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# Species-specific efficiency in PM<sub>2.5</sub> removal by urban trees: From leaf measurements to improved modeling estimates



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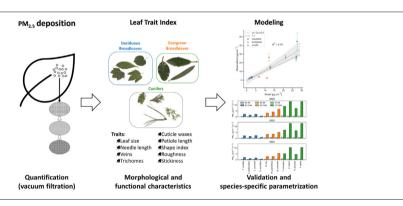
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#### HIGHLIGHTS

#### Modifying i-Tree Eco model based on leaf traits and experimental measures.

- Improved model matches experimental measures for conifers and deciduous broadleaves.
- PM<sub>2.5</sub> deposition was 4.6 to 32.2 higher than the conventional model estimates.
- Results provide more reliable quantification of air quality improvement by trees.

## GRAPHICAL ABSTRACT



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### ABSTRACT

The growing population in cities is causing a deterioration of air quality due to the emission of pollutants, causing serious health impacts. Trees and urban forests can contribute through the interception and removal of air pollutants such as particulate matter (PM). The dry deposition of PM by vegetation depends on air pollutant concentration, meteorological conditions, and specific leaf traits. Several studies explored the ability of different plant species to accumulate PM on leaf structures leading to the development of models to quantify the PM removal. The i-Tree Eco is the most used model to evaluate ecosystem services provided by urban trees. However, fine particulate matter (PM2.5) removal is still calculated with a poorly evaluated function of deposition velocity (which depends on wind speed and leaf area) without differentiating between tree species. Therefore, we present an improvement of the standard model calculation introducing a leaf trait index to distinguish the species effect on PM net removal. We also compared model results with measurements of deposited leaf PM by vacuum filtration. The index includes the effect of morphological and functional leaf characteristics of tree species using four parameters: leaf water storage, deposition velocity, resuspension rate and leaf washing capacity. Leaves of 11 common urban tree species were sampled in representative areas of the city of Ferrara (Italy) and at different times of the year from 2018 to 2021. This includes four deciduous broadleaf trees (Tilia cordata, Platanus acerifolia, Acer platanoides, Celtis australis), three evergreen broadleaf trees (Quercus ilex, Magnolia grandiflora, Nerium oleander), and four conifers (Thuja orientalis, Cedrus libani, Pinus pinaster, Picea abies). The results provide significant advancement in assessing PM removal using decision support tools such as models to properly select tree species for future urban tree planting programs aimed at improving air quality.

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#### 1. Introduction

As urban population is dramatically increasing worldwide, life quality and health in urban areas play a role of paramount importance for human well-being. Unfortunately, the latter is seriously affected by air pollution originating from a variety of anthropogenic sources such as motor vehicles, indoor heating, industries, etc. According to the EEA Report No 10/2019, fine particulate matter (PM2.5) caused 412,000 premature deaths in Europe in 2018, being the air pollutant with the highest impact in terms of mortality in EU. In fact, exposure to high PM<sub>2.5</sub> levels is associated to elevate risk of cancer, cardiovascular and respiratory mortality (Pun et al., 2017) and cause severe economic costs for the society (Trejo-González et al., 2019). The exposure to high levels of PM<sub>2.5</sub> air concentrations also seems to increase the mortality due to COVID-19 during pandemic (Brunekreef et al., 2021). In order to improve air quality levels, EU legislation imposed legal limits for different air pollutants (EU Directive 2008/ 50), which targets were often not achieved or criticized as insufficient to protect human health (Wise, 2018; Yamineva, 2017). Therefore, effective measures are urgently needed to improve the urban environment and protect its inhabitants.

In this regard, Urban Green Infrastructures (UGI) and particularly urban trees (Livesley et al., 2016) are gaining popularity as a cost-effective solution to improve the environmental quality of cities (Keesstra et al., 2018), supplying a comprehensive set of ecosystem services to support citizens' well-being, including air purification, storm water retention, temperature regulation, recreation and habitat provision (Capotorti et al., 2020). Among these, the capacity of urban vegetation to intercept and remove airborne particulates has been identified as an important process in polluted cities (Baldacchini et al., 2017; Manes et al., 2016). Airborne PM removal by vegetation is a result of multiple processes encompassing background pollution levels (i.e. PM air concentrations), meteorological and climatic conditions (e.g. wind speed, temperature and precipitation intensity) (Beckett et al., 2000; Nowak et al., 1998) and specific plant morphological characteristics (Corada et al., 2021; Sgrigna et al., 2020). In particular, two main processes can be identified: i) the gravimetric sedimentation of particles on leaves and ii) their subsequent retention on leaf surface to avoid resuspension until a precipitation event.

In the last decade, a growing number of studies demonstrated that PM removal depends on specific morphological traits of tree leaves and crown (Corada et al., 2021; Grote et al., 2016; Sgrigna et al., 2020) and that, consequently, plant species perform differently in removing airborne PM. However, the available literature provides a fragmented knowledge of the interaction between morpho-functional traits and PM removal, as well as their upscaling towards a quantification of PM removed by vegetated areas. Some leaf attributes, as trichomes density and cuticle wax content, were associated with a higher fine PM removal capacity. Foliar trichomes have the first contact with atmospheric PM thus favoring their adsorption and reducing resuspension (Zhang et al., 2019a). In addition, it seems that the presence of trichomes also affects leaf wettability (Bickford, 2016) which, in turn, influences the potential amount of PM deposits (Wang et al., 2013). Leaves with higher content of cuticle waxes can adsorb and retain (i.e. avoid or mitigate resuspension phenomena) larger amounts of fine PM (Dzierzanowski et al., 2011; Sgrigna et al., 2015), while no analogous effects were clearly demonstrated for coarser PM (Popek et al., 2013). Some authors also stated that composition and structure of the wax may influence the holding capacity of PM (Leonard et al., 2016; Wang et al., 2015a), further supporting the evidences for species-specific properties. Similar effects can be also attributed to leaf secretions. Even though more investigations are needed to clarify their role, some studies observed that sticky leaf surfaces capture and retain more PM than smooth leaves (Corada et al., 2021; Sgrigna et al., 2020). PM removal is also influenced by macro-level morphological and geometrical traits. Higher dry deposition levels are associated with leaves with roughness surfaces, due to the presence of veins, wrinkles and ridges (Zhao et al., 2019), as well as complex leaf shape (Son et al., 2019). Also, petiole length was found to be negatively correlated with PM deposition velocity (Zhang et al., 2021): a

shorter petiole may reduce leaf movement thus enhancing the velocity of particle deposition (Prusty et al., 2005), while a longer petiole enables leaves to flutter and causes resuspension (Leonard et al., 2016).

A further parameter that influences PM removal capacity is the washing performed by rainfall. In fact, intense precipitation events cause PM to be washed off from leaves, recovering their capacity to filter airborne PM. Even though some studies demonstrated that rainfall intensity and duration are most relevant parameters affecting overall PM removal (Weerakkody et al., 2018; R. Zhang et al., 2019b), it should also be noted that plant can retain part of the deposited PM according to their leaf traits after a rain event (Xu et al., 2017) - particularly in the case of fine PM (Xu et al., 2019). Leaf traits such as presence of sticky secretions, epidermal waxes, trichomes density and patterns, stomatal density, shape of epidermal cells, and leaf water status, may help avoiding a complete leaf washing during moderate rain events possibly due to their influence on leaf wettability (Wang et al., 2015b).

The application of UGI as nature-based solutions for improving air quality and its implementation in policy requires reliable information of PM retention at a large scale (e.g. city or neighborhood levels). The biophysical upscaling of airborne PM<sub>2.5</sub> removal assessment is often based on the use of dedicated models and tools as a basis for decision-making processes. Nonetheless, the reliability of such assessments depends on model accuracy, which is seldom validated with field measurements and is seriously affected by simplistic model assumptions. Among such models, i-Tree Eco (www.itreetools.org) is the most popular software suite for the estimation of a number of ecosystem services provided by urban trees and has been widely used to estimate PM<sub>2.5</sub> removal (Pace et al., 2018; Selmi et al., 2016; Song et al., 2020), but doesn't differentiate between leaf properties (Nowak et al., 2013), which has been criticized as an oversimplification (Pace and Grote, 2020). Model estimates were also compared with experimental measurements at the canopy and leaf level of a Mediterranean holm oak forest (Pace et al., 2021), but we couldn't find any studies that systematically evaluated and compared the impact of different tree species properties on the calculation of PM removal.

With the aim to rank and predict the PM removal performance of different urban tree species, some authors combined a set of leaf morphological traits by means of statistical models (Chen et al., 2017; Li et al., 2019; Muhammad et al., 2019), ranking criteria (Yang et al., 2015) or to compute indexes that quantify PM removal (Sgrigna et al., 2020). However, such efforts were not implemented in existing models to improve the assessment of air purification capacity of UGI to support decision processes. In fact, despite the rapidly increasing body of literature demonstrating their relevance, to date there is no clear and effective application of morphofunctional leaf traits in PM removal modeling (Corada et al., 2021).

The integration of experimental data on leaf particulate accumulation and the detailed differentiation of leaf traits on PM deposition-resuspension-removal processes can be used to improve model parameterization and evaluation. The aim of this work is thus to suggest such improvements to be used in the i-Tree Eco or similar models by testing and implementation of different species-specific properties. Therefore, leaf and crown traits of species were integrated to compute a score-based index or semi-mechanistic model, which was then validated/calibrated using primary measures obtained from field samplings of different urban tree species. The index is based on four model parameters that influence  $PM_{2.5}$  deposition on leaves: i) potential leaf water storage, ii) dry deposition velocity, iii) resuspension rate and iv) rain washing. We assume that this work represents a significant advancement on PM removal modeling that is urgently needed for improving our estimates on PM removal and to guide planners and management to the choice of the best tree species for this purpose.

## 2. Material and methods

## 2.1. Study area and laboratory measures

The city of Ferrara is located on the Po plain (Fig. 1), an extended flat area in Northern Italy which is subjected to high concentration levels of

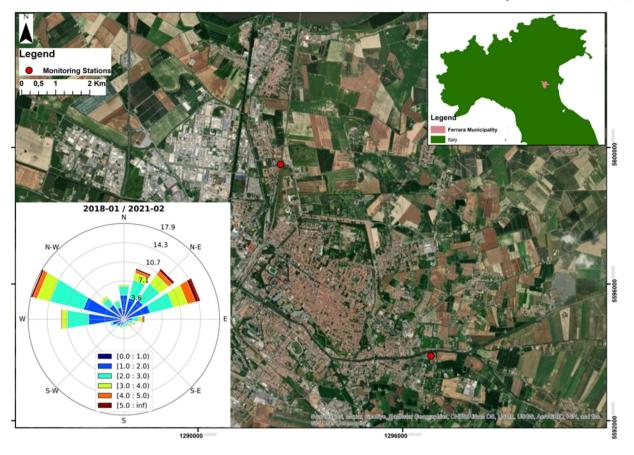


Fig. 1. Ferrara municipality and the two monitoring stations for PM<sub>2.5</sub> air concentration and meteorological data. The panel in the back-left reports wind speed and direction for the sampling period (summer 2018 – winter 2021).

air pollutants, due both to the regional emissions and its particular geomorphological conformation. The municipality of Ferrara has about 132,000 inhabitants living in an area of 405.16  $\rm km^2$ , at approximately 9-13 m a.s.l. The annual precipitation is about 814 mm yr  $^{-1}$ , while the mean annual temperature is 14.6 °C. The area is classified as "Cfa" (humid subtropical climate) according to the Köppen-Geiger climate classification (Peel et al., 2007). Prevalent wind directions are W in winter and NE in summer, with an annual mean speed of 3.4 m s  $^{-1}$ . According to the municipality census of public trees (Ferrara Municipality, 2018), *Celtis austalis*, *Tilia europea*, *Platanus acerifolia* are the most abundant species.

Meteorological data were obtained from the database of the Regional Environmental Agency (https://simc.arpae.it/dext3r/), while PM<sub>2.5</sub> concentrations were derived by two monitoring stations (Fig. 1). Deposition measurements were taken at 11 urban tree species that were selected according to their frequency of occurrence, differences in leaf traits, and closeness to the monitoring stations. Among the species were four deciduous broadleaves (*Tilia cordata, Platanus acerifolia, Acer pseudoplatanus, Celtis australis*), three evergreen broadleaves (*Quercus ilex, Magnolia grandiflora, Nerium oleander*) and four evergreen conifers (*Thuja orientalis, Cedrus libani, Pinus pinaster, Picea abies*). Of each species, 4 individuals were selected in order to cancel out differences regarding the position of emission sources.

Samplings were carried out between June 2018 and February 2021 with deciduous trees measured during summer after new leaves were fully developed (June/July) and evergreens during winter (January/February). An additional measure for *Q. ilex* (later winter) and *T. orientalis* (summer) was performed to obtain a second value for evergreen and conifer species considering their longer leaf-on period. Twelve samples of outer branch leaves that covered a mean leaf area of 3151 cm<sup>2</sup> per sample (min 691 cm<sup>2</sup>, max 14,182 cm<sup>2</sup>), were taken from each tree species (three samples x four trees) for analysis of deposition load as well as determining leaf traits. They are stored in plastic bags, placed in portable fridges, and

quickly taken to the laboratory. Leaves were sampled at >5 m height to avoid effects due to particle resuspension from the ground. The amount of PM<sub>2.5</sub> deposited on leaf surface was measured by vacuum filtration method (Sgrigna et al., 2015, 2020). This approach enables to directly weigh the amount of PM accumulated on the leaves and compare it with the values estimated from the model flux calculation (Pace et al., 2021). Leaves were washed one by one in 500 ml of micro-distillated water. The solution with suspended particles was filtered in a plastic sieve (100 µm pore size) to remove coarse material and subsequently forced through filters using a vacuum filtration system. PM  $> 10 \mu m$  was removed from the solution using filters with 10-13 µm porosity (Anoia S.A., Barcelona, code 1250). Coarse PM (between 2.5 and 10 µm) was collected using filters with 2.5 µm size (Whatman No. 42 Cat No. 1442-055, Maidstone, UK), while fine PM fraction (<2.5 μm) was collected on nitrocellulose filter with  $0.2 \, \mu m$  pore size (Whatman NC20). All filters were dried at 50 °C for 72 h before and after the vacuum filtration and then stabilized in glass boxes before being weighted through electronic precision balance (precision 0.0001 g, Crystal 100 CAL, Gibertini). The difference between the first and second weight expresses the sum of PM deposits on the analyzed amount of leaves of the same sample. The  $\mathrm{PM}_{0.2\text{--}2.5}$  load was assumed to be represent tative 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. The results of PM accumulated at the whole leaf/needle were then normalized by dividing the PM mass by the reference leaf area and expressed in  $\mu g\ cm^{-2}.$  Because of the different morphology of their leaves, the reference leaf area of each sample was derived differently for broadleaves and conifers. For broadleaf samples it was assumed to be the projected leaf area, directly measured by ImageJ (Schneider et al., 2012) in order to be consistent with model assumptions. The i-Tree Eco model, as well as other modeling approaches, uses LAI (the total one-sided green leaf area per unit per ground surface area) to calculate PM removal rates (Janhäll, 2015). Projected leaf area of conifers was

estimated according to the leaf blade image collection method (patent n. CN104990530A, 2015, Muresan et al., 2022), except for *T. orientalis* samples that were measured by leaf scanning due to their specific leaf conformation. This method was adopted after laboratory testing of reliability in comparison with a different approach based on conversion from needle-projected surface to leaf area (Räsänen et al., 2013) resulting in similar results. Specifically, leaf area of conifers was estimated by assuming that needles have a cylindrical shape. The surface of a single needle was then estimated by multiplying needle length by needle girth. The latter was measured making the cross-sectional slice in the middle part of the needle. The individual surfaces of 20 needles were measured to obtain a mean value and then multiplied by the number of needles present in each branch sampling. In addition, diameter at breast height and crown diameter were measured in situ for each sampled tree.

#### 2.2. Removal index and model implementation

The PM<sub>2.5</sub> deposition flux was calculated according to the method implemented in the i-Tree Eco model (Nowak et al., 2013). Morphological and functional leaf traits were combined to obtain an index that, in turn, was implemented in the model to include the species-related effects in PM<sub>2.5</sub> removal. The index encompasses four parameters (j, k, i, w) that influence deposition velocity (j), resuspension (k), leaf washing (w) and canopy water storage (i):

$$\begin{split} f_t &= Vd_t \cdot j \cdot C \cdot LAI \\ R_t &= (A_{t-1} + f_t) \cdot \frac{rr_t}{100 \cdot k} \\ \\ A_t &= \begin{cases} A_{t-1} + f_t - R_t & \text{if } rain \leq plws \cdot LAI \cdot i \\ (A_{t-1} + f_t - R_t) \cdot \left(1 - \frac{1}{w}\right) & \text{otherwise} \end{cases} \\ \\ F_t &= \begin{cases} f_t - R_t & \text{if } rain \leq plws \cdot LAI \cdot i \\ 0 & \text{otherwise} \end{cases} \end{split}$$

where  $f_t$  is the PM<sub>2.5</sub> flux at time t (g m<sup>-2</sup> s<sup>-1</sup>), Vd<sub>t</sub> is the deposition velocity at time t (m s<sup>-1</sup>), C is the PM<sub>2.5</sub> air concentration (g m<sup>-3</sup>), LAI is leafarea index,  $R_t$  is the PM<sub>2.5</sub> flux resuspended in the atmosphere at time t (g m<sup>-2</sup> s<sup>-1</sup>),  $A_t$  is PM<sub>2.5</sub> mass deposited on leaves at time t (g m<sup>-2</sup> ground) depending on previous hour deposition ( $A_{t-1}$ ) as well as precipitation,  $r_t$  denotes a 'resuspension class', which is the relative amount of deposited PM<sub>2.5</sub> that is resuspended at a specific wind speed at time t (%), *plws* is the potential leaf water storage (mm) and  $F_t$  is the net PM<sub>2.5</sub> removal at time t after considering resuspension. The leaf area index of tree species has been calculated based on the Beer-Lambert Law (Nowak et al., 2008):

$$LAI = \frac{\left[\frac{\ln (1 - x_s)}{-k}\right] \cdot \pi r^2}{CC}$$

Where  $x_s$  is the species-specific shading factors, k is the light extinction coefficient (0.52 for conifers and 0.65 for hardwoods) (Nowak, 1996), r is the crown radius (m), and CC is the crown covered area (m<sup>2</sup>).

Deposition velocities ( $vd_t$ ) and resuspension classes ( $rr_t$ ) both depend on wind speed and are defined based on the i-Tree Eco model standards (Nowak et al., 2013). When precipitation events are higher than the maximum water storage of the canopy (Ps in mm), which is calculated according to the potential leaf water storage plws (0.2 mm) and LAI (Ps = plws \* LAI), PM<sub>2.5</sub> deposition on leaves (A<sub>t</sub>) is assumed to be washed off according to the washing rate (w), and F<sub>t</sub> are set to 0.

Leaf on (DOY 63) and leaf off (DOY 320) dates were set based on the dates of the first and last frost events, assuming a 30-day transition period in spring when trees produce leaves reaching maximum LAI and in fall when they lose leaves leading LAI to 0.

The assessment of leaf traits was performed by either direct measurements or information based on literature (Kattge et al., 2020; Keating, 2009) as specified in Table 1. The values measured on the species samples with variations were reported in Table S1. Leaf size, needle length, shape index and petiole length were derived from data obtained by scanned images and needle measurements. Trichome density (number of trichomes per area unit) and surface roughness (% of leaf surface covered by groves, ridges and wrinkles) were quantified by means of electron microscopy (SEM) images at a magnification rate of 500 (derived from previous tests at different magnifications). Images from leaves were visually inspected, trichomes per area were counted and surface areas were classified as 'rough' surfaces if groves, ridges and/or wrinkles were visible. After drying and mounting, the leaf specimens were cut into squares (1 cm<sup>2</sup>), coated with gold in an Edwards S150 sputter coater and examined with a Zeiss EVO 40 SEM. Three images of each species were captured as replications. Presence of veins and cuticle waxes were considered based on literature information (Keating, 2009; Kattge et al., 2020). Stickiness was evaluated by direct observations (presence of sticky organic material on leaves)

A score was attributed to each leaf trait according to Table 2 and then computed to obtain a final value for each of the four parameters (i, j, k, w) as following:

$$\begin{split} i &= \begin{cases} \textit{leaf area} & \textit{if broadleaves} \\ \textit{neddle length} & \textit{if conifers} \end{cases} \\ j &= \begin{cases} \frac{(\textit{veins} + \textit{trichomes} + \textit{waxes})}{3} & \textit{if broadleaves} \\ 3 & \textit{if conifers} \end{cases} \\ k &= \begin{cases} \frac{(\textit{petiole lenght} + \textit{shape index} + \textit{roughness})}{3} & \textit{if broadleaves} \\ 3 & \textit{if conifers} \end{cases} \\ w &= \begin{cases} 2 \times \textit{Stickiness} & \textit{if broadleaves} \\ 3 + \textit{Stickiness} & \textit{if conifers} \end{cases} \end{split}$$

Table 1
Set of leaf traits used in model improvement. Method of assessment, scores rationale and assumption on which they were attributed are reported.

Leaf trait	Information source Description		Assumptions					
Leaf size	Direct measure	Measured with leaf scanning	Larger leaves need larger water amounts for a complete washing					
Needle length	Direct measure	Directly measured	Larger needles need larger water amounts for a complete washing					
Veins	Keating (2009)	Veins order (from 1 to 4)	Veiny leaves have higher deposition velocity due to their rougher surfaces					
Trichomes	Direct measure	Measured by SEM images	Trichomes increase particles capture by increasing their deposition velocity					
Epicuticular waxes	Kattge et al. (2020)	Abundant/Mod. present/absence	Waxes increase particles capture by increasing their deposition velocity					
Petiole length	Direct measure	Measured with leaf scanning	Longer petiole favor particle resuspension by increasing leaf movement					
Shape index	Direct measure	Measured with leaf scanning (length/width ratio)	Narrow leaves are less subjected to resuspension phenomena					
Roughness	Direct measure	Measured by SEM images	Presence of groves, ridges and wrinkles on leaf surface prevents resuspension					
			of trapped particles					
Stickiness	Direct observation	Presence/absence	Sticky secretions can retain PM even after rain events					

Table 2 Model parameters and the respective leaf traits used to attribute the respective scores. Class differentiation is according to Keating, 2009 (leaf area and vein orders), Sgrigna et al., 2020 (trichomes and roughness classes), Kattge et al., 2020 (waxes), and according to the number of group members (petiole length and leaf shape).

Model parameter	Leaf trait	Classes	Score
Canopy water storage (i)	Leaf area (mm²)	<225	1
		225-4500	2
		>4500	3
	Needle length (cm)	<2	1
		2-4	2
		>4	3
Deposition velocity (j)	Veins	1° order	1
		2° order	2
		3°- 4° order	3
	Trichomes (number/mm <sup>2</sup> )	<33	1
		33-66	2
		>66	3
	Waxes	Absent	1
		Moderately present	2
		Abundant	3
Resuspension (k)	Petiole length (cm)	>2	1
		1-2	2
		<1	3
	Shape index (adimensional)	>3	1
		1.5-3	2
		<1.5	3
	Roughness (% coverage)	<33	1
		33-66	2
		>66	3
Washing (w)	Stickiness	Presence	1
		Absence	0

#### 2.3. Comparative analysis

In order to test the performance of the leaf trait-based approach, the experimental measures were matched with the  $PM_{2.5}$  deposited flux

computed by the modified model. Additionally, the latter was also compared with the results of the conventional i-Tree Eco model to detect the differences induced by the modified approach. Both models were run for the period ranging from 1st January 2018 to 22th February 2021. This period covers more than three complete years until the date of the last field sampling, thus to evaluate the seasonal performances of models for the different species. Moreover, the results of the modified model allowed the calculation of the mean annual PM<sub>2.5</sub> removal per space unit (g m $^{-2}$  yr $^{-1}$ ) for each species. Such outcome can inform environmental managers about the removal performance of different species and address urban planning towards the planting of the most effective species to improve air quality in cities.

#### 3. Results

#### 3.1. Meteorological conditions and experimental analysis

The meteorological conditions recorded during the sampling period are shown in Fig. 2. Mean monthly  $PM_{2.5}$  concentrations followed seasonal patterns, with higher values observed in winter because of heating and traffic emissions.  $PM_{2.5}$  concentrations in Barco and Villa Fulvia stations were comparable, even though the first recorded slightly higher concentrations in the whole period. For the years 2018, 2019 and 2020, the mean annual  $PM_{2.5}$  concentrations were respectively 21.8, 21.1 and 23.3  $\mu g \ m^{-3}$  for Barco station and 16.4, 17.0 and 16.9  $\mu g \ m^{-3}$  for Villa Fulvia station. These values fall below the 25  $\mu g \ m^{-3}$  legal limits foreseen by EU Directive 2008/50/CE, but exceed the recommended WHO threshold (10  $\mu g \ m^{-3}$ ). Mean monthly wind speed for the sampling period was 2.21 m s  $^{-1}$ , ranging from 1.71, to 2.77 m s  $^{-1}$ . The highest monthly precipitations were 162.6 mm in May 2019 and 160 mm in November 2019.

The results of PM<sub>2.5</sub> mass deposited on leaves (At) are reported in Table 3. The higher depositions were found in *T. orientalis* (44.6  $\pm$  10.1  $\mu$ g cm<sup>-2</sup>), while the lowest were measured in *P. x acerifolia* (2.1  $\pm$ 

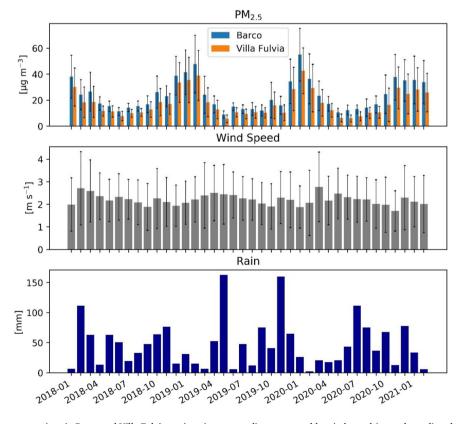


Fig. 2. Mean monthly  $PM_{2.5}$  concentrations in Barco and Villa Fulvia stations (upper panel), mean monthly wind speed (central panel) and monthly precipitations (lower panel). Values represent mean daily values ( $\pm$ st.dev) for PM concentration and wind speed, while cumulated values for rain.

**Table 3**Mean PM<sub>2.5</sub> mass per leaf unit (At) for each sampled species. Sampling stations and dates are reported.

Species	Type	Station	Sampling date	Mean $A_t$ ( $\pm$ St.err.) ( $\mu g$ cm <sup>-2</sup> )
Tilia cordata	deciduous	Barco	6/21/2018	3.5 (±0.6)
Quercus ilex	evergreen	Villa Fulvia	1/17/2019	$2.5 (\pm 0.5)$
Acer platanoides	deciduous	Villa Fulvia	7/26/2019	$2.6 (\pm 0.6)$
Celtis australis	deciduous	Villa Fulvia	7/26/2019	$3.2 (\pm 0.7)$
Platanus x acerifolia	deciduous	Villa Fulvia	7/26/2019	$2.1 (\pm 0.8)$
Thuja orientalis	conifer	Barco	7/26/2019	$14.2 (\pm 2.7)$
Cedrus libani	conifer	Villa Fulvia	1/13/2020	25.0 (±3.0)
Pinus pinaster	conifer	Villa Fulvia	1/13/2020	$26.9(\pm 2.7)$
Magnolia grandiflora	evergreen	Barco	1/13/2020	$14.6 (\pm 1.0)$
Picea abies	conifer	Villa Fulvia	2/22/2021	$26.3 (\pm 2.4)$
Nerium oleander	evergreen	Villa Fulvia	2/22/2021	$26.2(\pm 4.3)$
Quercus ilex	evergreen	Villa Fulvia	2/22/2021	$3.6 (\pm 0.5)$
Thuja orientalis	conifer	Barco	2/22/2021	44.6 (±10.1)

 $0.8~\mu g~cm^{-2}$ ). It is worth to be noted that these values do not represent the air depuration capacity of species but rather the PM<sub>2.5</sub> deposited on leaves at the sampling date, as a function of previous meteorological conditions and air PM concentrations at the sites. Therefore, the observed values were used to test and calibrate the improved version of the model.

### 3.2. Leaf traits and model results

The four model parameters (i, j, k and w) were computed on the basis of the specific leaf traits classified for each of the 11 considered species, as are reported in detail in Table 4. Conifers have larger values for i, j and w indices that respectively influence deposition velocity (vd), resuspension rate (rr) and washing. Some traits were specific to only some of the considered species. For example, an abundant amount of sticky substances was evident in leaves of T. cordata and T. orientalis. Higher coverage of rough structures on leaf surfaces was observed in N. oleander and, to a lesser extent, in P. acerifolia and A. platanoides. Interestingly, these two species were also those with lower trichome density. Afterwards, the modified model was run to be tested with the experimental At reported in Table 3. Its performance is shown in Fig. 3, which contrasts experimentally observed vs. model predicted amounts of PM deposits. Figs. 4, 5 and 6 respectively show simulated PM<sub>2.5</sub> deposits for deciduous broadleaves, evergreen broadleaves, and conifers. The improved model successfully predicted PM loads at deciduous and conifer species, while was less accurate for evergreen broadleaves. The model underestimated both experimental measures of Q. ilex. In contrast, PM deposition of M. grandiflora and N. oleander, as well as the second measures of T. orientalis were slightly overestimated.

#### 3.3. Model comparison and species performances

The modified model was also compared with the conventional i-Tree-Eco model to describe differences in  $PM_{2.5}$  deposition for different species. The standard i-Tree-Eco model markedly underestimates PM

mass deposited on all considered species, except for *Q. ilex* (Fig. 4). In particular, conifers accumulated higher amounts of PM by one order of magnitude (Fig. 5). In general, PM load is significantly higher in conifers than deciduous broadleaves.

The results also highlight differences in PM loads among species and tree types. Conifers accumulated more PM in all seasons when compared with evergreen and deciduous broadleaves. Evergreen broadleaves revealed contrasting results. The amount of deposited particles at *Q. ilex* was similar to those found at deciduous species, while that at *N. oleander* was similar to those at conifer species. Interestingly, a higher PM deposition flux does not necessarily correspond to a higher PM removal rate, as demonstrated by computing the total PM<sub>2.5</sub> removal per year for each species (Fig. 7). For instance, although *T. orientalis* showed higher PM deposition (Fig. 5), higher annual PM removal values were observed for *P. abies*, *C. libani* and *N. oleander*. It has to be mentioned, however, that the total PM removal reported in Fig. 7 is calculated for the whole tree and is therefore influenced by the related leaf area index, while PM accumulation fluxes are normalized per leaf area unit.

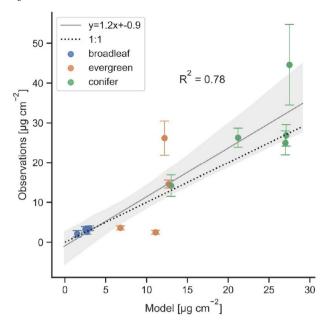
### 4. Discussion

# 4.1. Model performance and uncertainties

The results demonstrate the importance of including the properties of different tree species when assessing the contribution of UGI to air depuration. While the role of specific leaf traits in determining PM removal by trees is often demonstrated in literature (Chávez-García and González-Méndez, 2021; Xu et al., 2021), the introduction of a comprehensive set of traits into PM removal modeling is done here for the first time. Despite difficulties, this model approach has been evaluated under conditions of a wide range of environmental dynamics, such as wind speed and rainfall, considering washing and resuspension as separated processes that affect fine particulates to a larger extent than coarse material (Yan et al., 2019;

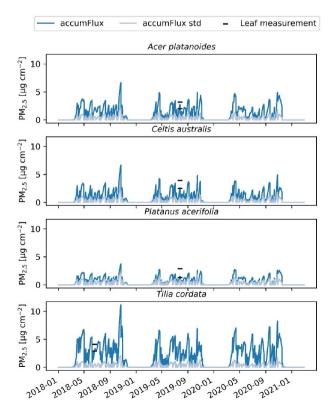
Table 4
Scores attributions for the four parameters (i, j, k and w) based on leaf traits values (Shape index = leaf length/leaf width). A 'minus' indicates that no reports of the specific traits could be found for these species. In this case, a '0' is assumed for calculations.

Species	plws			Vds			Rr				Washing		
	Leaf size (mm²)	Needle length (mm)	i-Index	Veins	Trichomes (n./mm²)	Cuticle waxes	j-Index	Petiole length (cm)	Shape index	Roughness (%)	k-Index	Stickiness	w-Index
Tilia cordata	2174.4	_	2.0	4°	>100	Absent	2.5	3.02	1.24	0	2.0	Present	2.0
Quercus ilex	664.1	-	1.0	$2^{\circ}$	>100	Moderate	2.0	1.22	2.74	0	2.0	Absent	0.0
Acer platanoides	5981.5	-	3.0	3°	16	Absent	2.0	10.00	0.93	60	2.0	Absent	0.0
Celtis australis	237.2	-	1.0	$2^{\circ}$	83	Absent	2.0	1.53	2.83	5	2.0	Absent	0.0
Platanus x acerifolia	6936.4	_	3.0	$1^{\circ}$	0	Absent	1.0	1.79	1.08	65	2.5	Absent	0.0
Thuja orientalis	_	118.6	3.0	_	_	-	3.0	_	-	_	3.0	Present	4.0
Cedrus libani	_	24.4	1.0	_	_	-	3.0	_	-	_	3.0	Absent	3.0
Pinus pinaster	_	144.2	3.0	_	_	-	3.0	_	-	_	3.0	Absent	3.0
Magnolia grandiflora	8047.9	-	3.0	$1^{\circ}$	>100	Abundant	2.5	2.35	1.97	0	1.5	Absent	0.0
Picea abies	-	16.8	1.0	-	-	-	3.0	-	-	_	3.0	Absent	3.0
Nerium oleander	1908.0	-	2.0	4°	>100	Moderate	2.5	0.77	6.99	85	2.5	Absent	0.0

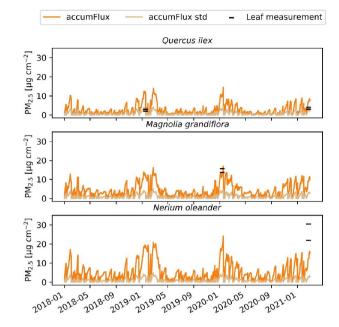


**Fig. 3.** Mean leaf PM load values observed experimentally (bars represent standard errors) vs. values predicted by the improved model. The dashed line represents the ideal condition of perfect match between observed vs. predicted values (1:1 ratio). The solid line represents the best fitting linear regression (grey area corresponds to 95 % confidence interval) ( $\mathbb{R}^2 = 0.78$ , p < 0.05).

Zhang et al., 2018). This work therefore provides important advances towards a more reliable quantification of  $PM_{2.5}$  removal by trees, significantly improving the popular i-Tree-Eco tool.



**Fig. 4.** PM $_{2.5}$  deposition fluxes for deciduous broadleaf species during the considered period (01–2018; 02–2021) as modelled by standard (light blue line) and modified (bright blue line) models. The experimental measures were shown in black lines (upper and lower limits of confidence interval).



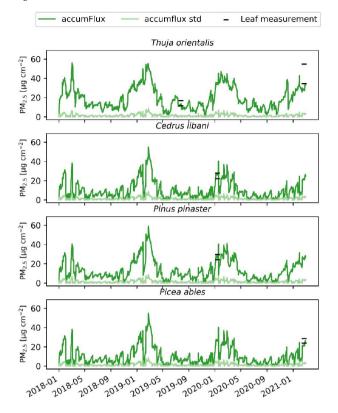
**Fig. 5.** PM<sub>2.5</sub> deposition fluxes for evergreen broadleaf species during the considered period (01–2018; 02–2021) as modelled by standard (light orange line) and modified (bright orange line) models. The experimental measures were shown in black lines (upper and lower limits of confidence interval).

The effects of specific leaf traits on  $PM_{2.5}$  accumulation and removal were implemented by means of multi-criteria scores that, in turn, adjust the responses to the four environmental processes considered by the model. This approach allows accounting the composite interactions between leaf traits and environmental conditions, a challenging aspect for the quantification of the PM removal service provided by urban trees.

The results obtained with the modified model were generally close to the experimental PM<sub>2.5</sub> accumulation measurements of deciduous broadleaves and conifers, thus supporting the view that it is a useful addition to current tools. In fact, the comparison with the conventional i-Tree-Eco model showed that the latter strongly underestimates PM<sub>2.5</sub> deposition on leaves and fails to distinguish species effects on it. In particular, deposition values were found to be a factor of 4.6 to 10.3 higher in deciduous broadleaves, 7.6 to 11.9 in evergreen broadleaves, and 23.9 to 32.2 in conifers, than the conventional model estimates (Figs. 4, 5 and 6). The generally higher PM loads on conifer species is corroborating results of previous studies (Chen et al., 2017; Muresan et al., 2022; Zhang et al., 2019a, 2019b), showing that longer and narrower needles may be more easily hit by airborne particles than large and flat leaves (Sæbø et al., 2012).

These outcomes are also reflected in the annual PM<sub>2.5</sub> removal values of sampled trees (Fig. 7), which further highlights the very good performance of the model particularly regarding *P. abies* and *C. libani*. Although *T. orientalis* showed the highest accumulation values in winter season, the total annual removal was lower than for *P. abies and C. libani*. Also, the observations of Yang et al. (2015) suggest higher PM loads on cupressaceous species, which might be related to the presence of sticky substances on leaf surfaces of these species that are retaining fine particles also during intense precipitation events. This feature demonstrates that PM deposition and removal should be considered separately, using overall removal as indicator for air depuration (a regulation service) instead of deposition rates (Chung et al., 2021). The lower PM removal of *P. pinaster* can be explained by its lower leaf area measured on sampled plants, compared to the other considered conifers.

Among the other species, *N. oleander* is the only species reaching levels of PM<sub>2.5</sub> deposition similar to those of conifers (Table 3). This unexpected result could be due to the combined performance in terms of deposition velocity and avoided resuspension, related to the presence of fourth order veins, roughness surface and short petiole length. Interestingly, the



**Fig. 6.**  $PM_{2.5}$  deposition fluxes for conifer species during the considered period (01–2018; 02–2021) as modelled by standard (light green line) and modified (bright green line) models. The experimental measures were shown in black lines (upper and lower limits of confidence interval).

conventional model represents deposition values of *Q. ilex* better than the modified model, despite the low scores obtained for potential leaf water storage and deposition velocity parameters (Table 4). In general, the shorter vegetation period and the overall low trait scores are the reasons for the lower values of deciduous broadleaves. Among those, the highest deposition values are reached by *T. cordata*, as also observed by Steinparzer et al. (2022,) which are explained by higher deposition velocity and presence of sticky substances on leaf surface. The effect of the latter trait on total annual PM removal was partially compensated by the lower leaf washing capacity of *Tilia* species.

Some deviations between measurements and simulations using the improved model, particularly regarding evergreen broadleaves, may originate in the history of their leaf. Older leaves tend to accumulate larger amounts of fine PM because their exposure time to air pollutants is longer (Memoli et al., 2020) and their leaf microstructure is more differentiated (Niu et al., 2020). At the same time, some traits, such as the presence of epicuticular waxes, can prevent PM wash off after rain events over time, thus leading to larger PM loads in older leaves. Accordingly, younger leaves are supposed to have less PM deposits. In this regard, sampled branches of evergreen species may have included younger and older leaves causing a bias in the results of leaf PM accumulation and may partly explain the relatively poor performance of the model for evergreen broadleaf species. A major impact due to different leave ages, however, seems unlikely according to some recent studies which demonstrated that leaf age is only correlated with the amount of PM deposits if stratified by other factors, e.g. local emission sources (air pollution intensity) or chemical composition of particulates (Esposito et al., 2020; Memoli et al., 2020; Niu et al., 2020). Still, uncertainties remain since it cannot be excluded that sample sites were affected by different amounts or compositions of air pollution.

Another uncertainty related to one (or two) sampling periods per year is the fact, that leaf properties change during the period of leaf expansion and senescence. Taking properties of fully developed leaves as representative

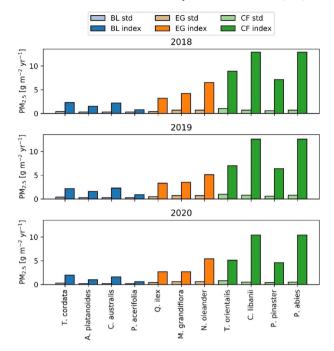


Fig. 7. Total annual  $PM_{2.5}$  removal of the different species as calculated by standard (std) and modified (index) models (BL = deciduous broadleaves; EG = evergreen broadleaves; CF = Conifers).

for the whole year will lead to deviations between measurements and simulations particular for deciduous broadleaves in spring and autumn. In cases were trees are developing leaves continuously (e.g. *M. grandifolia*), the sampling error might be smaller, since selected leaves might contain a representative average of younger and older leaves at any time. Still, sampling at multiple times of the year that cover different leaf phenological phases and an explicit dynamic development of leaf properties would increase the accuracy of model parametrization and the realism of the model process representation.

Additionally, it should be considered that leaf washing before vacuum filtration may not completely wash off the fine PM attached on epicuticular wax layer, which maybe a likely explanation for the overestimation of experimentally defined deposition values in case of Q. ilex, although this issue was not observed with seemingly relatively similar leaves of M. grandiflora, for which the model underestimated the measured deposition. Furthermore, the soluble part of the PM deposited at the leaves which is probably dissolved in the washing solution may be missed with the vacuum filtration method (Ristorini et al., 2020). The error, however, might be small as indicated by comparison with other advanced methods which produced similar results at least for the fine particulate component which we concentrated on (Baldacchini et al., 2019; Pace et al., 2021). Another uncertainty on the measurement of PM accumulation could be related to the measurement of leaf surface area, particularly for conifers because of their needle morphology. Furthermore, the level of accuracy in the measurement and classification of leaf traits could be improved by using more objective methods defined for particular scales when measuring leaf characteristics such as roughness (Andrade et al., 2022).

### 4.2. Management implications

Nature-based solutions are now mainstream considerations in environmental management to improve ecosystem services provision and protect human health (Keesstra et al., 2018). Their application requires a reliable evaluation of the ecological performances which, however, is not available for many currently applied decision support tools (Kangas et al., 2018; Schröter et al., 2015). Our results supported the notion that simply by neglecting the species-specific leaf traits, models may often and significantly

underestimate the contribution of urban trees to air quality, thus limiting their reliability for application in urban management. The problem is that the average deposition velocity that is used for all tree species alike, seems to be too low in most cases, at least for Mediterranean urban species (Pace et al., 2021). We therefore recommend using the improved model for assessing PM<sub>2.5</sub> removal for decision making in urban green management in order to select the most effective taxa for reducing air pollution levels. Regarding the presented model setup, conifers are generally the most effective plants in terms of PM<sub>2.5</sub> removal because of both their higher accumulation capacity per leaf unit and higher leaf area. However, species selection needs to consider local climate and air pollution conditions. Advantages of broadleaf trees include their ability to regenerate leaves each year regaining their full deposition capacity and potentially provide renewed leaf structures to interact with particulate matter. This may particularly provide advantages compared to evergreens in environments where air pollution removal is more important in the summer.

A further highlight of this study is that it emphasizes the close dependence of particulate removal on leaf washing capacity. Species with sticky substances such as waxes or resin are able to accumulate more particulate matter than others, but, at the same time, they do not allow for complete leaf washing and thus reducing the amount of particulate matter removal. This feature is particularly important when considering climate conditions for selecting tree species to promote PM removal. Species that accumulate and retain particulates are recommended for dry environments, where precipitation events are sparse and concentrated, while leaves with better wetting characteristics are recommended in rainy environments, where a lower accumulation is balanced by continuous PM removal. Such information can be suitable for planning reforestation measures as for instance those targeted by the European Strategy for 2030 (European Commission, 2019), which should contribute to mitigate air pollution effects on human health. Nonetheless, it is worth mention that although greening measures offer opportunities for reducing air pollution levels, a solution of the air quality problem requires the integration with other actions targeted to reduce pollution emissions (Jennings et al., 2021).

Furthermore, urban trees provide a set of other valuable services that should be considered as well in urban green management. For example, although less effective in PM removal when compared to conifer species, deciduous broadleaves contribute largely to tropospheric ozone absorption (Fusaro et al., 2017; Muresan et al., 2022) and produce higher benefits in terms of urban heat island mitigation (Yilmaz et al., 2018). For these reasons, a functional selection of tree species requires analyzing the ecosystem service demand, since mismatches between urban services supply and demand generate ineffectiveness and inequalities in urban planning strategy (Maragno et al., 2018; Sebastiani et al., 2021). In this sense, conifer plantations should be primarily destined to zones with higher PM emissions and presence of most vulnerable people, while deciduous species may be better suited for areas with higher temperature during summer season. Another aspect for species selection may be to focus on autochthonous species to support local biodiversity. In the case of Ferrara municipality, 2 out of 4 conifers sampled in this analysis are non-native species.

In general, species should be always selected according to the local environmental context. In fact, local climatic conditions not only interact with vegetation in regulating ecosystem services provision (Cimburova and Berghauser Pont, 2021) but also affect plant health, thus determining the success of plantation efforts and maintaining total leaf area over time. These aspects can perhaps best be captured by considering the potential vegetation of the area (Blasi et al., 2010). Finally, species selection should be coherent with urban cultural history and landscape configuration to support cultural services, as sense of place, aesthetic values, recreation, and education.

#### 4.3. Future research

Improving current models for the quantification of air depuration is challenging. The presented work provides important model improvements by introducing species-specific sensitivity based on leaf traits that can be obtained by both direct measurements as well as literature sources. The

relatively certain availability of leaf trait knowledge increases model accuracy, although some traits may be better known than others. We thus recommend future research also considers the interaction among different traits, as well as interactions with local environmental conditions. Moreover, the integration of trait-based and fluid dynamics models could be an additional field of investigation (Vos et al., 2013). The results of this study also highlighted the importance of experimental measures to calibrate commonly used models. Finally, future studies are called for that enable the evaluation of model processes regarding PM deposition and removal processes.

#### 5. Conclusions

The combination of experimental measurements and modeling enabled the development of an index, based on morphological and functional characteristics of leaf traits that can serve for a more accurate semi-mechanistic modeling of species-specific fine particulate removal. The outcome provides a significant advancement for improving decision support tools such as the i-Tree-Eco model and represents a basis for a broader classification of species useful for improving urban air quality. These results are considered relevant to planning and management of urban green spaces, and important for more comprehensive analyses of plant-PM interactions. We recommend that future studies should focus on the leaf history of deposited PM, the characteristics of the particulate matter, leaf structures, as well as the saturation levels of leaves and their ability to regenerate after precipitation events.

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## CRediT authorship contribution statement

Mattias Gaglio: Conceptualization, Methodology, Data curation, Formal analysis, Writing – original draft. Rocco Pace: Conceptualization, Methodology, Software, Visualization, Writing – original draft. Alexandra Nicoleta Muresan: Methodology, Data curation, Formal analysis, Visualization. Rüdiger Grote: Methodology, Software, Writing – review & editing. Giuseppe Castaldelli: Conceptualization, Resources, Writing – review & editing. Carlo Calfapietra: Conceptualization, Supervision, Writing – review & editing, Funding acquisition. Elisa Anna Fano: Conceptualization, Supervision, Writing – review & editing, Funding acquisition.

## 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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