Special Steels Terni) steel factory, located in the western part of the city, occupying an area of approximately 158 ha (in red in Fig. 1). Twelve tree species were sampled, located within an urban park downwind to the steel factory (in green in Fig. 1). The selected species, described in Table 1, are: *Acer saccharinum* L., *Catalpa bignonioides* Walter, *Cedrus atlantica* (Endl.) Manetti ex Carrière, *Celtis australis* L., *Magnolia grandiflora* L., *Platanus acerifolia* (Aiton) Willd., *Populus nigra* L., *Populus tremula* L., *Prunus cerasifera* Ehrh., *Quercus pubescens* Willd., *Robinia pseudoacacia* L. and *Tilia cordata* Mill.

For each species, three branches were collected from the same tree. The branches were gathered from the exterior of the crown, at 6 m height from the ground. Successively, leaves were separated for both V/F and SEM/EDX analysis. All branches were collected at similar cardinal direction, in order to select leaves under similar wind conditions, e.g. influenced by the same pollution sources: east-north-east side of the crown, which is the direction of major steel factory influence in “Le Grazie” park (Sgrigna et al., 2016). Leaf sampling was conducted on April 15th, May 15th and June 15th, in 2017. The most important rain event preceding the final sampling occurred in May 20th, 2017, i.e. 25 day before the sampling, with a total amount of rain of 19 mm. Due to the long time distance between this rain event and the sampling date, it is expected to have no impact on the present study. Leaves from the final sampling (June 2017) were used to perform the PM quantitative analysis and to characterize the leaf traits, while leaves sampled in April and May were used to assess leaf expansion (expressed in terms of percentage of the mean single leaf area under expansion compared with the totally expanded single leaf area).

## 2.2. Vacuum filtration: quantification of leaf deposited PM

Between 300 and 700 cm<sup>2</sup> of total leaf area per species were selected from sampled branches; the procedure has been previously described by Sgrigna et al. (2015). The number of leaves per each species varied depending on the different peculiar dimensions (from <10 leaves for *C. bignonioides*, up to >100 for *C. atlantica*). Leaves on each branch were washed in 500 ml of micro-distilled water, with the leaves being singularly brushed by a synthetic paintbrush. The solution with the suspended particles was then filtered by a plastic sieve (100 µm porosity), in order to remove the coarsest material. Finally, the washing solution was forced through filters by a vacuum filtration system.

PM > 10 µm was removed from the solution using filters with a porosity between 10 and 13 µm (Anoia S.A., Barcelona, code 1250). Coarse PM (between 2.5 and 10 µm) was collected on filters with a porosity between 2 and 4 µm (Anoia S.A. Barcelona, code: 1244); then fine PM fraction (between 0.2 and 2.5 µm) was collected on nitrocellulose membrane (0.2 µm porosity - Advanced Microdevices Pvt. Ltd. India). All filters were dried in oven at 65 °C for 60 min, and then stabilized in glass boxes for 30 min before weighing. Filters had been pre-weighed through precision balance with four decimal places (Sartorius – model r180d) and were re-weighed after drying. The arithmetic difference between the second and the first weight (expressed in µg) is the PM deposition relative to the analyzed amount of leaves from each branch. To obtain the normalized value expressed in µg/cm<sup>2</sup> (the PM load), PM mass was divided by the total leaf area of leaves from the same branch. Leaf area, here, has been considered as double sided. Foliar measurements for the average leaf area have been performed through ImageJ, an open source software (Schneider et al., 2012), upon a direct leaf scan. PM deposition values were then obtained per PM<sub>0.2–2.5</sub> and PM<sub>2.5–10</sub> size fractions. The PM<sub>0.2–2.5</sub> load was assumed to be representative of the total PM<sub>2.5</sub>, while PM<sub>10</sub> loads were obtained by summing the PM<sub>0.2–2.5</sub> and PM<sub>2.5–10</sub> size fraction loads.

## 2.3. Scanning electron microscopy imaging: characterization of leaf deposited particulates and leaf surface micromorphology

SEM imaging was performed on three different leaves per species, each leaf being selected from a different sampled branch, randomly chosen. For each leaf, two portions of 0.5 cm<sup>2</sup> were cut in a comparable blade specific portion, considering the varying shapes of leaves for different species: in the mid part of lamina area, close to the main rib, and/or avoiding the presence of large veining. The two 0.5 cm<sup>2</sup> portions were used for the analysis of the abaxial and the adaxial leaf surface, respectively.

A Phenom ProX (Phenom-World, The Netherlands) scanning electron microscope was used, equipped with X-ray analyzer and a charge-reduction sample holder suited for biological samples. Leaf portions were mounted within the sample holder by using double coated carbon conductive PELCO Tabs (Ted Pella, Inc.) and then fluxed with compressed air. SEM Imaging was performed in backscattered electron configuration, with an incident electron energy of 5 keV, in order to limit the surface charging. The sample surface was randomly imaged by 150 µm wide scans (approximately 1800× magnification), at a resolution of 1024 × 1024 pixels. These imaging conditions have been chosen as the best compromise among a large scanning area (which means high number of observed particles per image) and a minimum particle resolution of 0.2 µm, which is comparable with the cutoff of the filters used in V/F. For each leaf portion, five images were acquired, resulting in a total of 10 images per replicates (5 per leaf portion), and 30 images per species.

Images acquired through SEM were used for particle counting and measuring, and for observing leaf micromorphological traits. PM can

**Table 1**

Scientific and common names and main botanical characteristics of the sampled tree species. In the foliage description, “DEC” is for “deciduous” and “EVE” for “evergreen”.

Tree species	id code	Common name	Family	Foliage	Leaf macro-characteristics
<i>Acer saccharinum</i>	A. s.	Silver maple	Aceraceae	DEC	Simple and palmately veined, with deep angular notches between the five lobes
<i>Catalpa bignonioides</i>	C. b.	Indian-bean-tree	Bignoniaceae	DEC	Large and heart shaped, smooth margins
<i>Cedrus atlantica</i>	C. a.	Atlas cedar	Pinaceae	EVE	Needle-like, smooth margins, concentrated at the proximal end of the long shoots
<i>Celtis australis</i>	C. au.	Mediterranean hackberry	Ulmaceae	DEC	Narrow and sharp-toothed, rugose above and tomentose below
<i>Magnolia grandiflora</i>	M. g.	Southern magnolia	Magnoliaceae	EVE	Simple, broadly ovate, smooth margins, stiff, scurfy underneath with pubescence
<i>Platanus acerifolia</i>	P. a.	Planetree	Platanaceae	DEC	Deeply lobed and palmate or maple-like
<i>Populus nigra</i>	P. n.	Black poplar	Salicaceae	DEC	Diamond-shaped or triangular, slightly lobed margin
<i>Populus tremula</i>	P. t.	Aspen	Salicaceae	DEC	Nearly round, slightly wider than long, coarsely toothed margin
<i>Prunus cerasifera</i>	P. c.	Cherry plum	Rosaceae	DEC	Ovate shape, serrate margins
<i>Quercus pubescens</i>	Q. p.	Downy oak	Fagaceae	DEC	Divided into pairs of deep or shallow blunt lobes, usually divided into few sublobes
<i>Robinia pseudoacacia</i>	R. p.	Black locust	Fabaceae	DEC	Compound, odd pinnate, roughly paired leaflets, smooth margins
<i>Tilia cordata</i>	T. c.	Small-leaved lime	Tiliaceae	DEC	Ovate or cordate, serrate margin and soft texture

be easily distinguished as bright particles on the leaf surface, since the contrast of SEM images depends on the elemental composition of the imaged object. Thus, by using Gwyddion v. 2.49, an open source software program (Nečas and Klapetek, 2012), a color threshold was applied to each image, producing a rapid and repeatable selection of particulates against the leaf surface background, obtaining the number of particles per unit leaf area and their size distribution (Baldacchini et al., 2019). The size of particulates was estimated by the equivalent sphere diameter ( $d_{eq}$ ), automatically calculated by the software grain analysis, by averaging the minimum and maximum Feret diameters (Merkus, 2009). SEM images further allow the direct observation of leaf microstructural traits, with different patterns, typologies and abundances. We focused on three micro-morphological characteristics that have been reported to be important for PM leaf deposition: stomata, trichomes and surface roughness. Stomata density has been quantified by counting the number of stomata per unit leaf area in the SEM images. Due to the complexity of the trichome layer, it was not possible to estimate the surface density, as was done for stomata. Instead, the percentage of the leaf area covered by the trichome layer was estimated. Trichome density has been evaluated by measuring the percentage of the leaf area covered by the trichomes, by using the open source software ImageJ (Schneider et al., 2012). The degree of the leaf surface roughness has been further assessed by evaluating three parameters: (a) the type of surface structures, which have been classified as smooth (S), peaks (P), valleys (V), and presence of grooves (G); (b) the percentage of the leaf area covered by these surface structures; (c) the mean width of the grooves (when present). The two latter quantities have been evaluated from the SEM images by using ImageJ (Schneider et al., 2012). Examples of these characteristics are reported in Fig. 2.

#### 2.4. Accumulation index

In order to disclose the relationship between leaf morphology and PM load, measurements and categorized characteristics have been combined to create an index able to describe the combinations of different morphological characteristics positively or negatively related to PM deposition. Beyond the three previously mentioned micromorphological leaf characteristics (surface roughness, stomata density, trichomes density), two further macromorphological characteristic (leaf blade margin complexity and leaf shape) and two macroscopic parameters (leaf growth expansion, LGE, and foliage character) have been identified as driving characteristics for PM capture, according to previous studies (Cai et al., 2017; Liu et al., 2017; Mitchell et al., 2010; Mo et al., 2015; Mori et al., 2018; Shi et al., 2017; Weerakkody et al., 2018a).

For each species, as shown in Fig. 2, the complexity grade of margins has been described as smooth (SM), serrate (SER) or lobate (LOB), while the leaf shape has been classified as elliptical (ELL), compound (COM), acicular (ACI), or palmate (PAL). These characteristics has been merged in one macromorphological characteristic called "Morphology".

The LGE (referred to by relative percentages) at the sampling date has been evaluated by comparing the mean leaf area of leaves used for PM analysis with that of leaves collected, from the same tree, 30 days and 60 days before the final sampling.

Finally, the selected leaf characteristics were ranked, and grading criteria are reported in Table 2: grades ranged between 0 and 3, for each selected characteristic. The higher the effect on the PM capture capability of the specific characteristic, the higher the associated score. Also, values were linked to the variable abundance/morphology, or to the presence/absence of specific characteristics. This is the first study where all characteristics are considered and joined together to create an index. It should be noted that in this study the same weight has been associated to all characteristics. Specifically, high scores have been associated with leaf surfaces with high percentages of rough area (Zhang et al., 2017b), trichome covered area (Räsänen et al., 2013), high stomata density (Liang et al., 2016), or small groove dimensions (L. Chen et al., 2017). Therefore, leaves with serrate/lobate margin and

palmate shape were associated with higher scores (complex leaf shape); leaves with smooth margins and ellipsoid shapes were assigned lower scores (simple leaf shape) (Weerakkody et al., 2018b). Exposure time of leaves to air during development, here described through LGE, has also been considered as an influencing factor in PM deposition: high scores were associated with species with unripe development of leaves. Similarly, evergreen species were differentiated from deciduous ones with higher scores (Cai et al., 2017).

Scores corresponding to the different characteristics were summed in order to obtain a cumulative value for each species, here defined as Accumulation Index ( $A_i$ ):

$$A_i = (Rt + Ra + Gd + Sd + Td + Mc + Ls + LGE + F)$$

Where the micro characteristics are represented by: roughness type (Rt), rough area (Ra), grooves dimension (Gd), stomata density (Sd) and trichome covered area (Td); while the macro characteristics are margin complexity (Mc), leaf shape (Ls), leaf growth expansion (LGE) and foliage (F).

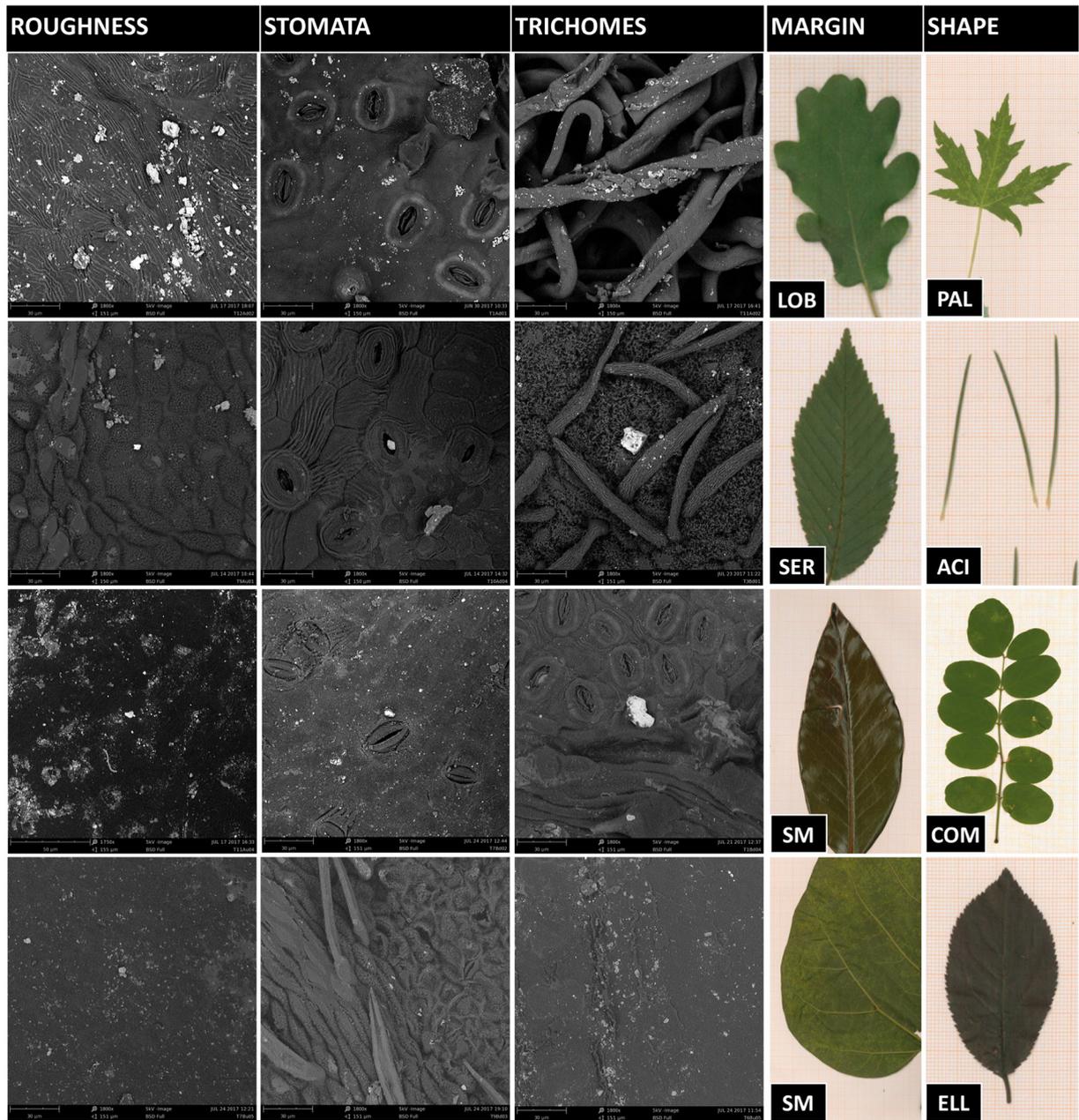
#### 2.5. Energy-dispersive X-ray spectroscopy (EDX) for quali-quantitative PM assessment

EDX was used to obtain the elemental composition of leaf surface deposits. To provide particulate elemental analysis, five points of the leaf surface under investigation were randomly selected. Then, for each point, a high-resolution SEM image was acquired, at 50  $\mu\text{m}$  lateral size and 15 KeV of electron accelerating voltage. Ten particles were randomly chosen for each image, resulting in 300 particles per sampled species (50 particles per leaf side, for 2 sides and 3 replicate leaves). The EDX spectrum of the selected particles was obtained by positioning the laser beam in the particle center. The main elements identified in the particles were C, N, O, F, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Mo, Sn, Sb, Ba, W, Bi. Particles showing only C, N and O were excluded from the analysis, due to the uncertainty of EDX in quantifying those light elements, difficulties introduced by the use of leaves as substrates, and the presence of other organic materials like cellulose (Sgrigna et al., 2016; Wilkinson et al., 2013). A first estimation of the elemental composition of the total leaf deposited PM, per replicate, was obtained by calculating the weighed volume percentage ( $W_{\%x}$ ) occupied by each element x over the selected particles, as previously described by Baldacchini et al., 2017 and Sgrigna et al., 2016.

Then, for each replicate, the  $W_{\%x}$  obtained per each element x were multiplied by the corresponding elemental atomic mass per volume (also known as solid-state density, taken from <https://www.webelements.com/periodicity/density/>) and by the particle volume per unit leaf area. The sum of obtained quantities is an estimation of the amount of leaf deposited PM per unit leaf area (Baldacchini et al., 2019). PM deposition values were calculated for PM0.2–1, PM1–2.5 and PM2.5–10 size fractions. Total PM10 and PM2.5 loads were then obtained by summing the size fractions.

#### 2.6. Statistical analysis

Statistica software, version 7 (TIBCO Software Inc., 2018) was used for statistical analysis in this study. PM weight variability on different species, for different PM sizes (PM10 and PM2.5 for V/F and PM10, PM2.5 and PM1 for SEM/EDX), was analyzed using one-way ANOVA, following confirmation of normality using the Shapiro-Wilk test. Significant differences, thus the identification of homogeneous groups, were identified using a post-hoc test, namely a multiple comparison test, which was used to determine the significant differences among the twelve species in an analysis of variance setting. Fisher LSD – test revealed the homogeneous groups, which are evidenced by letters: species that share the same letter, thus the same group, do not present significant differences ( $p < 0.05$ ).



**Fig. 2.** Selected images of micro (surface roughness, stomata and trichomes) and macrostructural (margin complexity and leaf shape) characteristics of sampled leaves. For each micromorphological trait, four examples are shown, corresponding to species with a different complexity degree of the specific characteristic. For each macromorphological characteristic, four examples are reported: margin complexity has been classified as smooth (SM), serrate (SER) and lobate (LOB); the observed leaf shapes have been classified as acicular (ACI), elliptical (ELL) and palmate (PAL). According to the figure, the shown species (by following the order: roughness, stoma, trichomes, margin, and shape) are, from upper to lower row: first row - P. c., C. au., M. g., Q. p., A. s.; second row - R. p., P. a., A. s., C. au., C. a.; third row - M. g., P. n., C. au., M. g., R. p.; fourth row - P. n., R. p., C. a., C. b., P. c. (species id codes are reported in Table 1).

**Table 2**

Associated scores and criteria assignment corresponding to foliar micro and macro morphological characteristics. Stomata and trichomes densities are estimated as reported in Materials and Methods. The surface structures are classified as smooth (S), peaks (P), valleys (V), grooves (G). The margin complexity are described as smooth (SM), serrate (SER) and lobate (LOB). The leaf shape is classified as elliptical (ELL), compound (COM), acicular (ACI), palmate (PA). The leaf growth is quantified through the estimation of the Leaf Growth Expansion (LGE, see Materials and Methods). The foliage character can be either deciduous (DEC) or evergreen (EVE).

SCORE	Micromorphology					Macromorphology			
	Roughness		Stomata	Trichomes	Morphology		LGE	Foliage	
	Type	Area (%)			Margin	Shape			
			Density (#/mm <sup>2</sup> )	Covered area (%)		% (days before sampling)			
0	S	<1	>2	<100	< 1	SM	ELL	<50% (30 days)	DEC
1	P/V	1–33	1.5–2	100–250	1–33	SER LOB	COM	>50% (30 days)	/
2	G/V	33–66	1–1.5	250–350	33–66	/	ACI PAL	>90% (30 days)	/
3	G	>66	<1	>350	>66	/	/	>70% (60 days)	EVE

The comparison between deposition values ( $\mu\text{g}/\text{cm}^2$ ) obtained by SEM/EDX and V/F was performed through regression analysis.

PCA was applied to associate specific morphological characteristics with quality (in terms of PM size fraction) and quantity (in terms of percentages per dimensional class) of accumulated particles on leaves.

### 3. Results

#### 3.1. Leaf deposited PM load

The average value ( $\pm$  standard error) of deposited PM10 and PM2.5, as obtained by the two techniques over the twelve species, are quite similar. Indeed, PM10 mean load from V/F is  $3.1 \pm 0.4 \mu\text{g}/\text{cm}^2$ , while for SEM/EDX is  $5.2 \pm 1.1 \mu\text{g}/\text{cm}^2$ . The average value of PM2.5 from V/F is  $1.3 \pm 0.2 \mu\text{g}/\text{cm}^2$ , almost exactly the same as  $1.3 \pm 0.3 \mu\text{g}/\text{cm}^2$  obtained by SEM/EDX.

The particle load for all species and PM fractions, measured with both techniques are summarized in Table 3, grouped according to the ANOVA results (see Materials and Methods).

The twelve species showed different PM deposition quantities on their leaves, and the two techniques showed comparable results for six out of twelve species, but for the remaining species it is rather different (or even opposite) when V/F and SEM/EDX results are compared. V/F PM10 ranges from group "a" (which includes only *P. tremula*, with deposition value of  $0.89 \pm 0.24 \mu\text{g}/\text{cm}^2$ ) to group "f" (which is represented only by *T. cordata* that shows a PM10 deposition of  $6.69 \pm 0.32 \mu\text{g}/\text{cm}^2$ ). Similarly, it is observed for PM2.5 deposition by V/F: *P. tremula*, with deposition value of  $0.33 \pm 0.03 \mu\text{g}/\text{cm}^2$ , represents the species with the lowest load (group "a"), while species with the highest load (group "e") still includes *T. cordata* (having a PM2.5 deposition of  $2.99 \pm 0.19 \mu\text{g}/\text{cm}^2$ ). On the other hand, the species showing the maximum and minimum SEM/EDX PM load are completely different: the lowest SEM/EDX PM10 load ( $1.10 \pm 0.25 \mu\text{g}/\text{cm}^2$ ) was obtained for *T. cordata* (group "a"), while *P. acerifolia* and *P. cerasifera* (having PM10 load of  $11.96 \pm 0.53 \mu\text{g}/\text{cm}^2$  and  $11.49 \pm 1.92 \mu\text{g}/\text{cm}^2$ , respectively) are the species with the highest loads (group "e").

However, for six out of the twelve species, the same ANOVA classification was concluded at all PM size fractions with both techniques: *C. bignonioides*, *C. australis* and *Q. pubescens* always show low PM values ("a" and "b" classes), *A. saccharinum* and *M. grandiflora* always belong to the intermediate classes ("c" and "d" classes), while *P. cerasifera* shows always a high PM deposition ("e" or "d" class).

The average species-specific PM10 load values obtained by the two techniques are compared in Fig. 3, through a scatterplot graph. When data from the two techniques are related through regression analysis,

no significant relationship is found. However, if the dataset is restricted to the six species mentioned above (namely, *A. saccharinum*, *C. bignonioides*, *C. australis*, *M. grandiflora*, *P. cerasifera* and *Q. pubescens*), a significant relationship exists between the two techniques ( $r^2 = 0.77$ , regression line is shown in Fig. 3). On the contrary, two species (*P. tremula* and *P. acerifolia*) show considerably lower PM10 loads when measured by V/F than by SEM/EDX, while four species (specifically, *C. atlantica*, *R. pseudoacacia*, *P. nigra* and *T. cordata*) show the opposite results.

#### 3.2. Micro and macromorphological traits of leaves

Surface roughness, stomata, trichomes density and LGE results are reported in Table 4, together with macromorphological traits of leaves (leaf blade margin complexity, leaf shape and leaf foliage).

In the majority of observed species, the dominant surface structure was Grooves. Groove covered area was measured as abundant ( $> 66\%$  of observed area) for four species: *P. acerifolia*, *P. cerasifera* and *P. tremula*, while 50% of observed species showed absence or a very low presence of rough area. The surface roughness was totally absent in *M. grandiflora* and *C. atlantica*, and in two cases (*P. nigra* and *T. cordata*) the measurement of this trait was impossible due to the large quantity of honeydew on the leaf surface. Groove dimensions varied from a minimum of  $0.66 \mu\text{m}$  up to a maximum of  $1.72 \mu\text{m}$ .

The abaxial side of leaves showed a significant variability of stomata and trichomes cover. Number of stomata per  $\text{mm}^2$  varied between 44 and 435. *A. saccharinum* showed the highest presence of these structures, followed by *P. cerasifera* ( $>350$  per  $\text{mm}^2$ ). The lowest values recorded for this characteristic were observed in *P. nigra* and *R. pseudoacacia* (97.2 and 44.5 stoma/ $\text{mm}^2$ , respectively). Due to the complexity of the trichome layer, it was not possible to estimate the surface density, as was done for stomata. Instead, the percentage of the leaf area covered by the trichome layer was estimated. Trichomes varied from being totally or nearly absent (*P. acerifolia*, *C. atlantica*, *P. cerasifera*, *C. australis*, and *P. nigra*), or very sparse (*C. bignonioides*, *R. pseudoacacia*, *A. saccharinum*, *Q. pubescens*, and *T. cordata*) up to covering almost entirely the leaf surface (*P. tremula* and *M. grandiflora*). Regarding LGE characteristic, species like *P. cerasifera*, *A. saccharinum*, *M. grandiflora* and *C. atlantica* showed higher than 70% leaf development at 60 days before final sampling, therefore these leaves had been exposed to air pollutants for longer time than others. On the other hand, species like *P. tremula* and *T. cordata* showed a leaf development close to 60%, and in the *C. bignonioides* case leaf expansion was  $<50\%$ , at 30 days before final sampling.

**Table 3**  
Mean particulate matter (PM) loads on leaf surfaces with standard errors, at different size fractions, as obtained by vacuum filtration (V/F) and scanning electron microscopy coupled with energy dispersed X-ray spectroscopy (SEM/EDX), for twelve trees species sampled in Le Grazie park in the city of Terni (Italy). Homogeneous groups were calculated through Fisher LSD – test and are represented by the letters (a–f), being "a" the group with the lower PM deposition, in every size fraction. The id codes of the sampled tree species correspond to those reported in Table 1.

Species	PM load by V/F ( $\mu\text{g}/\text{cm}^2$ )		PM load by SEM/EDX ( $\mu\text{g}/\text{cm}^2$ )							
	PM10	PM2.5	PM10	PM2.5	PM1					
<i>A. s.</i>	$2.76 \pm 1.06$	<b>bcd</b>	$0.69 \pm 0.15$	<b>ab</b>	$9.16 \pm 3.49$	<b>de</b>	$2.47 \pm 0.71$	<b>cd</b>	$0.67 \pm 0.23$	<b>d</b>
<i>C. b.</i>	$1.92 \pm 0.94$	<b>ab</b>	$0.68 \pm 0.09$	<b>ab</b>	$1.95 \pm 0.52$	<b>ab</b>	$0.49 \pm 0.09$	<b>a</b>	$0.23 \pm 0.06$	<b>abc</b>
<i>C. a.</i>	$3.71 \pm 0.38$	<b>cde</b>	$0.92 \pm 0.05$	<b>abd</b>	$2.94 \pm 0.66$	<b>ab</b>	$1.21 \pm 0.17$	<b>ab</b>	$0.47 \pm 0.02$	<b>bcd</b>
<i>C. au.</i>	$2.13 \pm 0.76$	<b>abc</b>	$0.71 \pm 0.33$	<b>ab</b>	$4.12 \pm 1.72$	<b>ab</b>	$0.80 \pm 0.23$	<b>a</b>	$0.23 \pm 0.05$	<b>abc</b>
<i>M. g.</i>	$2.75 \pm 0.57$	<b>bd</b>	$1.45 \pm 0.26$	<b>cde</b>	$4.67 \pm 0.42$	<b>cb</b>	$1.97 \pm 0.38$	<b>bc</b>	$0.65 \pm 0.19$	<b>d</b>
<i>P. a.</i>	$1.64 \pm 0.24$	<b>ab</b>	$0.58 \pm 0.22$	<b>ab</b>	$11.96 \pm 0.53$	<b>e</b>	$3.15 \pm 0.27$	<b>d</b>	$0.51 \pm 0.04$	<b>cd</b>
<i>P. n.</i>	$4.17 \pm 0.38$	<b>de</b>	$1.79 \pm 0.02$	<b>ce</b>	$1.93 \pm 0.39$	<b>ab</b>	$0.69 \pm 0.05$	<b>a</b>	$0.28 \pm 0.03$	<b>abc</b>
<i>P. t.</i>	$0.89 \pm 0.24$	<b>a</b>	$0.33 \pm 0.03$	<b>a</b>	$7.69 \pm 2.24$	<b>dc</b>	$1.23 \pm 0.04$	<b>ab</b>	$0.38 \pm 0.02$	<b>abcd</b>
<i>P. c.</i>	$5.08 \pm 0.46$	<b>e</b>	$1.87 \pm 0.55$	<b>ce</b>	$11.49 \pm 1.92$	<b>e</b>	$2.69 \pm 0.66$	<b>cd</b>	$0.67 \pm 0.15$	<b>d</b>
<i>Q. p.</i>	$1.94 \pm 0.45$	<b>ab</b>	$1.26 \pm 0.13$	<b>bcd</b>	$3.38 \pm 0.59$	<b>ab</b>	$0.65 \pm 0.05$	<b>a</b>	$0.19 \pm 0.02$	<b>ab</b>
<i>R. p.</i>	$3.80 \pm 0.96$	<b>de</b>	$1.98 \pm 0.45$	<b>c</b>	$1.80 \pm 0.49$	<b>ab</b>	$0.42 \pm 0.11$	<b>a</b>	$0.15 \pm 0.03$	<b>a</b>
<i>T. c.</i>	$6.69 \pm 0.32$	<b>f</b>	$2.99 \pm 0.19$	<b>e</b>	$1.10 \pm 0.25$	<b>a</b>	$0.64 \pm 0.16$	<b>a</b>	$0.17 \pm 0.03$	<b>a</b>

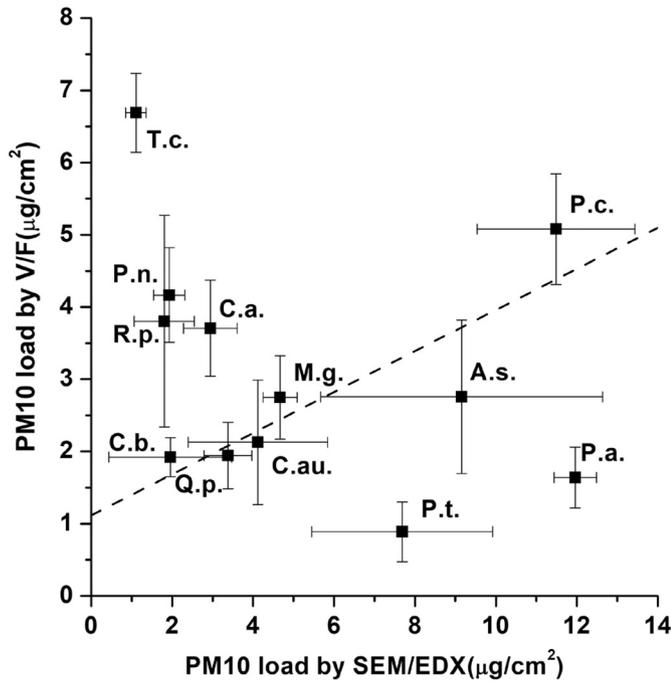


Fig. 3. Scatterplot comparison of two PM10 quantitative analysis techniques: scanning electron microscopy (SEM/EDX) and gravimetric analysis through vacuum filtration (V/F).

4. Discussion

4.1. Species-specific PM loads

Differences in PM deposition as a function of tree species has been documented previously by many studies (L. Chen et al., 2017; Mo et al., 2015; Shi et al., 2017; Weerakkody et al., 2017). However, we have discovered here that the observed differences can be, in some cases, also technical-dependent, otherwise different results are affected by the different techniques.

Six over the twelve species belong to the same ANOVA classification, at every PM size fraction and technique: *C. bignonoides*, *C. australis* and *Q. pubescens* always show low PM values, *A. saccharinum* and *M. grandiflora* always belong to the intermediate classes, while *P. cerasifera* shows always a high PM load.

Popek et al. (2013) reported similar conclusions for *C. bignonoides*: a relatively low capture efficiency was reported, with a minimum of  $3.7 \pm 0.2 \mu\text{g}/\text{cm}^2$ . This value is further comparable with those we have obtained ( $2.6 \pm 0.94 \mu\text{g}/\text{cm}^2$  and  $2.67 \pm 0.52 \mu\text{g}/\text{cm}^2$ , for both V/F and SEM/EDX, respectively), despite being observed in different background environments. Similarly, Paoletti and Bardelli (2011) confirmed low values of deposited PM by *C. australis* leaves. Nevertheless they suggested Mediterranean hackberry as a suitable tree species for urban environments. Blanusa et al. (2015) reported medium-low values with reference to *Q. cerris*, and related its capability in PM capture to the sclerophyllous habitus of leaves, superficial roughness, presence of trichomes, convex epidermal cells and presence of wax crystals.

The species *A. saccharinum* showed the highest variability among replicates, with large standard errors in both techniques. Nevertheless, based on the LSD post-hoc test, this species is partially related to the group with higher capability in PM capture (Table 3, “ab” group). This species has not been discussed in the literature, but many studies reported results for other *Acer* genus species, mostly *A. campestre* (Beckett et al., 2000; Dzierzanowski et al., 2011; Freer-Smith et al., 2004). Most of these studies did not show significantly high PM deposition values for this genus, but Sæbø et al. (2012) evidenced the efficiency of *A. campestre* and *A. pseudoplatanus* as compared to other species. *M. grandiflora*, one of the only two evergreen species included in this study, showed intermediate PM deposition values. The species has been mostly described in studies from China (Liang et al., 2017; Yang et al., 2014; Yin et al., 2011). Xie et al. (2018) described in detail its PM capture as the worst in a comparative study of six tree species, and related the low performance to a simple crown structure.

Finally, *P. cerasifera* is the species with high PM deposition values, which also showed a correlation between V/F and SEM/EDX techniques. In a comparative study among tree species, in contrast to present results, Shi et al. (2017) included cherry plum (*P. cerasifera*) in the group of lower PM deposition group. However, from the same study, the same species showed a major PM capture capability in a more polluted environment. Mo et al. (2015) included *P. cerasifera* in the intermediate-high PM deposition group, while Li et al. (2019) also reported relatively high values for this species.

On the other hand, for the remaining six species, the two techniques gave different results for the same species. The first possible explanation is the variable but important honeydew presence on leaf surface (detected by SEM images; data not shown): *R. pseudoacacia*, *P. nigra* and *T. cordata* showed an abundant quantity of sticky organic material probably produced by parasitic insects. As a result, PM is probably

Table 4

Micro and macrostructural characteristics recorded during leaf surface observation. The selected micromorphological characteristics are the abaxial surface roughness (which includes: type of surface structures described as smooth S, peaks P, valleys V, and grooves G; average percentage of the leaf area covered by structures; groove mean size), the mean stomata density, the average percentage of leaf surface area covered by trichomes. Macromorphological characteristic are also reported, such as margin complexity (described as smooth SM, serrate SER, lobate LOB), leaf shape (classified as elliptical ELL, compound COM, acicular ACI, palmate PAL), leaf growth expansion (LGE: percentage of leaf developed at 60 or 30 days before the final sampling), and foliage character (either deciduous DEC or evergreen EVE). The id codes of the sampled tree species correspond to those reported in Table 1. (NR: not recorded; s.e.: standard error).

Species	Micromorphology									Macromorphology						
	Roughness				Stomata		Trichomes			Morphology		LGE		Foliage		
	Type	Area	s.e.	Grooves	s.e.	Density	s.e.	Covered area	s.e.	Margin	Shape	60 days before	s.e.		30 days before	s.e.
	%		µm		#/m <sup>2</sup>		%				%		%			
<i>A. s.</i>	G	59.9	22.5	0.66	0.03	453.5	18.5	12.6	1.3	SM	PAL	77.9	15.2	100	/	DEC
<i>C. b.</i>	G/V	65.2	15.1	1.46	0.06	174.3	20.6	6.6	1.2	SM	ELL	0.6	0.2	49.8	19.5	DEC
<i>C. a.</i>	S	0	/	NR	/	NR	/	NR	/	SM	ACI	75.7	4.6	96.3	4.4	EVE
<i>C. au.</i>	G	4.5	1.4	1.72	0.06	334.4	28.1	0.4	0.2	SER	ELL	37.5	9.3	100	/	DEC
<i>M. g.</i>	S	0	/	NR	/	288.5	11.6	99.5	0.1	SM	ELL	89.4	23.9	93.2	/	EVE
<i>P. a.</i>	G	66.3	3.7	1.05	0.04	266.9	9.2	NR	/	SM	PAL	13.9	2.2	90.6	13	DEC
<i>P. n.</i>	NR	0	/	NR	/	97.2	7.4	0.9	0.5	LOB	ELL	33.2	5.8	100	/	DEC
<i>P. t.</i>	G	73.9	21.5	0.93	0.03	211.3	7.9	83.3	2.9	LOB	ELL	28	4.2	64.7	7.3	DEC
<i>P. c.</i>	G	84	9.6	0.79	0.03	351.9	11.9	0.9	0.3	SER	ELL	71.9	10.3	100	/	DEC
<i>Q. p.</i>	G	16	3.7	1.63	0.05	283.6	57.8	20.4	8.3	LOB	ELL	35.3	7.4	100	/	DEC
<i>R. p.</i>	P/V	57	1.7	1.42	0.06	44.5	3.6	4.5	0.4	SM	COM	26	4.2	100	/	DEC
<i>T. c.</i>	NR	0	/	NR	/	181.3	7	19.3	1.1	SER	ELL	34.9	12.2	68.2	7.5	DEC

overestimated by the V/F technique, since honeydew residuals have been weighted, together with washed PM. Furthermore, it should also be taken into account that honeydew could hinder PM particles from being detected by SEM/EDX analysis.

PM deposition on *T. cordata* has been described by Dzierzanowski et al. (2011) and Sæbø et al. (2012), and similar results were obtained by both studies to the SEM/EDX results in this study. Dzierzanowski et al. (2011) showed a PM<sub>2.5–10</sub> value between 2 and 3 µg/cm<sup>2</sup>, more comparable with our SEM/EDX results than V/F results. Also in a 22 tree species comparison, Sæbø et al. (2012) included *T. cordata* in the less efficient species group in PM capture capability. The species *P. nigra* has been observed by Beckett et al. (2000), which recorded PM deposition rates in a controlled environment (wind tunnel). In that study a remarkably fine PM retention capability for black poplar was observed, similarly with our results, in which PM<sub>1</sub> deposition represents a large part of the PM<sub>10</sub> load for *P. nigra*. Sæbø et al. (2012) and Muñoz et al. (2017) analyzed deposition on *R. pseudoacacia*; in both cases the black locust was classified as relatively inefficient in PM capture.

*C. atlantica*, the only coniferous species of our study, did not show high PM depositions. However many studies reported major PM capture capability by coniferous species if compared with broad-leaf trees (Freer-Smith et al., 1997; Räsänen et al., 2013), due to the high Leaf Area Index (LAI) of coniferous trees (Liang et al., 2016). Regarding *C. atlantica* there are no published studies related to PM deposition on leaves: Al-Alam et al. (2017) used Atlas cedar as biomonitoring tree in urban environment. The PM capture capability a different species but same genus (*C. deodara*) was described by Wang et al. (2013), which actually reported a significantly minor PM deposition quantity if compared with *P. acerifolia*.

Finally, both *P. tremula* and *P. acerifolia* showed high SEM/EDX PM<sub>10</sub> load values but very low V/F quantitative results. *P. tremula* was described by Sæbø et al. (2012), and similarly to present study was classified in the group with average PM deposition capability. Furthermore both studies showed its capacity in coarse PM capture. Different studies reported *P. acerifolia*'s efficiency in PM capture (Baldacchini et al., 2017; Gandolfi et al., 2017; Hofman et al., 2013) but they did not directly compare different species, except for Liang et al. (2017) which compared tree species in urban environment and describe their capability in heavy metal removal from air pollution. A similar comparison among species was presented by Muñoz et al. (2017), which reported *P. orientalis*, a species with an analogous foliar morphological structure of *P. acerifolia*, and ranked it as the best species in PM capture if compared with *A. negundo* and *R. pseudoacacia*. These previous results are consistent with our SEM/EDX quantification results, but not with our V/F results. On the other hand, Dzierzanowski et al. (2011), which used the V/F technique and described *P. hispanica* as the least effective in PM capture. One possible explanation of such discrepancy is the presence of epicuticular waxes. As reported by other studies (Mori et al., 2015; Przybysz et al., 2014; Sgrigna et al., 2015), PM of different size fractions can be easily included in epicuticular waxes: the electron beam used during the analysis (5 and 15 KeV), was probably able to penetrate the wax layer and measure all PM, while V/F technique not coupled with specific solvents (chloroform) cannot measure these particles. Thus, based on our results, and according with the literature (Baldacchini et al., 2019), SEM/EDX appears to be more appropriate than V/F for PM load analysis on leaves, since it allows to better distinguish either PM dimensional classes and its chemical content, and to better evaluate the overall PM amount.

#### 4.2. Correlation among leaf traits and PM capturing capability

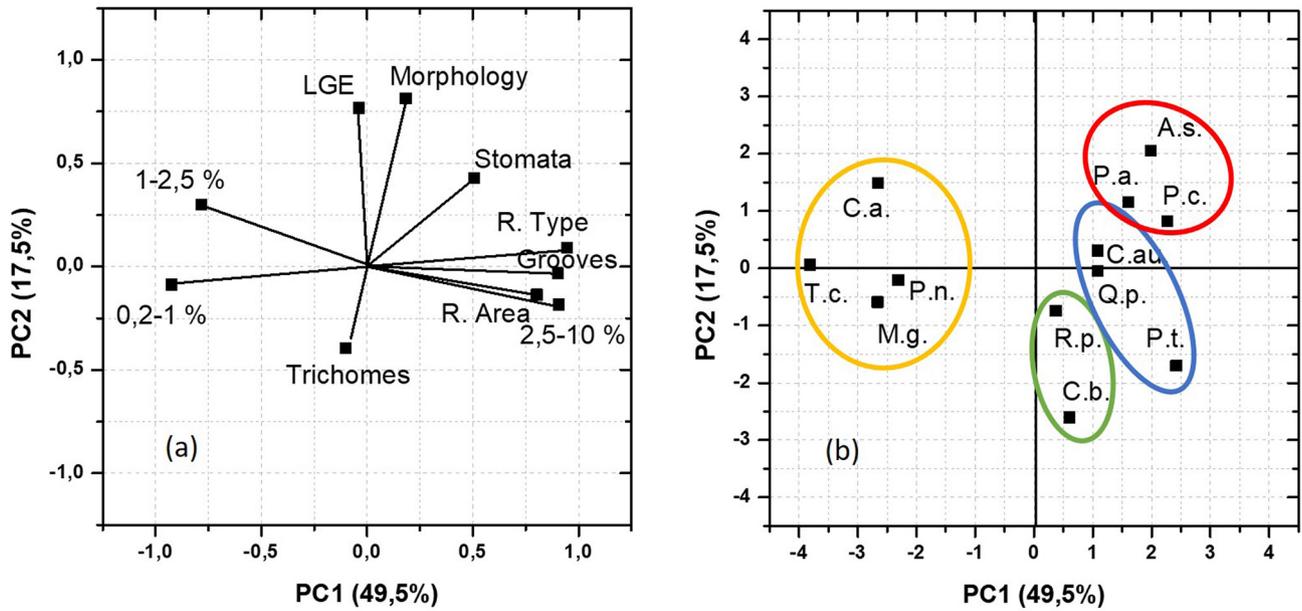
Leaf characteristics observed in this study showed great morphological diversity among the selected tree species, which, in turn, showed variable PM capturing capability.

In order to identify the relative importance of morphological characteristics to the three PM size fraction loads (PM<sub>0.2–1</sub>; PM<sub>1–2.5</sub>; PM<sub>2.5–10</sub>), a PCA was performed over the data collected for the twelve tree species. Variables used as input for the PCA were: SEM/EDX PM loads for the three size classes, expressed in relative percentages; numerical values associated to measurable morphological characteristic (namely, Rough Area, Stomata Density and Trichome covered area, from Table 4); scores values as derived by the values in Table 4, according to the scores described in Table 2, for Roughness Type, Groove size, LGE and the input variable called "Morphology" obtained by the sum of Margin complexity and Leaf shape scores. Foliage parameter was excluded from the PCA: it is characterized only by two possible values (0 or 3; respectively for DEC or EVE) and this would drive the whole species discrimination. The multivariate analysis results in nine principal components (PCs), where the cumulative eigenvalues for the first five PCs and the first three PCs represent the 94.3% and the 80.0% of variance, respectively. The factor coordinates and the factor scores of the two first PCs are plotted in Fig. 4. The eigenvalues of the two PCs, measuring proportion of variance, are at 49.5% and 17.5%, respectively for PC1 and PC2.

Based on factor coordinates (Fig. 4a), PC1 mainly describes the clustering of species with higher proportional quantity of PM<sub>1–2.5</sub> and PM<sub>0.2–1</sub>, comparing to the other cluster of species with higher deposition of PM<sub>2.5–10</sub>; the latter is also related to the presence of roughness characteristics. PC2, instead, differentiates species with high trichome density from those with high stomata density, which is also associated with a high LGE.

According to the above described PCs, tree species are grouped in four different families (Fig. 4b). The species *A. saccharinum*, *P. acerifolia* and *P. cerasifera* belong to the same group (red cluster), with positive high values for both PCs. They present similar PM deposition at each size fraction, and are characterized by high load of coarse particles (average percentages are: 74.4% ± 1.9% for PM<sub>2.5–10</sub>; 19.7% ± 2.2% for PM<sub>1–2.5</sub>; and 5.8% ± 1.5% for PM<sub>0.2–1</sub>) and low presence of trichomes. The other micro and macromorphological characteristics are well represented by these species: PM deposition is enhanced by the contribution of Rough characteristics (Area, Type and Grooves dimensions), Stomata density, Leaf Morphology and LGE. The influence of characteristics related to roughness on PM deposition has been observed in many studies (Shao et al., 2019; Wang et al., 2015; Weerakkody et al., 2018a). Smaller groove dimension and its connection with PM deposition is probably related to higher level of roughness expressed by this characteristic: smaller grooves are related to a higher number of ridges per unit area. Similar conclusions are also drawn by Liang et al. (2016) and J. Chen et al. (2017). There is still not a clear explanation to the correlation between high PM deposition and stomata density, probably because higher number of stomata enhances the total roughness trait of leaf surfaces. The position in the first quarter (Fig. 4b), for the red cluster, describes also a quantitative equilibrium of two PM classes, PM<sub>2.5–10</sub> and PM<sub>1–2.5</sub>, located on the fourth and second quarters of variables, respectively (Fig. 4a).

On the opposite side the two species *R. pseudoacacia* and *C. bignonioides* (green cluster), show a similar pattern for PM relative percentages (75.7% ± 1.1% for PM<sub>2.5–10</sub>; 14.2% ± 1.4% for PM<sub>1–2.5</sub>; and 10.1% ± 2.5% for PM<sub>0.2–1</sub>) but, differently from species in the red cluster, the two species showed general small contribution of all considered characteristics, except for Rough area micromorphological characteristic and a slight presence of trichomes. PC2 evidenced the effect of trichomes on PM capture, and a slight connection especially with smaller particles retention. Nevertheless PM deposition mechanisms linked to the abundance of trichomes and hairs are still unclear. Both techniques (V/F and SEM/EDX) implemented in this case study have limitation in identifying particles trapped within complex structures of trichomes such as i.e. *M. grandiflora* or *P. tremula*: microscope images at 5 KeV cannot penetrate inside the intricate mesh formed by everted structure on the abaxial side, and vacuum filtration technique may not be effective for PM resuspension in any solvent.



**Fig. 4.** Biplot of the factor coordinates of variables (a) and of factor scores (b) of the two first PCs obtained by correlation PCA of PM loads per size fraction (expressed as percentage of the PM10 load), and morphological characteristics of leaves.

The blue cluster (Fig. 4b) includes *C. australis*; *Q. pubescens* and *P. tremula*. The three species are characterized by a dominant presence of coarse particles ( $81.7\% \pm 1.9\%$  for PM2.5–10) and low levels of finer PM classes ( $12.8\% \pm 1.5\%$  and  $5.3\% \pm 0.4\%$  for PM1–2.5 and PM0.2–1 respectively). As described for the red cluster, coarse particles abundance is linked to Rough characteristics (Area, Type and Grooves dimension), and is more evident for *P. tremula* than the other species. On the other hand, *C. australis* and *Q. pubescens* showed a shorter expansion period and a larger number of stomata (Table 4). This is in contrast to the yellow cluster (Fig. 4b), where *C. atlantica*, *M. grandiflora*, *T. cordata* and *P. nigra* are grouped. The four species in the yellow cluster show the smallest amount in coarse PM deposition ( $59.4\% \pm 3.4\%$ ) and the highest presence of fine particles ( $25.6\% \pm 3.5\%$  and  $14.9\% \pm 0.8\%$  for PM1–2.5 and PM0.2–1 respectively). Finally, on Fig. 4b, we can observe that the only two evergreen species are grouped in the second quarter (still pertaining to the yellow cluster). A slight connection brought by PC2 between persistence of leaves and medium particles is evidenced on Fig. 4a.

Finally, PCA showed the synergy among different leaf traits: the more characteristics coupled in one species the more efficient that

species is in capturing PM. The presence of only one or two leaf trait is not a sufficient condition to achieve high efficiency for PM capture.

4.3. Particulate Accumulation index

The correlation highlighted by PCA among leaf morphological characteristics and PM deposition can then be used to obtain a morphology-based descriptive ranking of the PM capturing capability of the analyzed species, called the Accumulation index (Ai). According to the criteria described on Table 2, the scores for each species, shown in Table 5, include all the morphological characteristics that contribute to Ai. Then Ai is calculated by summing all the scores for each species (Table 5).

A wide range of Ai values are obtained, over the twelve selected species, ranging from 3 (*P. nigra*) to 17 (*A. saccharinum*). By plotting the PM10 load as a function of Ai (Fig. 5), the data fit well as an exponential curve (corresponding formula:  $y = 0.72e^{0.16x}$ ) where  $r^2 = 0.88$ , and  $p < 0.05$ .

**Table 5**

Single scores corresponding to the micro and macromorphological characteristics, with respect to their effects on the leaf PM accumulation, for twelve tree selected species. The id codes of the sampled tree species correspond to those reported in Table 1. Scores were derived from observation and measurement of the leaf characteristics, by combining the data in Table 4 with the scores in Table 2.

Species	MICROMORPHOLOGY					MACROMORPHOLOGY				
	Roughness			Stomata	Trichomes	Morphology		Foliage	LGE	
	Type	Area	Grooves	Density	Covered area	Margin	Shape			Total Ai
<i>A. s.</i>	3	2	3	3	1	0	2	0	3	17
<i>C. b.</i>	2	2	2	1	1	0	0	0	0	8
<i>C. a.</i>	0	0	0	0	0	0	2	3	3	8
<i>C. au.</i>	3	1	1	2	0	1	0	0	2	10
<i>M. g.</i>	0	0	0	2	3	0	0	3	3	11
<i>P. a.</i>	3	3	2	2	0	0	2	0	2	14
<i>P. n.</i>	0	0	0	0	0	1	0	0	2	3
<i>P. t.</i>	3	3	3	1	3	1	0	0	1	15
<i>P. c.</i>	3	3	3	3	0	1	0	0	3	16
<i>Q. p.</i>	3	1	1	2	1	1	0	0	2	11
<i>R. p.</i>	1	2	2	0	1	0	1	0	2	9
<i>T. c.</i>	0	0	0	1	1	1	0	0	1	4

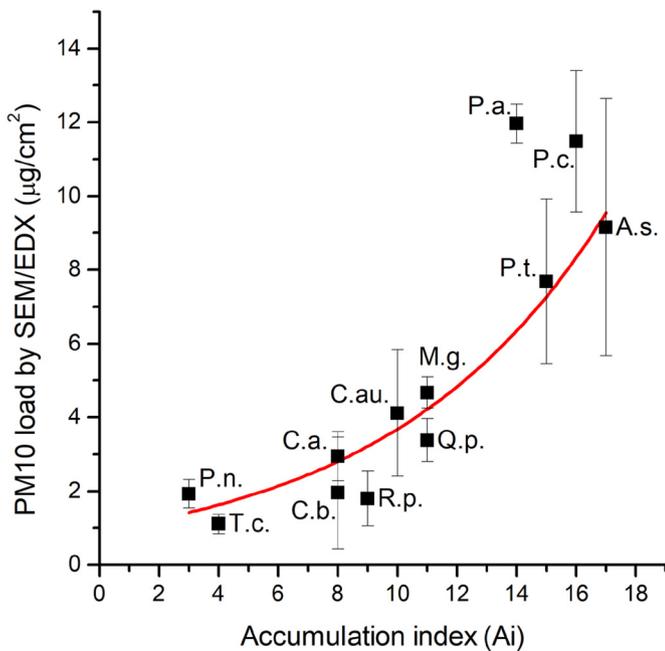


Fig. 5. PM10 load ( $\mu\text{g}/\text{cm}^2$ ) from SEM/EDX analysis related to Accumulation index (Ai) based on micro and macromorphologies observed and measured for twelve trees species. The regression line is represented by an exponential curve ( $r^2 = 0.88$ ;  $p < 0.05$ ).

The Ai was tested also with PM<sub>2.5</sub> and PM<sub>1</sub> loads as obtained by SEM/EDX (Figures SM1 and SM2), finding that an exponential trend can be observed, but with very low correlation coefficients. The low significance of Ai when associated to fine PM classes is likely due to the fact that fine particles are more easily dispersed (and resuspended) in the atmosphere with respect to coarser ones. Instead, when testing Ai with the PM loads obtained by V/F, rather scattered plots, without any correlation, are obtained (see SM3 and SM4); this further confirming that V/F is weaker technique in studying leaf deposited PM with respect to SEM/EDX.

Thus, the newly introduced Ai appears as a good descriptive tool for coarse PM accumulation capability of tree leaves. However, it is affected by some drawbacks that should be ameliorated by future studies. First of all, the choice of assigning to each character the same scoring range is arbitrary and should be refined, by weighting the score ranges. Also, a more robust result could be obtained by incrementing the number of included characteristics and of testing species.

## 5. Conclusions

Micro and macromorphology of leaves may have significant impact on their ability of capturing and retaining all particle size fractions. Some characteristics, such as trichomes density, is noteworthy but not crucial in PM capture: their presence is a co-existing factor and if isolated do not significantly contribute to PM deposition. The combination of different traits is a key factor to enhance PM capture. Rough leaf, with complex shapes, high stomata density and longer persistence with leaf blade totally developed were the combined characteristics related to the highest PM deposition values. The newly introduced Ai efficiently describe the PM capturing capability of tree leaves, taking into account a wide range of morphological characteristics.

Since a concrete action against PM pollution in urban environment is needed, we should focus not only on the reduction of emissions but also on natural and low cost solutions, such as the new and science-based urban greening approach. The results from this study can inform practitioners, urban managers and planners on the selection of tree species so that air particle capture and retention can be maximized. The presented Ai index can be a very useful tool for this purpose. It will be

possible to create a list of efficient PM capturing trees species, suitable for specific latitudes or environments.

## 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.scitotenv.2020.137310>.

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