

Methods to quantify particle air pollution removal by urban vegetation: A review

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ABSTRACT

Among the ecosystem services provided by urban forests, the air quality amelioration is particularly relevant. The high level of air pollution in modern cities and the indirect involvement of particulate matter (PM) in the spread of COVID-19 have exacerbated the air quality issue worldwide. However, in the estimation of urban vegetation effectiveness in particle air pollution removal, there is a lack of a standard procedure. Different methods are used for this purpose, making the comparison across different studies difficult. Therefore, there is a need of an extensive review, aimed at: i) identifying the existing direct methods to quantify this ecosystem service, ii) assessing their pros and cons, accuracy and reliability, sustainability, and iii) laying the foundations to create a standard method, commonly and universally recognized. We identified and meticulously assessed five main direct metrics: the gravimetric method (G, 40%), aerosol monitor (AM, 20.5%), wind tunnels and deposition chambers (WT&CH, 19.5%), Scanning Electron Microscopy (SEM, 14%) and Saturation Isothermal Remanent Magnetization (SIRM, 6%). This work provides a crystal picture and a critical framework of the last thirty years literature on this topic and lays the foundations to create a common and shareable approach to quantify the air PM mitigation potential of the urban vegetation. This will be useful to guide researchers and urban planners in shaping greener, healthier, and more sustainable cities.

1. Introduction

Air pollution is one of the major health concerns in urban areas, leading to 7 million premature deaths every year, 91% of which is caused by particulate matter (PM) (Wróblewska and Jeong, 2021; and reference therein). Air pollution is directly mentioned in 2 of the 17 Sustainable Development Goals (SDGs) of the 2030 Agenda for Sustainable Development, signed in 2015 by United Nations (UNs) members: SDG 3.9 (substantial reduction of health impacts from hazardous substances) and SDG 11.6 (reduction of adverse impacts of cities) (United Nations, 2023). However, its implications on other Goals can also be considerable.

PM is an air contaminant which consists in a mixture of solid and liquid particles (with an aerodynamic diameter in the range 0.001–100 μm) (Xu et al., 2020) of different chemical composition and shape, produced in cities mainly by vehicles, industrial plants, power plants and heating systems. It is usually classified in PM_{10} , $\text{PM}_{2.5}$, PM_1 , and $\text{PM}_{0.1}$ according to particle size (aerodynamic diameter <10 μm , <2.5

μm , <1 μm and <0.1 μm , respectively) and its risk for human health is higher for finer particles, which more easily trap toxic substances (such as polycyclic aromatic hydrocarbons and heavy metals) and penetrate the respiratory tract until reaching the alveolar and the blood circulation system, causing a variety of human systemic diseases (Hofman et al., 2013; Popek et al., 2013; Qiu et al., 2018; Wróblewska and Jeong, 2021). Moreover, PM has also environmental effects, since it can reduce visibility (by forming haze) (Yin et al., 2020), affect ecosystems (by contaminating water and soil), damage stone and other materials (including statues and monuments) and warm or cool climate depending on its components (e.g., black carbon by absorbing sunlight contributes to global warming, while particulate sulfates cool the earth's atmosphere) (WHO, 2023; EPA, 2023; Wang and Shi, 2021).

Although current policies are trying to reduce emissions, the air PM concentration is still very high, making mitigation measures urgent. Air purification devices, such as mechanical air filters (high volume) or powered electronic air cleaners (ionizers), were developed for this purpose but do not provide additional functions and are short-lived

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(Donateo et al., 2021).

Nature-based solutions, instead, represent an authentic public service, able to provide multiple ecosystem services in the long-run, such as carbon sequestration, micro-climate regulation, noise reduction, rain-water drainage, psychological and social values and, last but not least, air pollution mitigation (Hofman et al., 2013). A large body of literature, from over a century (Diener and Mudu, 2021; Litschke and Kuttler, 2008), pointed out the air quality amelioration effect of vegetation, considered as an important terrestrial sink for atmospheric particles, able to reduce outdoor but also indoor concentration of pollutants (Jang et al., 2021; Maher, 2013), thus lowering human exposure. The local PM levels reduction due to the urban green coverage was estimated to be in the range 0.2–26%, as reviewed by Wróblewska and Jeong (2021); Abhijith et al. (2017) reported that vegetation barriers along open roads, compared to the vegetation barrier free condition, could provide 15–60% reductions in PM and other pollutants concentrations. Maher, 2013 found 50% less PM inside houses screened by a temporary tree line. However, many different factors can influence this ecosystem service, and they are related to pollutants (e.g., diameter, shape, composition), plant characteristics (e.g. leaf-traits, vegetation type, structure, and configuration) and environmental variables (e.g., humidity, rain, wind speed, pollutant concentration, urban configuration) and their interactions. The scientific research, which is still working on untangling this complex topic, should collaborate with urban planning to maximize this benefit in cities. For example, in mid-latitude countries (e.g., Europe, China and USA), it is essential to consider the seasonal difference between vegetation dynamic and pollutant emission in a year. Indeed, both vegetation cover and air quality are generally higher in summer than in winter, when deciduous plants are leafless and the fossil fuel consumption rises (Han et al., 2020). A larger use of evergreen species, therefore, should be considered to reduce the gap between the “source” and the “sink”. In the complex relationship between the urban vegetation and the air particulate pollution, two main mechanisms are known: deposition and dispersion.

Deposition, the most studied process (Diener and Mudu, 2021), indicates the particle accumulation on plant surfaces (i.e., bark and leaves, the latter considered the most important), driven by rain and snow (wet deposition) or by gravity, inertia and Brownian motion (dry deposition) (Yin et al., 2020). Deposited PM can be absorbed into the plant tissues (e.g., through the leaf stomata) or, more frequently, retained on the plant surfaces (surface PM) and/or encapsulated in epi-cuticular wax layers (in-wax PM). Surface PM can be resuspended into the air under the wind force, washed off by rain or fell to the ground for instance during defoliation (Popek et al., 2013; Yan et al., 2016b), providing a relative short-term removal (Bertold et al., 2019). In-wax PM is more immobilized, providing a relative long term-removal, although wax desquamation occurring during leaves lifetime could resuspend the trapped PM into the environment (Przybysz et al., 2014). If resuspended in the air, particles can redeposit on plant organs and if reaching the ground, organic components may be decomposed by micro-organisms, inorganic components may be immobilized in the soil, or particles can be resuspended again (Vigevani et al., 2022).

The dispersion mechanism indicates how the plants, simply acting as a physical entity (obstacle), can modify air wind speed and turbulence, influencing the local concentration of pollutants. Vegetation imposes a drag on the air moving through the leaves and branches, which causes some air to move up and around the canopy. This can change the particles trajectory and velocity and reduce the air PM concentration downwind of the vegetation (Diener and Mudu, 2021; Ranasinghe et al., 2019). The vertical mixing which is created within the canopy, moreover, can favor the particle deposition on the vegetation surfaces. Thus, dispersion is a process which may hardly be disentangled from deposition, as pointed out by Diener and Mudu (2021). On the other hand, the drag action can cause a windbreak effect, especially in the case of long and thick green barriers, resulting in lower wind speed and turbulence behind the vegetation. This windbreak effect reduces both the dispersion

and the rate at which pollutants can be advectively moved away, potentially augmenting the air PM concentration downwind of the vegetation (Ranasinghe et al., 2019).

In addition to the known deposition and dispersion processes, a third mechanism is the modification, which includes any modification of the pollutant particles with possible alterations in the exposure risk for human health. Modification may relate to the PM size, due to chemical coagulation forces (such as Brownian or van-der-Waals ones, which determine the aggregation of fine or ultrafine particles in larger ones on plant surfaces), and to the PM composition, due to the selective sorption of plants (e.g., some species accumulate more easily some specific metals than others) or due to the microbiological influence (particles modification through microbes present on plant surfaces) (Diener and Mudu, 2021). However, this mechanism is still little investigated and it complicates the study of the overall PM removal by urban plants. For example, Yin et al. (2020) identified a coagulation effect of ultrafine particles (UFPs) on plant leaves, suggesting that coagulation and dry deposition were two processes that occurred simultaneously and interacted.

The scientific research has widely demonstrated the potential of different urban species (e.g., Beckett et al., 2000a; Popek et al., 2013; Vigevani et al., 2022) and, to a lesser extent, of different planting configurations (e.g., Ozdemir, 2019; Qiu et al., 2018, 2019) in the air PM mitigation in urban areas. However, some studies discovered that the differences among species, for example, can be not only species but also technical-dependent (Sgrigna et al., 2020). The multitude of metrics and metrics modifications existent in literature, the variety of experimental conditions and the confusion about terms and concepts makes the comparability across studies really difficult to perform and underlines the need to address in a more clear and organic way the topic. Thus, the aim of this work is to carry out an integrative review (i.e., a review with the aim to synthesize, assess and critique the literature on a research topic in a way which enables new theoretical frameworks and perspectives to emerge, as defined by Snyder, 2019) on the direct methods to evaluate the effectiveness of urban vegetation in particle air pollution removal. Specifically, the aims are: i) identifying the existing direct methods to quantify this ecosystem service, ii) assessing their pros and cons, accuracy and reliability, sustainability, and iii) laying the foundations to create a standard method, commonly and universally recognized. This work will improve the knowledge on this topic by providing an organic and critique framework, fundamental to guide researchers and urban planners towards a more green and healthy future.

2. Methodology

The review approach laid its foundations on primary and secondary questions (Knight et al., 2016). The primary question, from which the literature research and relative results originates, was: “which are the existing direct methods to quantify the effectiveness of urban vegetation in particle air pollution removal?”. The secondary ones, which determine a critical analysis of the findings obtained in the first, were: “Which are their pros and cons? How accurate and reliable are they? How are sustainable from the environmental, economic and social point of view? Is there a standard procedure to quantify the plant species removal potential? If not, is it possible to create it, refining and optimizing existing methods?”.

After setting these questions, a literature analysis was carried out on the scientific databases Scopus, Web of Science and Google Scholar by using four relevant keywords (“Urban vegetation”, “Particulate matter”, “Accumulation”, “Method”) searched in combination using the AND operator, on title, abstract and keywords (except for Google Scholar, in which the survey was carried out on full paper due to database limitations). One additional keyword (“Air pollution”) was added to exclude pollution of other environmental compartments (e.g., water and soil). Per each keyword, a set of correlated search terms was fixed and used with OR operator. While for Scopus and Web of Science only one run was

conducted, for Google Scholar several runs were applied (using several combinations of the different search terms) due to text length requirements.

Since the initial survey provided a large body of literature (a total of about 8700 results), search terms and inclusion criteria were tested (as suggested by Snyder, 2019) until the databases were able to provide a manageable number of articles which could effectively be about the subject of interest (final search terms can be found in Table 1). In this way, 1092 papers (52 reviews and 1040 articles; date: March 18, 2022) were obtained. A first selection of these papers was made by reading the title (and the abstract when the title was ambiguous), providing 343 papers (22 reviews and 321 articles). A second selection was made by reading the full text of the reviews (to check if these included methodological approaches) and abstract and methods of the articles, to check if they effectively reported direct methods to quantify the effectiveness of urban vegetation in particle air pollution removal. This second screening returned 144 papers (3 reviews and 141 articles). Twenty relevant papers (3 reviews and 17 articles), known by the authors but not returned by the literature survey, were added. In this way a list of 164 papers (6 reviews and 158 articles) was obtained. The entire process was carried out by two reviewers, as suggested by Pullin and Stewart (2006). The final list was made by papers selected based on the following inclusion criteria.

1. To be a full text papers (including original research and reviews), peer-reviewed, available in English;
2. To be published between 1992 and 2022 from any geographic location;
3. To include a relevant subject: anyone reporting direct methods (in real or controlled conditions) to quantify the urban vegetation effectiveness in particle air pollution removal (PM deposition on plants, also in relation to PM dispersion due to the presence of plants).

The final list did not include (exclusion criteria).

1. Papers which only cover pollens, elements, heavy metals, ions or polycyclic aromatic hydrocarbons (PAH) or held in non-urban forests or crops;
2. Papers based on models;

Table 1

Main and additional keywords and their relative sets of search terms used in the literature review.

	Keywords	Sets of search terms
Main	Urban vegetation	"vegetat*" OR "urban forest*" OR "greening" OR "woody species" OR "tree*" OR "shrub*" OR "green hedge" OR "green barrier*" OR "green infrastructure*" OR "park*" OR "garden" OR "green space*" OR "green area*" OR "green wall*" OR "green roof"
	Particulate matter	"particulate matter" OR "particle matter" OR "PM" OR "PM10" OR "PM2.5" OR "PM1" OR "PM0.2" OR "PM0.1" OR "particulate pollution" OR "airborne particle*" OR "airborne particulate*" OR "atmospheric particle*" OR "ultrafine particle" OR "ultrafine particulate*" OR "fine particle*" OR "fine particulate*" OR "coarse particulate"
	Accumulation	"purification" OR "purify" OR "adsorp*" OR "mitigat*" OR "deposit*" OR "disperse" OR "dispersion" OR "retain" OR "accumulat*" OR "capture" OR "reduce" OR "reduction" OR "trap*" OR "sequestration" OR "sink" OR "filter"
	Method	"method*" OR "technique*" OR "procedur*" OR "approach*" OR "process*" OR "quantitative assessment" OR "quantitative analysis"
Additional	Air pollution	"air borne" OR "ambient air" OR "airborne" OR "outdoor air" OR "air quality" OR "air pollution" OR "air pollutant*" OR "atmospher"

3. Papers which only cover PM dispersion or that treated the theme from a biomonitoring (and source attribution) or pollution tolerance (PM effects on plants growth and physiology) or planning (e.g., guidelines) perspective;
4. Papers based on synthetic plants or organs or idealized crowns;
5. Papers unclear, especially in methods.

The process of including and excluding papers was documented carefully by using a bibliographic software (Mendeley) and per each paper excluded the reason for exclusion was recorded. During the second selection phase, each paper was tagged with preliminary keywords related to.

1. Type of pollutant (PM, PM elements, etc.)
2. PM removal mechanism (deposition, dispersion or modification)
3. Method used (gravimetry, Scanning Electron Microscopy, aerosol monitor, wind tunnels and deposition chambers, Saturation Isothermal Remanent Magnetization)
4. Parameters (air PM concentration; deposition velocity; deposition amount)
5. Approach (real conditions, controlled conditions, models)
6. Type of vegetation (urban vegetation, roadside vegetation, green walls, etc.)
7. Scale (plant level, city level, regional level)
8. Other (e.g., biomonitoring)

This step was useful to provide a preliminary categorization of the papers and to deal with the full text reading in a valuable way. Indeed, addressing the reading by grouping the papers by method used was extremely convenient.

The list of 164 papers (6 reviews and 158 articles) was reduced to 146 papers (6 reviews and 140 articles) after removing 18 papers which did not match the set inclusion criteria.

3. Results

In this review a quantitative and qualitative data extraction was performed for the articles (all the data extracted are available in supplemental material), while only qualitative data extraction (i.e., general information and considerations reported about methods) was carried out on the reviews.

3.1. General overview

Regarding the geographical context, the studies examined were mostly performed in Asia (48%, of which 35% in China and 6% in Korea) and Europe (38%, of which 13% in UK, 8% in Poland and 6% in Italy), while less works came from America (8%, of which 6% in USA) and Oceania (6%, all in Australia) (Fig. 1A). In the time period selected for the survey (1992–2022), only 2% of the documents was published in the decade 1992–2001, 7% in the decade 2002–2011, while 91% in the period 2012–2022 (of which 86% in the decade 2012–2021 and 5% in the first three months of 2022, when this literature survey ended) (Fig. 1B). Concerning the type of green infrastructure, most of the experiments were performed on urban vegetation (39%, in this review intended as all the vegetation located in urban areas, such as residential green areas, university campus, etc.) and roadside vegetation (25%, intended as all the vegetation located along the streets), while few studies were carried out on urban parks (12%), green walls (11%), and other types of greening (e.g., indoor vegetation, green barriers, wetlands, green roofs, urban woodlands and wastelands, <5% each one). Trees were the type of vegetation most investigated (47%), followed by shrubs (30%), herbaceous (12%), climbers (9%), mosses (1%); 1% of the articles did not specify this aspect. The experimental conditions of the studies were mainly real (*in situ*, in urban sites, 71%), followed by controlled (25%) and nursery (3%, considered as a condition between

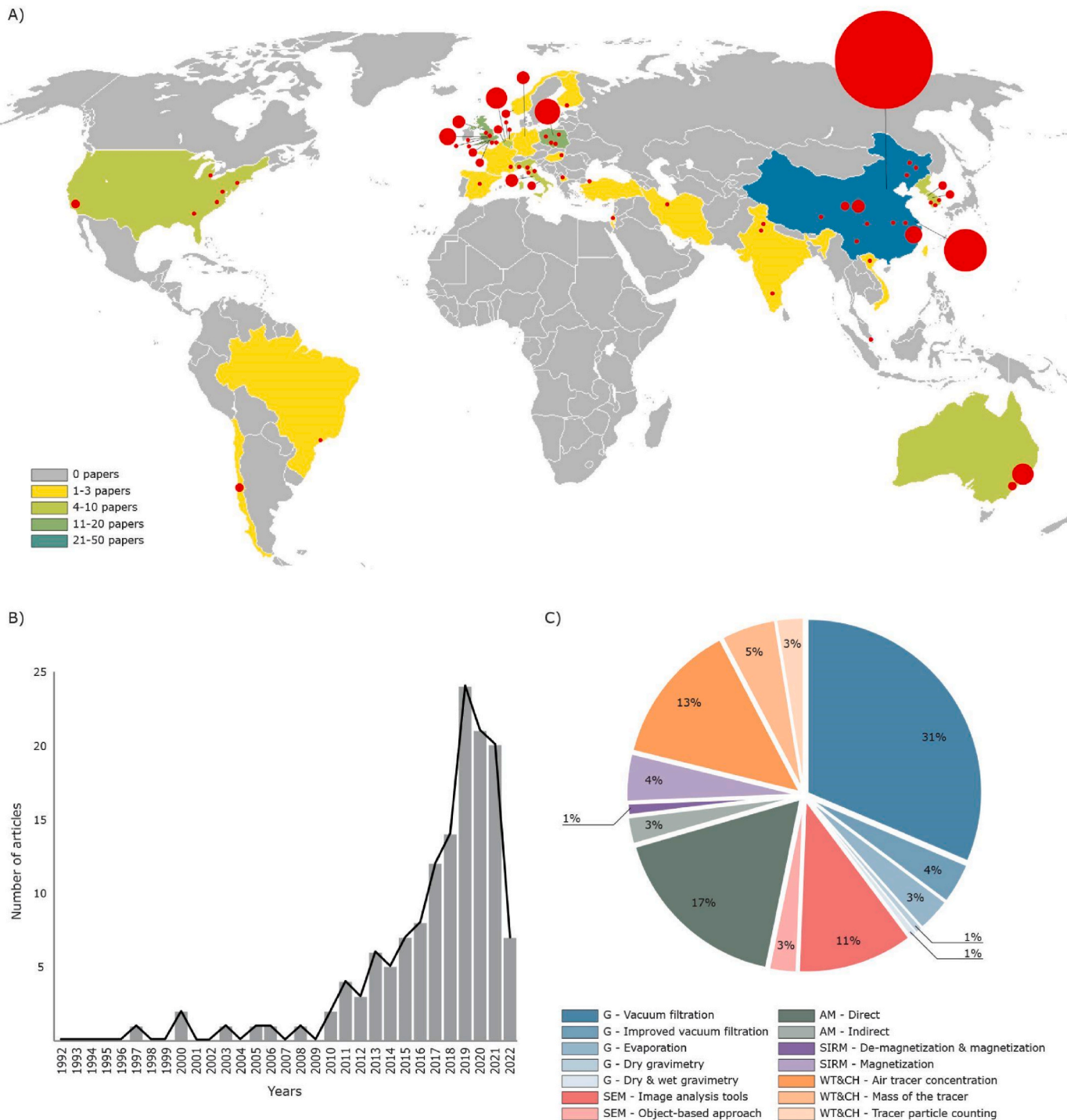


Fig. 1. – A) Map of the geographical context of the articles analyzed in this review. Different colors represent the different number of articles found in a specific country. The red dots represent, according to their size, the number of items found for a given city. B) Number of articles found per year from 1992 to 2022. The line represents the trend during the thirty-year period. C) Percentage distribution of the reviewed articles based on methods (and their declinations).

real and controlled); only 1% of the works was carried out in both, real and controlled conditions.

Concerning the method used to assess the vegetation effectiveness in PM removal in urban areas, the methods and their declinations, which will be later explained, were in the following order: the gravimetric method was the most found (G, 40%) in accordance with Corada et al. (2021), followed by aerosol monitor (AM, 20.5%), wind tunnels and deposition chambers (WT&CH, 19.5%), Scanning Electron Microscopy (SEM, 14%) and Saturation Isothermal Remanent Magnetization (SIRM,

6%) (Fig. 1C). In some cases we found different methods within the same paper. In this review we named ‘combined methods’ all the secondary methods (classified in this review, i.e., G, SEM, AM, WTs & CHs, SIRM) or instruments or techniques which strictly depended on a main method and which allowed to: i) refine the main method; ii) obtain parameters different from those that would be obtained using the main method only. We found that 24% of the articles used combined methods. Specifically, by considering the overall articles, the combined methods were found mostly in WT&CH (16%, of which 15% AM and 1% G), followed by G

(4%, of which 2% SEM, 1.5% AM and 0.5% SIRM), AM (3%, all G) and SIRM (1%, all AM); in this review the SEM method was never found in combination with other methods. Finally, we found that 9% of the articles used more than one method for the quantification of the vegetation effectiveness in PM removal (named here 'parallel methods').

3.2. Gravimetric method (G)

A gravimetric analysis identifies a set of techniques used in analytical chemistry for the quantitative determination of an analyte based on its mass. Therefore, in this review we grouped all the articles based on PM mass measurements under the 'gravimetric method'.

3.2.1. Experimental conditions

Most of the analyzed studies were conducted under real conditions (82%), i.e. on the vegetation planted in urban areas (along roads, in parks or forests, wetlands, wastelands, green walls, experimental fields and university campus) in open field. Few works were carried out under controlled conditions (8%), i.e., on vegetation grown in indoor controlled environments (e.g., greenhouses or rainfall simulation systems). 3% were performed under both real and controlled conditions. We grouped experiments carried out on potted or planted plants, irrigated and or fertilized in nursery, under the nursery category (7%), which represent a condition between real and controlled.

3.2.2. Sampling and preparation procedures

Sample material included different plant organs: mostly leaves (95%, e.g., Freer-Smith et al., 1997; Sæbø et al., 2015; Vigevani et al., 2022) but also bunches (Wang et al., 2013; Wang and Shi, 2021), branches (Xu et al., 2019b; Zhang et al., 2019), twigs (Kwak et al., 2020; Xu et al., 2017, 2019a), shoots (Przybysz et al., 2014, 2020) and trunk (Catinon et al., 2012; Xu et al., 2019b). One study (Cai et al., 2019) sampled the throughfall (rain passing through tree crowns or canopies). The sample dimension varied across plant organ area (46%, cm²), number (36%, n.) and mass (3%; g of fresh weight). Few studies reported only length of twigs and shoots or number of internodes for branches (Xu et al., 2019a, 2019b), while others did not report information about sample dimension. The weather conditions during sampling, when specified, were mainly dry, and many authors performed the sample collection after a certain number of non-rainy days (here in the range 1–37 days).

The gravimetric method requires a cleaning step, which consists in the mechanical removal of PM from the sample surface. This phase was carried out in different ways. Most of the studies only wash the sample surface with water (59%), while others only brush (3%) or clean ultrasonically (3%). In addition, some studies combined more than one cleaning method: the most common combination was wash and brush (28%), while the less used were wash and ultrasonic cleaning (3%), wash, brush and ultrasonic cleaning (3%), wash, brush and tissue paper (1%). Distilled water was widely used in wash cleaning, followed by deionized and unspecified water; other less used types were ultrapure, microfiltered and deionized, mineralized ultrapure distilled, purified and reverse osmosis. Different approaches were used in water wash cleaning: soaking, manual or automatic agitation (with oscillators, magnetic stirrers, vortex stirrers, shakers) and centrifugation. The brush tool, when specified, was generally no-hair loss, non-depilatory, soft and fine and the made material was natural (camel hair) or synthetic (nylon or plastic); some specific tools mentioned were paint and banister brushes. Ultrasonic cleaning describes a procedure in which the sound energy of the ultrasonic frequency is converted into mechanical vibration of a fluid, providing a cleaning effect. It should be noted that the above-mentioned procedures remove only particles deposited on plant organ surfaces (surface PM), while particles trapped in the leaf wax layer (in-wax PM) can be extracted by chloroform washing, as performed by 43% of the authors.

3.2.3. Declination of gravimetric method

Within the gravimetric method, we identified five declinations (based on common procedures adopted): the main was the Vacuum Filtration (VF; 80%), followed by Improved Vacuum Filtration (IVF; 10%), Evaporation (E; 8%), Dry Gravimetry (DG; 1%) and Dry and Wet Gravimetry (DWG; 1%). We named the different declinations with names known, introduced by the authors or created by us.

3.2.3.1. Vacuum filtration (VF). Vacuum filtration (VF) is a technique for separating a solid from a liquid; the solid is trapped by a filter and the remaining solution is removed by the action of a vacuum pump, which increases the rate of filtration providing an air force on the solution in addition to the gravity force. Therefore, this procedure allows the quantification of the PM captured on plant organs by the difference in weight of filters before and after filtration.

Filters (especially hydrophilic ones) require stable conditions (T and RH) to reduce potential errors linked to environmental changes during the weighing process (usually carried out with a precision balance). Most of the studies dried filters in ovens or drying chambers (40°–105 °C; 30 min–3 days) and then stabilized them in rooms, boxes (even desiccators) or chambers (T 20°–25 °C and RH 25%–50%; 10 min–48 h). In some cases, only one of these two steps were carried out. To avoid electrostatic charges on the filters, some authors passed them through a deionizer gate before weighing.

The sampled area for vacuum filtration was generally in the range 100–700 cm² and it was determined by authors mainly after filtration (only in few cases it was done before; e.g., Muhammad et al., 2022) with the methods described in the section below.

For the wash cleaning (also when combined with the other cleaning methods), every sample was placed in a container with water (50–800 ml) and soaked and/or agitated for a variable time period (1–80 min). Authors which used ultrasonic cleaning used 150–250 ml of water for a time range of 1–20 min. For brush cleaning, authors usually washed, scrubbed and then flushed sample surfaces with additional water (5–100 ml) or simply scrubbed samples dipped in the water. After washing with water, some authors washed leaf samples with chloroform (50–300 ml) for a few seconds (10–60 s) to dissolve the epicuticular wax layer from leaf tissues and to wash out particles embedded in waxes. Due to the toxicity of chloroform, some authors performed this step under a fume cupboard (Li et al., 2020).

The obtained solution was then ready for the filtration process. To eliminate larger particles, many authors used a sieve (usually with a mesh diameter of 100 µm). The solution was next filtered on different types of filters (pre-weighed) often using a 47 mm glass filter funnel connected to a vacuum pump. Filters can be of different size porosity (100 µm, 20–22 µm, 6.5–13 µm, 2–4 µm, 0.1–0.45 µm) and material (mainly paper but also cotton, polycarbonate, nitrocellulose, nylon, mixed cellulose ester, cellulose nitrate, alumina, glass, PTFE) but most of the authors filtered in sequence through 10 µm, 2.5 µm and 0.2 µm, providing 3 p.m. fractions (large: PM_{10–100}; coarse: PM_{2.5–10}; fine: PM_{0.2–2.5}) and the main material was paper (for 10 µm and 2.5 µm filters) and PTFE (for 0.2 µm ones). When filtering the water solution, usually a few droplets of isopropyl alcohol were placed on the PTFE membranes before filtration to reduce surface tension and speed up the process. The filtration procedure of the chloroform solution was the same as for the water solution, except for isopropyl alcohol which was not used (Dzierzanowski et al., 2011). Particular attention is needed for the filter material, which could be damaged by chloroform, as nitrocellulose filters, which were replaced with PTFE ones for this reason (Xu et al., 2018).

At this stage, filters could be weighed as did before filtration to calculate the mass of PM per difference. Some authors used blanks, i.e., control filters, which ensured any changes in humidity that could affect filter weight were accounted for (Haynes et al., 2019).

3.2.3.2. Improved vacuum filtration (IVF). Improved vacuum filtration (IVF) is a 2-step process, where vacuum filtration is preceded by drying (in oven or vacuum freeze dryer) a sub-sample of solution (stored in beakers, petri dishes, plastic bags or test tubes), at a variable temperature (40–105 °C), until the water evaporated completely, to determine the mass of total suspended particles (TSP). It was followed by a second phase of vacuum filtration of the remaining solution to determine the mass of large and coarse PM fractions (PM_{\times}), using filters of 10 and 2.5 μ m porosity, respectively. TSP and PM_{\times} were obtained by proportion to the total solution, while the fine fraction (<2.5 μ m) was obtained by difference. This method was applied by the authors only for the quantification of surface PM, not for in-wax PM.

3.2.3.3. Evaporation, dry gravimetry, dry & wet gravimetry (E, DG, DWG). Evaporation (E) is a basic gravimetric technique to determine the total mass of PM, without the possibility to discriminate among different PM size fractions, unless with an additional instrument (such as laser particle analyzer or laser granularity instrument). It consists in drying the washing solution and in quantifying PM by the weight difference of the boxes containing the washing solution (beakers, petri dishes or centrifugal tubes) before and after drying.

We called dry gravimetry (DG) a very simple technique, used by Paull et al. (2020), carried out by weighting the sample before and after a dry removal (brushing) of particles from its surface.

The last method identified was named dry & wet gravimetry (DWG), performed only by Singh et al. (2020), which estimated the PM accumulated on the sample surface averaging the results of three gravimetric methods: the first is equal to dry gravimetry above mentioned, the second is the same but with wash cleaning instead of brush cleaning, and the third is based on the weight difference between the box filled with the washing solution and the same box containing only water. The analyzed area for these three declinations were in the range 200–400 cm^2 .

3.2.4. Combined methods

We found that 10% of the papers classified in the gravimetric method performed gravimetric analysis in combination with the other methods classified in this review. Some authors used SEM in combination with VF (Beckett et al., 2000b; Freer-Smith et al., 1997; Sillars-Powell et al., 2020) or fluorescence microscope in combination with DG (Paull et al., 2020) to discriminate different PM size fractions, measuring particles diameter. Moreover, Freer-Smith et al. (1997) and Sillars-Powell et al. (2020) used SEM also to provide particle number density instead of or in addition to PM mass, respectively. Other authors used AM to quantify air PM concentration and provide, in combination to VF, the deposition velocity (Freer-Smith et al., 2005; Muhammad et al., 2022; Sillars-Powell et al., 2020) or used SIRM to quantify the magnetic signal of PM retained on filters obtained by VF (Muhammad et al., 2022). In the pool of papers analyzed in the gravimetric method, we found that 15% used additional techniques in combination with gravimetric analysis to quantify the different types of deposited PM.

'Surface PM' represents particles retained on the plant surfaces which can be subjected to dry (blowing off by strong winds) or wet (washing off by rain) removal and thus retained temporarily. These particles could be water insoluble or soluble and inorganic or organic. In this review, when not specified, we referred to insoluble PM. 'Water soluble PM' included inorganic ions and organic material. Authors quantified inorganic ions, such as Cl^- , NO_3^- , NO_2^- , PO_4^{3-} , SO_4^{2-} and F^- (anions) and Ca^{2+} , Mg^{2+} , K^+ , Na^+ and NH_4^{+} (cations), in different ways. Beckett et al. (2000b) and Freer-Smith et al. (2005) quantified anions and cations on the residual solution obtained after the vacuum filtration for insoluble PM with ion chromatography and atomic absorption spectrometry, respectively, while Xu et al. (2018) and Cao et al. (2022) quantified both anions and cations with ion chromatography. Ristorini et al. (2020) analyzed the leaf washing solution by ion chromatography

for the detection of anions, and by inductively coupled plasma mass spectrometry (ICP-MS) and UV-Visible spectrophotometer, the latter for NH_4^{+} , for the detection of cations. They also compared the total ionic concentration obtained to the Electrical Conductivity (EC) of the leaf washing solution measured by a conductivity meter, to test its suitability as a proxy for the quantification of the water-soluble and ionic fraction of leaf deposited PM. Regarding the water-soluble organic material, Ristorini et al. (2020) quantified it using a non-purgeable organic carbon procedure and Cao et al. (2022) using a total organic carbon analyzer. Wang and Shi (2021), instead, quantified the total dissolved solids (TDS) in the elution after vacuum filtration with a conductivity meter, referring to TDS as all inorganic and organic substances contained in a liquid in molecular, ionized or microgranular suspended forms which can survive filtration through a filter/membrane with 2 μ m pores. All the above-mentioned works quantified the 'water insoluble PM' with vacuum filtration and the 'water soluble PM' with the above-mentioned techniques. In the papers which quantified 'water insoluble PM' with evaporation, instead, some studies separated the supernatant from the precipitation to distinguish the water-soluble PM from the water insoluble PM, respectively (Yue et al., 2021), while other works remove the supernatant and quantify only insoluble particles (Catinon et al., 2012; Heshmatol Vaezin et al., 2021).

'In-wax PM' represents particles embedded in epi-cuticular wax layers in which resuspension by wind or removal by rain would be negligible (Muhammad et al., 2022). All the papers analyzed quantified it through vacuum filtration method, using chloroform instead of water to dissolve the wax layer and extract the trapped PM.

3.2.5. Sample area measurement

In most of the studies, the PM amount retained by the sample was expressed per unit sample area. When leaves were sampled, authors quantified one or two sides of the leaf surface (15% and 18%, respectively), even if in the remaining 67% of the studies this information was not available, not clear in method details or not applicable (e.g., when sample material was different from leaves). Considering petioles as part of the leaf area measurements was another point often unspecified, except for few studies which expressly reported to measure only the leaf blade (Li et al., 2020; Paull et al., 2020; Popek et al., 2017; Yue et al., 2021; Zhou et al., 2020).

For broad-leaved species, the leaf area of each sample was quantified mainly by an Image Analysis System (64%), which used a scanner (or a video camera, or seldom an optical microscope) and a software to measure the scanned leaf area. Softwares used, in order of frequency, were Image J, Photoshop, Skye, WinRHIZO, image analyzer, WinDIAS, Black spot, Zhejiang Advanced Instrument, WinFOLIA, Leaf Area Measurement, DLT-Cam Viewer. Some studies (20%) used a Leaf Area Meter (mostly Licor, in only in two cases a DeltaT-device was used), an instrument which directly digitized area, length, and width of leaves. Few works (5%) used both the above-mentioned methods, mainly due to the limitations of some leaf area meters in measuring small leaves. Only one research used the graph paper method, a basic technique which used a millimeter graph paper to quantify the leaf surface area.

For needle- and scale-leaved species, the leaf area was calculated mainly by Image Analysis System and equations or water displacement method (15% or 11% of all the analyzed papers, respectively). Image Analysis Systems allowed to obtain geometric parameters needed for leaf area calculation. Some studies used based-shape equations. For flat needles and scales, only scanned area (one side or two sides) was used (e.g., Li et al., 2021, for *Metasequoia glyptostroboides*; Yue et al., 2021, for *Sabina chinensis*). Leaf area of needles with semi-circular cross-section (e.g., *Pinus tabuliformis* in Cao et al., 2022; Yue et al., 2021) was measured according to the following equation: $S = PA + 1/2 SA$, where PA is the projected area (the longitudinal surface of the flat portion) and SA is the surface area (the longitudinal surface of the semi-circular portion). For cylinder-shaped needles (*Pinus nigra* in He et al., 2020a; He et al., 2020c) half of the lateral surface of a cylinder was calculated as

follows: $S = \pi dl/2$, where d is needle diameter and l is needle length. For cone-shaped needles, Liu et al. (2018) and Yue et al. (2021) calculated the leaf area by using the diameter and length of the leaves manually measured. Beckett et al. (2000b) and Freer-Smith et al. (2005) used Needle Pair Area (NPA) method for *Pinus nigra*: $NPA = \pi dl + (2 \times dl)$, where n is number of needles, d is needles diameter and l is needle length. Finally, Vigevani et al. (2022) used Leaf Mass per Area (LMA) for *Pinus nigra* and *Pinus pinea*, an approach based on measuring weight (after drying and weight stabilization) and leaf area of a sub-sample to obtain the total leaf area by proportion. Water displacement was the other method mainly used. It consisted in measuring the water displacement for leaf volume and converted the volume to leaf area according to the following formula: $S = 2L \cdot (1 + \pi/n) \cdot \sqrt{(nV/\pi)}$, where S is leaf area, V is water displacement volume as the substitute of needle–leaf–volume, n is the number of needles in a single bundle and l is the average length of the needles.

When samples were branches, authors considered them as cylinders, calculating their surface measuring length and diameter with a Vernier Calliper (Xu et al., 2019b; Zhang et al., 2019). The two articles which treated the trunk sampled a fixed area of bark tissue of known geometric shape (Catinon et al., 2012; Xu et al., 2019b).

3.2.6. Parameters

The gravimetric method mainly provided parameters expressed as mass of PM per unit sample area (93%), mainly per unit leaf area ($\mu\text{g cm}^{-2}$, mg cm^{-2} , g m^{-2} , mg m^{-2}), but also per bark unit area ($\mu\text{g cm}^{-2}$, mg dm^{-2}) or upscaled at tree level (g tree^{-1}) or at a larger scale ($\text{mg green barrier section}^{-1}$, kg hm^{-2} green area). In some cases, the mass of PM per unit leaf surface was also expressed per unit time, providing the daily PM accumulation (8%; $\mu\text{g cm}^{-2} \text{day}^{-1}$, $\text{g m}^{-2} \text{day}^{-1}$, $\text{mg cm}^{-2} \text{day}^{-1}$). In most cases, this parameter was calculated mainly by sampling after a certain number of days after an effective rainfall (i.e., the one assumed to clean the leaves) and dividing PM accumulation per those days (Chen et al., 2016; Guerrero-Leiva et al., 2016; Vigevani et al., 2022). Only Singh et al. (2020) directly measured the daily accumulation by cleaning marked leaves, which were analyzed after 24 h of accumulation. In one case the mass of PM was expressed per unit dry weight (Haynes et al., 2019; $\text{mg dry weight}^{-1}$ of tree leaves or mosses). Combining different methods to the gravimetric one allows to obtain different parameters comparable to those obtained by the other methods. Freer-Smith et al. (1997) and Sillars-Powell et al. (2020), for example, combined SEM analysis to the VF procedure providing the particle number density parameter (number of particles per unit filter surface) instead or in addition to PM mass, respectively. Other authors used AM to quantify air PM concentration and provide, in combination to VF, the deposition velocity (cm s^{-1} , Freer-Smith et al., 2005; Muhammad et al., 2022; Sillars-Powell et al., 2020) or used SIRM to quantify the magnetic signal of PM retained on filters obtained by VF (Muhammad et al., 2022). The fractions of PM assessed ranged from ultrafine particles to total suspended particles.

3.2.7. Considerations

The gravimetric method cleans the vegetation organs to remove the trapped PM and measure its mass by weighing, mainly providing the mass of PM per unit sample area.

- *On sampling: a standardization of leaf sample area is needed; not only leaves should be studied; weather conditions for sample collection depend on study aims*

Although other vegetation organs could play a role, which should be further investigated, leaves are considered responsible for most of the PM trapping (Vigevani et al., 2022). Thus, it was not surprising that leaves were the most used sample material, which in our mind offers the possibility to make comparisons across different plant species regardless of the vegetation type (e.g., trees, shrubs or herbaceous). A fixed

dimension, however, should be selected, in order to be representative, suitable for comparisons and for the method declination procedures. Some authors reported collecting a certain number of leaves, others a target leaf area, and some others carried out preliminary tests to select the most appropriate standard area (e.g., Yue et al., 2021), which is preferable. It should be noted that the weather conditions during sampling should be selected depending on the aim of the study. For example, if the intention is to quantify the species efficiency in PM removal, standard weather conditions should be selected; if the intention is to investigate how the meteorological factors affect the PM cycle on vegetation, diversified weather conditions and temporal scales should be considered.

- *On preparation procedures: adding ultrasonic cleaning (UC) to water and brush cleaning (WC and BC) provides more accurate surface and in-wash PM quantification*

The cleaning (i.e., the mechanical removal of PM from the sample surface) is the first main step required by all the declinations of the gravimetric method. It can cause systematic errors. For example, appendages on the leaf surfaces can be broken during the cleaning and weighed as PM, causing over-estimations (Xu et al., 2020).

The water cleaning (WC), the most used also in combination to the brush cleaning (BC), does not allow PM from the abaxial and adaxial surfaces to be distinguished (Freer-Smith et al., 1997). Moreover, the amount of detectable PM depends on the type and quantity of water and on the washing approaches and times. Distilled water, the most used water type found in this review, seems to be the more appropriate choice due to the absence of saline components (as well as deionized water) and its sterility. These characteristics allow control of the variable conditions and reveal the dynamic changes of PM washed off by plant organs (Cai et al., 2019). Almost 40% of the authors performed a low intensity washing, agitating the samples in a variable quantity (150–300 ml) of water only for 1 min, obtaining a solution containing only the particles poorly bound to the sample surface (e.g., Dzierzanowski et al., 2011; Leonard et al., 2016; Popek et al., 2022). These represent the fraction of surface PM easily rinsed off by rain under natural conditions (Kończak et al., 2021), which, although suitable for across species comparison, does not allow to collect all the particles present on the sample surface, which is the premise to accurately evaluate the plant PM retention capacity (Yue et al., 2021). Thus, other authors performed a higher intensity washing, agitating (even automatically, and even performing a centrifugation at 3000 rpm or more) the sample in the water (in a quantity in the range 50–500 ml) for greater time (e.g., 3 min, 10 min, 1 h and 20 min Repeated 4 times, in Beckett et al., 2000b; Esposito et al., 2020; Freer-Smith et al., 1997; Simon et al., 2020; respectively), also performing preliminary tests (e.g., Muhammad et al., 2022). Regardless of the washing intensity, Wang et al. (2015) pointed out that the entire washing procedure should take less than 10 min, to minimize the dissolution of water soluble PM.

Adding brush to the water cleaning can increase the amount of PM removable from the sample surface. Some authors after that washed again the sample surface with an additional quantity of water (e.g., Beckett et al., 2000b; He et al., 2020a; He et al., 2020b; Wang et al., 2015) and/or used an additional quantity of water to remove any PM from the sample bag and other residual PM wash from equipment (e.g., Sillars-Powell et al., 2020). However, it should be noted that the BC, especially when performed alone (without water use), could damage the functional traits involved in PM trapping (e.g., trichomes) causing over-estimates (weighing as PM trichomes residues) and/or induce particles resuspension causing under-estimates (not weighing the resuspended PM). Liu et al. (2018) nevertheless, after performing WC and BC, still found 29%–46% of PM remaining on leaves of the different tested tree species, highlighting the inefficacy of conventional cleaning methods.

They found, instead, that after adding the ultrasonic cleaning (UC),

the residual PM (especially the smaller particles) was removed almost completely, as also indicated by the SEM photographs of the leaves performed at the different cleaning steps. Specifically, UC washed out the particles tightly adhered to the furrows and grooves of both broad-leaf and coniferous leaves. Moreover, Yue et al. (2021) demonstrated the efficacy of adding UC in improving the recovery of both, water-insoluble (WIPM) and water-soluble particles (WSPM) within all diameter classes from leaf surface. Specifically, they interestingly found that the UC elution effect (i.e., the eluted PM proportion) was much larger for WSPM (54%) than for WIPM (31%), maybe because, after water and brush cleaning, the WIPM could disperse into small particles, while WSPM could resolve into ion, thus having more chances to reenter the leaf surface furrows, grooves, or even stomata and then being washed out by UC. Thus, a relatively higher proportion of WSPM can be washed out when cleaning the leaves by ultrasonic after the conventional cleaning methods. Overall, adding UC to the conventional cleaning methods (WC and BC) can be extremely necessary to accurately quantify the PM retention capacity of plants. These procedures are only valid for the surface PM.

However, many studies have shown that PM in leaf waxes accounts for a significant part of PM retained by plant leaves. For example, 40% of the total PM can be embedded in the wax layer (Popek et al., 2013), even with differences among species (in Sæbø et al., 2012, *Betula pendula* and *Fagus sylvatica* accumulated 82.6% and 25% of PM in the wax layer, respectively). Some authors decided only to assess surface PM and not the in-wax PM due to environmental health concerns of using chloroform (e.g., Chen et al., 2016), which is able to dissolve the majority of the leaf waxes and to release the particles embedded in the cuticle (Haynes et al., 2019). However, surface PM results, even if suitable for species comparison, lead to underestimate the total plants PM removal. Thus, to accurately assess the vegetation effectiveness in air PM mitigation, both, surface and in-wax PM should be investigated. Also for the in-wax PM quantification, it is appreciable to select the amount and duration of chloroform washing based on preliminary tests (as performed, for example, by Dzierzanowski et al., 2011; Przybysz et al., 2020). Still, Yue et al. (2021) pointed out that the leaf wax PM can be overestimated if the leaf surface PM is not eluted completely. This is because the residual PM on the sample surface could be eluted into the chloroform when dissolving the leaf waxes with chloroform. For this reason and for the above-mentioned ones, ultrasonic cleaning seems to be an emerging and important procedure to accurately quantify the PM retention capacity of plants.

- *On vacuum filtration (VF): a standardization of filters stabilization and weighing phases is needed; particles geometrical diameter and filters saturation can cause errors in water insoluble particles (WIPM) estimation; water soluble particles (WSPM) can be evaluated only with a combined technique; PM_{2.5} quantification is approximated (particles smaller than the lower filter porosity can't be measured)*

Among the pioneers of the most used gravimetric method, the vacuum filtration (VF), there is the work of Dzierzanowski et al. (2011) (the most cited reference for the methodology in this declination), which defined a protocol to study the deposition of PM of different size fractions on the leaf surfaces and in waxes of urban forest species. One of the steps introduced compared to the previous works (Beckett et al., 2000b; Freer-Smith et al., 1997, 2005) was the use of a sieve, which is needed to exclude particles larger than 100 µm, which are not defined as PM. Later authors introduced some modifications to the protocol. For example, Zhang et al. (2019) and Wang and Shi (2021) soaked the filters before drying them to remove any possible soluble impurity deriving from manufacturing in the factory, which otherwise could have resulted in overestimates.

For stabilization and weighing of filters, Muhammad et al. (2022) followed the European Standard Guidelines (FprEN 12341:2013) which, even if referred to sampling PM on filters of aerosol monitor devices, it is

generally applicable to filters derived from any kind of procedure of collecting PM on filters in our mind. According to Muhammad et al. (2022), the procedure basically consists in stabilizing (in a climate-controlled room with an average relative humidity of 50% and temperature 21 °C) and subsequently weighing filters with a 1 µg precision balance (both, clean and loaded filters) 2 or 3 times in order to meet specific weighting criteria. Other authors measured every filter three times, to reduce the potential errors (e.g., Sillars-Powell et al., 2020; Wang and Shi, 2021; Zhang et al., 2019). Others performed an additional step in deionizer gates before weighing, to avoid electrostatic charges on the filters, which could lead to overestimates. Still, others used blank filters to control possible influence of the environmental variables on the filter's weight, especially hydrophilic ones. In conclusion, even if based on preliminary tests (which are preferable), the stabilization (mode and time) and weighing procedures were found to be performed in many different ways depending on the authors. This highlights the need to set a common and detailed procedure ever applicable.

Regarding the filtration process, an issue could be the saturation of the filters. This can occur if the solution is very concentrated, which could happen if an excessive quantity of polluted sample area is cleaned in a small amount of water. Saturation of the filters can lead to an overestimation of the larger PM particles, which can clog the pores of filters trapping with them also finer particles; subsequently, finer PM particles can be under-estimated. In order to obtain sufficient material to determine the finer PM and still avoid filter blockage by particles during filtration, Dzierzanowski et al. (2011) fixed a sample leaf area in the range 300–400 cm². However, a wide range of sample areas were found to be washed and filtered in this review, highlighting that this variable should not be fixed *ex ante* but rather selected depending on the specific conditions of every study. Other authors washed the sample in a large amount of water (e.g., 800 ml), then filtered only a sub quantity of the shaken washing solution (e.g., 100 ml) and finally normalized the resulting filter weight to the total volume and washed sample area (Hofman et al., 2014a). For the smallest porosity filters (e.g., 0.2 µm pore size), those more subjected to the saturation due to the little filter dimension and porosity, Hofman et al. (2014a) used two filters to avoid their saturation. These solutions are favorable, even because they save time. The vacuum filtration, for its intrinsic methodologic characteristics, is based on the geometrical diameter (Esposito et al., 2020) and not on the aerodynamic diameter of the particles which define the air PM size classes. The geometrical diameter is the diameter of a spherical particle that has an equal surface of the particle under consideration. The aerodynamic diameter, instead, is the diameter of a perfectly spherical particle of unitary density (1 g cm⁻³) which has the same inertial characteristics as the particle under consideration. Air PM particles are often not spherical and their shapes can be extremely diverse and irregular. This can cause an over- or under-estimation of the different PM fractions in the VF. Indeed, depending on how particles reach the filter's pores, they can pass through or remain trapped, being part of the larger or finer fraction. For example, if a particle with an oblong shape and a geometrical diameter >10 µm reaches the filter pore of 10 µm horizontally oriented, it can be trapped, being weighed in the right PM fraction. If, instead, it reaches the filter pore vertically oriented, it can pass through the pore, being weighed erroneously in the finer fraction. Sillars-Powell et al. (2020) performed a particle size analysis (PSA, based on using SEM and Image J software) on filters theoretically containing the 2.5–100 µm fraction obtained by VF and found a high proportion of particles with a physical diameter <2.5 µm. Although they hypothesized that this occurred due to the coarse measurement technique involved with their PSA method, we guess that the saturation concern discussed above and also the issue linked to the geometrical diameter could have played a role in their findings.

Another limit of the VF is that it mainly provides the isolation and measurement of the water insoluble particles (WIPM) (Wang et al., 2015). Water soluble particles (WSPM), which could dissolve in the

filtration solution (Cai et al., 2019; Ristorini et al., 2020), represent 9–50% of the total surface PM (Song et al., 2015; Xu et al., 2018; He et al., 2020b; Wang and Shi 2021) depending on plant species (Xu et al., 2018) and PM source (He et al., 2020b). The gravimetric method alone does not allow to distinguish WSPM in different size fractions and it is difficult to extract the PM dissolved in the large quantity of water deriving from the wash cleaning steps (Song et al., 2015). This aspect may result in an underestimation of the effect of plants on PM deposition (Wang and Shi, 2021) when gravimetric method alone is used. Moreover, during this procedure, the water-soluble ions present in the leaf may leak out causing a further variation of the results (Song et al., 2015). The only way to estimate the amount of the soluble fraction over the total surface PM is coupling the gravimetric with a combined technique (e.g., ion chromatography, atomic absorption spectrometry, inductively coupled plasma mass spectrometry and UV-Visible spectrophotometer, Electrical Conductivity). Among these, Electrical Conductivity performed on the washing solution, seems to be a fast and easy to apply technique to assess the WSPM, even if it seems to be reliable only when there is not a substantial presence of biological matrices on the leaf surfaces, such as honeydew or resins (Ristorini et al., 2020).

Finally, also not considering the lower porosity limit of the filters represents an approximation. Specifically, some authors assume that, for example, the fraction of PM in the size range 0.2–2.5 μm was equal to $\text{PM}_{2.5}$, thus approximating this fraction to all the particles with diameters below 2.5 μm even if particles smaller than 0.2 were not measured. Other authors stated that particles smaller than the lower porosity limit of the filters were not considered (e.g., Wang et al., 2015). In addition, Cai et al. (2019) underlined that shaking, washing, filtration, and drying in the laboratory, all processes included in the VF method, can determine particle morphology change, particle size conversion, and soluble PM dissolution because of erosion, dissolution and hydration, thus highlighting the potential uncertainty linked to the VF method.

- *Improved vacuum filtration (IVF) is a gravimetric declination which can overcome VF limitations on $\text{PM}_{2.5}$ quantification*

Based on the original protocol for VF of Dzierzanowski et al. (2011), the improved vacuum filtration (IVF) method was developed. The pioneers of this method appeared to be Liu et al. (2014) (cited in He et al., 2020a; 2020b; 2020c; not included in this review due to unavailability reasons) and Hong et al. (2015) (cited in Zhang et al., 2017; not included in this review because in chinese). Although many of the observations mentioned above are valid also for IVF, this method can overcome some of the VF limitations. Specifically, not using the smaller filter porosity (e.g., 0.2 μm) and quantifying $\text{PM}_{2.5}$ per difference between TSP and $\text{PM}_{2.5-100}$, allows the measurement of all the particles smaller than 2.5 μm , overcoming the issue linked to the lower porosity limit of the filters. Moreover, the clogging of the smaller filter porosity (e.g., 0.2 μm), the most sensitive to this problem, will not occur, saving time and reducing the use of laboratory consumable materials. However, the calculation of $\text{PM}_{2.5}$ per difference is based on the use of a subsample of solution (8–12%), assumed as representative of the entire solution. Thus, both for subsample analysis and for vacuum filtration it is important to use a homogeneous solution. IVF was used in few cases in this review, but it seems a promising gravimetric method compared to VF, even if comparative studies between the two methods should be performed.

- *The other gravimetric method declinations (E, DG, DWG) appear inaccurate*

The evaporation (E), dry gravimetry (DG) and dry & wet gravimetry (DWG) appear to us as simplified and inaccurate methods, which need combined techniques to distinguish particle size fractions. Even if they can result faster, a compromise method between accuracy and operational time should be used.

- *On sample area measurement: a standard approach is needed*

Leaves are the most used samples, and the different procedures to measure the leaf area of the samples used for the gravimetric analysis could be a source of variation in the results, making difficult the comparability of different works. For example, to measure the surface area of the cylindrical needles of the same species, *Pinus nigra*, three different methods emerged in this review: needle pair area (Beckett et al., 2000b; Freer-Smith et al., 2005), lateral cylinder surface formula (He et al., 2020a), leaf mass per area (Vigevani et al., 2022). There is no evidence on which method could be the best, but more consistency is needed to reduce the error in comparing results of different experiments. Regarding the leaf area measurements, a source of deviation in the results could be the instrument used, such as leaf area meter. Some models detect only one side of the leaf (Sæbø et al., 2012), while others measured both sides simultaneously (Paull et al., 2020); some models are portable and do not provide accurate measurements of small leaves (Li et al., 2021), while others are stationary and do not have this limit although they cannot be used directly in the field. Moreover, when an Image Analysis System was used, the surface area obtained from scanning the leaf is a proxy of projected area, because it does not consider the veins and other structures on the leaf blade in determining the surface area (Hwang et al., 2011). These aspects should be considered when comparing the results of different studies which use different approaches in measuring leaf area.

- *On parameters: one sample side (e.g., leaf) should be considered; PM daily deposition is more comparable among studies and upscalable in time; different PM fractions can be provided*

An issue linked to the largely used wash cleaning is the number of the leaf sides considered to express the final parameter. Indeed, the wash cleaning involves the whole leaf, isolating PM from both ad- and abaxial side (two sides). This amount could be referred to one (Dzierzanowski et al., 2011 and most of the studies reviewed here) or two sides (He et al., 2020b) of surface area. This step can largely influence the comparability of results and must be checked when different studies are compared. In our opinion, it is appropriate to refer the amount of PM obtained by the wash cleaning to a single side of the leaf, which captures PM from the two sides with different retention capacities (Weerakkody et al., 2017). This is usually performed also in other physiological studies (Dzierzanowski et al., 2011).

The mass of PM expressed per unit sample area was the parameter most found in the gravimetric experiments. It represents an instantaneous situation of the PM cycle, thus the samples must be properly collected considering different periods of the year and different environmental conditions, to be representative and comparable among the different works (Wang and Shi, 2021). To overcome this limit, some authors expressed PM mass or number of particles per unit leaf area and unit time, assessing the PM daily deposition (Chen et al., 2016; Guerrero-Leiva et al., 2016; Liu et al., 2018; Singh et al., 2020; Vigevani et al., 2022). This parameter also provides the possibility of upscaling the benefit in a specific time period (e.g., month, year).

Finally, the gravimetric method allows to divide PM into specific diameter classes (Yan et al., 2016b), such as 0.2–2.5 μm , 2.5–10 μm and 10–100 μm . However, using a sieve to exclude particles larger than 100 μm is a fundamental prerequisite to consider particulate matter (which, per definition, comprises particles with diameter less than 100 μm). On the other hand, we found a huge problem linked to the naming of the different PM classes. For example, some authors named PM_{10} (which, per definition, comprises particles with diameter less than 10 μm) a fraction which actually includes particles with diameters ranging from 10 to 100 μm (e.g., Chen et al., 2016) or from 2.5 to 10 μm (Jin et al., 2021). This aspect can cause misunderstanding in the process of comparison among different experimental results. Without a clear and universal definition, comparisons among studies are easily subjected to

misunderstanding.

3.3. Scanning Electron Microscopy (SEM)

Scanning electron microscopy (SEM) generates images by scanning a sample surface with a focused beam of high-energy electrons. In this review, we found that particle quantification on vegetation surfaces was carried out using different types of scanning electron microscopes: Scanning Electron Microscope (SEM), Environmental Scanning Electron Microscope (ESEM) and Field Emission Scanning Electron Microscope (FESEM). Samples are observed in high vacuum in a conventional SEM, while in low vacuum or wet conditions in ESEM; FESEM has higher depth of field, with a definition reaching the atomic level. Scanning electron microscopy was used by several authors also to observe micro-morphological traits involved in PM trapping (e.g., Zhang et al., 2019) and to characterize the elemental composition of particles (e.g., Shi et al., 2017), providing a qualitative analysis, not considered in this review which focuses on quantitative analysis.

3.3.1. Experimental conditions

Most of the analyzed studies were conducted under real conditions, on urban vegetation in open field. Only one work was carried out under controlled conditions, in a chamber equipped with a simulated rainfall system. Two studies worked in open field, also simulating the rainfall effect, and thus they can be considered as a condition between real and controlled.

3.3.2. Sampling and preparation procedures

Sample material included only leaves. Most of the authors analyzed both sides (ad- and ab-axial) of the leaves, few authors (Ottel  et al., 2011; Perini et al., 2017) investigated only the ad-axial side and others did not specify this aspect. Authors which investigated needle-like species analyzed one sample surface, since it was not possible to distinguish two leaf blades, and then estimated as twice the PM density on the imaged surface to make it comparable with the other species, although this could underestimate total PM density on cylindrical leaf needles (Weerakkody et al., 2018b). The weather conditions during sampling, when specified, were mainly dry, and many authors performed the sample collection after a certain number of non-rainy days (here in the range 2–30 days). After the plant material was collected, the leaves were usually prepared to be investigated by cutting samples in surfaces mainly of 0.25 or 1 cm². The position of the cut was usually carried out on the leaf blade, mainly avoiding edges, midrib, tip and leaf base. Smaller leaves were usually analyzed without cutting, mounted intact on SEM (Song et al., 2015; Weerakkody et al., 2018b). Other preparation procedures included: drying, coating with gold (in an ion sputter coater) or carbon, to strengthen electrical conductivity and increase the quality of the images. To obtain high-resolution images, some authors used Back-scattered electrons (BSE), beam electrons that are reflected from the sample by elastic scattering.

3.3.3. Declination of the SEM method

The amount of particles accumulated on vegetation surfaces was determined by counting particles on micrograph images obtained by the above-mentioned types of SEM. Particle counting could be carried out by visual inspection (i.e. counting of particles on SEM images by human operators) or by automatic approaches (i.e. counting of particles on SEM images by image processing softwares). All the papers grouped in this review under the SEM method used an automatic approach; in two cases (Yan et al., 2016a, 2016b) approach was compared to the visual inspection to quantify accuracy and time requirements. Within the SEM method, we identified two declinations based on image processing software and techniques adopted for the automatic quantification: Image analysis tools were the most used (81%), followed by the object-based approach (19%). In all the declinations, the magnification employed to take the micrographs was in the range 100–5000 × .

3.3.3.1. *Image analysis tools.* Within this declination, the Image J software was the most used tool for automatic quantification of particles on vegetation surfaces by SEM method (67%), followed by Gwyddion v. 2.49 (9%) and Pathfinder 2.0 X-ray Microanalysis softwares (5%).

Ottel  et al. (2010) were pioneers in the development of the automatic approach for counting particles on leaf surfaces using Image J software. After taking micrographs (magnification mainly in the range 100–1000 ×) on randomly chosen spots with a reasonable particle distribution, binary images (i.e., black and white) were imported in Image J to be analyzed. The automatic threshold function was used to separate particles from the background; the manually set threshold was avoided to prevent a user-bias in the analysis. The watershed function was used to separate particles slightly overlapped (segmentation), even if this second step was not always mentioned by authors (e.g., Weerakkody et al., 2017, 2018a, 2018b, 2018c, 2019). After these two steps, particles could be analyzed to obtain information regarding particle size and number. When different magnifications were used to count different particle sizes (e.g., 100 × for particles >10 μm, 250 × for particles 2.5–10 μm and 500 × for particles <2.5 μm, Ottel  et al., 2010; Perini et al., 2017; Shi et al., 2017), weighing factors were used to compensate for the loss of counting area (zoom effect). No boundary to the circularity value was given: all various shapes of particles were counted. Cross sectional diameter of each particle was calculated, assuming that a calculated area belongs to a certain aerodynamic diameter.

Gwyddion v. 2.49 and Pathfinder 2.0 X-ray Microanalysis softwares were used instead of Image J by Ristorini et al. (2020) and Sgrigna et al. (2020), and Abhijith & Kumar (2020), respectively. They employed these softwares to quantify the PM number density on leaf surfaces using procedures like the ones adopted in Image J, separating particles from the background and counting particles of different fractions. Micrograph magnifications were 1800× for Gwyddion v. 2.49 and 500–1200 × for Pathfinder 2.0 X-ray Microanalysis.

3.3.3.2. *Object-based approach.* Yan et al. (2016b) developed an automatic method, named object-based approach, to quantify the particle load on the leaf surfaces. After importing micrographs (magnification in the range 500–5000 ×) into eCognition Developer™ software, three steps were performed. First, a multi-resolution segmentation (MRS) algorithm embedded in the software was used to segment SEM micrographs into image objects. The algorithm consecutively merged pixels or existing image objects into larger objects based on relative homogeneity within the merged object. Three key parameters (scale, shape and compactness) could be set to determine the degree of homogeneity. Second, a classification and Regression Tree (CART) was used to generate classification rulesets, including spectrum, shape and texture, based on selected training samples. Finally, the ruleset-based classification was applied to the entire micrographs for particle matter identification and characterization.

3.3.4. Parameters

The SEM method mainly provided the number of particles per unit leaf area (n particles cm⁻² or mm⁻² or μm⁻² of leaf area), followed by the number of particles per specific leaf area, i.e., a known surface area (n particles), covering both the 86% of the articles. Other parameters provided were the percentage of leaf area covered by particles (PLA, %) and mass of PM per unit leaf area, obtained through equations based on particle diameters and density (mg mm⁻² or cm⁻² leaf area or μg cm⁻² leaf area). The reduction of PM densities on leaves exposed to rain (%) and the deposition velocity (cm s⁻¹), the latter obtained by integrating aerosol monitor and SEM data (Abhijith & Kumar, 2020), were seldom delivered. The fractions of PM assessed ranged from ultrafine particles to total suspended particles.

3.3.5. Considerations

The SEM method enables the study of PM load directly on the leaves,

providing the particles amount, size and shape.

- *Particles diameter can be measured; soluble and insoluble surface PM can be detected; in-wax PM could be detected using BSE; a qualitative characterization (elemental composition and morphological traits) can be performed*

A wide range of particle dimensions, from ultrafine to large, could be detected, even if some authors reported that only particles larger than a specific size (e.g., 0.2 μm) could be counted for image resolution limitations (Sgrigna et al., 2020; Song et al., 2015). Regarding the type of PM, this method can identify not only the surface PM but also the in-wax PM, especially when using BSE as hypothesized by Sgrigna et al. (2020). Moreover, it can potentially identify both water insoluble and water-soluble particles, since this method does not require water washing preparation procedures, which determine the dissolution of the soluble fraction.

Although not treated in this review, it is worth mentioning that this method could provide a qualitative characterization of PM. Firstly, when coupled with Energy-dispersive X-ray spectroscopy (EDS), SEM could deliver data about the elemental composition of the particles, useful for source and toxicity identification, even if some authors pointed out the difficulties of this hybrid method in discriminating PM composition from leaf composition (Sillars-Powell et al., 2020). Secondly, SEM could be used to observe the micro-morphological traits involved in PM trapping, even if some authors combined it with 3D surface profiling to overcome the limitations linked to the 2D nature of this technique, not able to detect the depth and height of some traits (e.g., grooves) in which particles can be deposited (Redondo-Bermúdez et al., 2021).

- *On preparation procedures: a significant number of micrographs is required to accurately quantify PM deposition; magnification should be carefully set and its impact should be further explored*

Regarding the sample preparation for the analysis, it should be noted that since the scanning area is much smaller than the leaf surface area, a statistically significant number of micrographs is required to have a representative sample of the overall PM deposition (Abhijith & Kumar, 2020; Paull et al., 2020; Ristorini et al., 2020). The magnification of the micrographs is a key point to consider, since it may affect the recognition of the particles and make the comparability across studies based on different scales difficult. Lin et al. (2018) found differences among the tested magnifications, even if relatively small in some cases (e.g., between 1000 and 2000 \times) and a sensitivity to the scale effect higher in the ultrafine and fine particles than coarse and large ones. However, this point should be further explored.

- *On SEM method declinations: automatic approaches (image analysis tools; object-based approach) are more efficient than visual inspection method; automatic functions of the softwares should be preferred compared to manual ones; further investigations on the applicability of object-based approach should be performed*

Regarding the two declinations of the method identified in this review, image analysis tools (with Image J representing the most used) and object-based approach, they both automatically extract the particles captured on the leaves, showing higher efficiency and objectivity than the visual inspection method, which is too subjective and time consuming for common use (Yan et al., 2016a). Specifically, Yan et al. (2016a) found that their object-based approach achieved similar overall accuracy to that of microscopic inspection (92.17% versus 95.53%), but microscopic inspection took fourteen times longer. Yan et al. (2016b) reported that the major difference between Image J and object-based methods, instead, is that Image J requires the image to be a “binary” image, in which a threshold must firstly be set to distinguish the particles from the background. Therefore, the automatic threshold function

should be preferred to the manually set threshold, despite the potential higher accuracy, in order to preserve the precision and allow the comparability of different studies based on the Image J method (Ottelé et al., 2010). Equally, the watershed function should be used to separate particles slightly overlapped. Yan et al. (2016b) reported that the Image J method was less applicable with increasing complexity of the leaf surface structure, and that the particle boundaries were less reliable than that derived from object-based classifications. However, they did not perform a direct comparison with Image J method in their study and tested only the leaves of one species with a relatively complex structure, *Broussonetia papyrifera*, and only one magnification (1000 \times). However, few subsequent works applied this method using different magnifications or different species (Lin et al., 2017, 2018; Yan et al., 2016a), by configuring this novel approach as promising, despite further evaluations should be carried out.

3.4. Aerosol monitor (AM)

We grouped under the aerosol monitor method all the works which quantify the vegetation effectiveness in particle air pollution removal by measuring air PM concentration around vegetation in real conditions.

3.4.1. Sampling

The air sampling was carried out near vegetated systems, mainly roadside vegetation, followed by vegetation in urban green areas, urban parks and green walls. The scale of the sampling area varied among the studies, ranging, for example, from green panels of 12 m² (Donateo et al., 2021), to vegetated road segments 150 m long (Vailshery et al., 2013) to urban park plots of 0.4–3 ha (Gao et al., 2020). Since the scale was usually larger than in other methods, no work was carried out at the individual plant scale, rather they examined mono- (rarely) or pluri-specific groups of plants. Moreover, some authors listed only dominant species or mentioned only green coverage or vegetation patterns without reporting any information about the species. The air sampling mainly occurred on working days during daylight and rush hours in good weather conditions, also if some authors also measured during night hours (e.g., Srbinovska et al., 2021) and in all weather conditions (e.g., Yang and Chen, 2021). Instruments used varied across the works (the most cited were particle counters, aerosol spectrometers and dust mate machines), their distance from vegetation ranged from 0.5 to 150 m and the measuring height was in the range 0.7–19 m (1.5 m, the breathing height, was the most used).

3.4.2. Declination of aerosol monitor

Based on the analytical procedure carried out to measure the vegetation influence on air PM concentration, we identified two declinations of aerosol monitor method: direct (87%) and indirect (13%). Works employing the direct procedure used instruments which sampled the air in the field, providing air PM concentration values directly. The indirect procedure requires an additional laboratory phase for the quantification of the air PM concentration values.

Most of the studies which performed the direct procedure carried out stationary measurements (85%), using devices mounted on supports (e.g., tripods or stopped vehicles with engine shut off to avoid sample contamination; Abhijith & Kumar, 2020; Lin et al., 2016) fixed in specific points. Only few works (15%; Gómez-Moreno et al., 2019; Hagler et al., 2012; Lin et al., 2016; Ranasinghe et al., 2019) carried out mobile measurements (e.g., operator walking with devices or instrumented vehicles moving) in addition to stationary measurements. Monitoring frequency varied from a few seconds (1–6 s), approximating a human resting inhalation rate (Chang et al., 2014), to some minutes (1–10 min). Within a sampling day, different monitoring sessions could be carried out (each ranged from 2 min to 6 h) for a total monitoring of 1.5–24 h.

In the few works which performed the indirect procedure (Mori et al., 2018; Ozdemir, 2019; Tiwary et al., 2008; Vailshery et al., 2013), the filters derived from aerosol monitor devices working in the field

were later analyzed in the laboratory with combined gravimetric methods: weight difference without any cleaning procedure (Ozdemir, 2019; Tiwary et al., 2008; Vailshery et al., 2013) or vacuum filtration (Mori et al., 2018). In the case of weight difference, air PM concentration was determined gravimetrically by subtracting the initial mass of the filter sample (before exposure) from the final mass (after exposure) and then divided by the volume of the air passed through it. The devices used in this procedure worked at a flow rate ranging from 0.005 to 3 m³ min⁻¹. Filters, made of PTFE or glass microfiber paper, were prepared before and after the exposure mainly by drying and the exposure varied from 8 to 36 h. In the case of vacuum filtration, Mori et al. (2018) used passive samplers to collect air PM on PTFE membranes exposed to air pollution in the field for at least 10 days from the last rainfall. These membranes were later analyzed in the laboratory by water cleaning followed by the filtration of the solution obtained on different size porosity filters, to provide different PM fractions.

3.4.3. Parameters

The aerosol monitor method mainly provided the air PM concentration, expressed as a mass of PM per unit air volume (88%, µg m⁻³) and/or number of particles per unit air volume (31%, n cm⁻³). These parameters were in few cases normalized and presented as a number between 0 and 1. Some articles also reported the removal efficiency (expressed in percentage) or other similar parameters. Few studies estimated the inhaled contaminant dose (µg or µg min⁻¹) (Jia et al., 2021; Ozdemir, 2019), assessing the cyclist–pedestrian pollution exposure levels in presence or in absence of green infrastructures. One article (Mori et al., 2018) reported the mass of PM but expressed per unit filter membrane (µg cm⁻²). The only article which reported deposition velocity (cm s⁻¹) was Donateo et al. (2021), which calculated it by plotting the flux of PM (obtained by the gradient–flux relationship of Dyer, 1974) against the average PM concentration (recorded by the AM instrument). The fractions of PM assessed ranged from ultrafine particles to total suspended particles.

3.4.4. Considerations

The AM method investigates the vegetation effectiveness in PM removal by measuring PM not on vegetation organs but in the inhalable air in its surroundings, providing air PM concentration results.

- *On sampling: position, time period and weather conditions of the measurements should be carefully considered*

Regarding the sampling, which in this method is often difficult to distinguish from the measuring, it is worth noting that the relative position of the measuring devices to the vegetation should be carefully considered, since it can affect the measured PM concentrations in a non-negligible way (Srbínovska et al., 2021). Naturally, it should be set depending on the scale of the sampling area and on the weather conditions, especially wind direction. For example, Tiwary et al. (2008) investigated the influence of a hawthorn hedge on air PM concentration by placing measuring devices at two-thirds of its height (considered representative of the entire stand filtration properties) on its upwind and downwind sides (in a way that allowed a uniform and perpendicular flow of air through the hedge faces). Qiu et al. (2018), instead, studied the effect of green spaces on air PM concentration performing the measurements at a height of 1.5 m (the average height of human respiration) in ten urban sites with different vegetation structures, with a control group at the hard ground in each of them. Since it could be easy to over- or under-weigh some points, for example due to the presence of high-emitting vehicles near the sampling points or because of including data when the wind direction was not perpendicular to the system, some authors developed and reported specific approaches to handle these limitations (Ranasinghe et al., 2019). These approaches are particularly needed if the aim is to derive accurate concentration profiles from a series of concentration measurements collected on different days and

under slightly different conditions, as in the case of Ranasinghe et al. (2019).

Another consideration regards the time period for the analysis and its weather conditions. Most of the authors performed the measurements during daylight hours (generally capturing both morning and evening traffic peaks; Abhijith & Kumar, 2020; Tong et al., 2015), the period in which the most movement is expected from both people and vehicles, and in good weather conditions. However, it should be noted that measurements carried out on night hours and in all weather conditions (Srbínovska et al., 2021) should not be avoided, as they can provide a more representative framework.

- *Measuring devices powered by renewable energy and integrated with CIoT should be preferred*

The instruments used can limit the possibility of measuring during all the day and in all weather conditions. For example, power supply and poor reliability of Grim recorders in high humidity (such as during evening and morning mist and dew) restricted the measurements of Freer-Smith et al. (2005) between the hours 08.00 and 20.00. However, instruments powered by renewable energy could be used to face the power supply issue, and their integration with Cognitive Internet of Things (CIoT) would increase their effectiveness and lower their costs, other points which should not be ignored. This was underlined by Srbínovska et al. (2021), which in their study employed a new generation of sensor systems with relatively small dimensions and mobility nature (WSN, wireless sensor networks), reporting also the characteristics and costs of different low-cost PM sensors compared to traditional PM monitoring stations. To choose the instrument, some authors used devices whose field performance compared to other ones had been evaluated in previous work (Ranasinghe et al., 2019). Finally, other authors set the instruments with specific parameters (e.g., Tiwary et al., 2008, selected a moderate flow rate to minimize the volatilization of organic PM), calibrated them (Hagler et al., 2012) or implemented data quality control strategy (Abhijith & Kumar, 2019). Finally, it should be noted that AM method employs a suite of instruments (e.g., particle counters, aerosol spectrometers and dust mate machines) which compared to the monitoring stations (i.e., permanently located monitors, not treated in this review) can be placed in the most suitable point for the investigation, does not impact local dispersion patterns (especially when they are particularly small in size) and are less subjected to vandalism attacks (Tong et al., 2015).

- *On AM method declinations: the direct procedure should be preferred, especially if performed with mobile measurements carried out with electrical vehicles*

Regarding the two declinations of the method identified in this review, it is worth noting that the indirect one, since requiring an additional step (the laboratory phase) compared to the direct one (which can detect air PM concentration values directly in the field) is more subjected to possible bias. By contrast, it has the advantage of requiring more simple equipment, especially in case of passive samplers as in the work of Mori et al. (2018).

About the measurement type, Lin et al. (2016) found that both stationary and mobile measurements could reduce the UFP concentration downwind to a vegetation barrier, thus providing a good comprehension of temporal and spatial variability of particles in the air. However, they found that the two types of measurement agreed to within 20%. This little accordance can be explained by the ability of mobile measurements to detect the PM concentration also in proximity to gaps and spacing over the entire length of the vegetation, which reduce the filter effect of the system. Even if potentially more representative, mobile measurements show limitations linked to the using of vehicles, which could affect the data acquired, especially if non-electric. Otherwise, this does not occur when the measurement is carried out by an operator walking

with devices, although it can cover only a limited sampling area and in more time.

3.5. Wind tunnels and deposition chambers (WT&CH)

In this section we grouped all the articles which quantified PM removal by vegetation in specific indoor controlled conditions, i.e., wind tunnels (WT) and deposition chambers (CH). Most of the articles (60%) were performed in CH, while 37% was carried out in WT and 3% in both conditions. It is worth noting that we classified WT&CH as a method, even if it is more properly a condition in which different techniques were applied for the quantification of PM removal by plants. However, given the high number of studies based on this condition found in this research, we decided to consider it as a stand-alone method.

3.5.1. Facility and set-up

A wind tunnel is an indoor equipment used to study the flow of a fluid (typically air) around a body and it consists of a semi-enclosed or closed pipeline (Wu et al., 2019; Yin et al., 2019). In this field, it is commonly used to study PM deposition onto vegetation and dispersion in space due to its presence. This tool allows to control flow parameters (speed and orientation), exposing samples mainly to a unidirectional flow (Hwang et al., 2011; Lin et al., 2012). The length of the working section (i.e., the part of the wind tunnel in which the vegetation sample is located) observed in the selected articles varied from 0.5 to 4 m and the cross-sectional shape was variable (e.g., rectangular, hexagonal, circular, square) and varied in height and width (from 0.15 to 1.5 m for both).

A chamber is an indoor closed or semi-closed system (Yin et al., 2019) used for the same purpose of the wind tunnel, differing in height to length ratio which is higher in chambers. This feature allows an omni-directional flow around the sample (Hwang et al., 2011). The three dimensions of the chambers were mainly similar, ranging in the analyzed articles from 0.3 to 3.5 m, and the cross-sectional shape was mainly square, followed by circular, rectangular and octagonal.

The two devices were mainly made of acrylic, followed by stainless steel, glass and stainless steel, Perspex®, plastic steel and toughened glass and FEP Teflon. WT and CH require a tracer as a proxy of PM in urban environment, which consisted in different materials: the most used was NaCl aerosol, followed by mosquito repellent incense smoke, incense smoke, cigarette smoke, engine exhaust gas, mixture of surface soil layer, commercial powder, silica micro powder (SiO₂), soot and tire wear particles, oil mist particles (DEHS (di-(2-ethyl-hexyl) sebacate)), smoke from diesel absorbed on paper, talcum powder, (NH₄)₂SO₄ aerosol and outdoor ambient air. In WT vegetation samples were exposed to the pollution source for an exposure time ranging from 5 s to 40 min at a wind speed ranging from 0.7 to 10 m s⁻¹, while in CH the exposure time was higher, ranging from 8 min to 21 days and the wind speed, when specified, was lower, ranging from 0.9 to 2 m s⁻¹. In CH was also specified the flow rate, mainly ranging from 2 to 63 L min⁻¹ (even if in Irga et al., 2017, flow rates reached 900 L min⁻¹), and the initial air PM concentration, ranging from 36 to 2200 µg m⁻³ or from 300 to 4100 particles cm⁻³ of PM_x.

3.5.2. Vegetation sample and preparation procedures

WT and CH allow to work with different vegetation samples, from entire potted plants (in some cases with shoots held in place externally with a clamp and stand or in other cases fixed in green walls modules) to several plant organs, like branches and twigs (fixed into floral foam bricks or arranged horizontally or vertically on supports like slim frames) and leaves (inserted into mesh supports or floral foam bricks, tied on hangers or fixed with clips on supports).

Before exposure to the PM flux, some authors cleaned (washing, brushing, or ultrasonic cleaning) and dried samples (sometimes in a laminar flow cabinet, which allows samples to dry quickly within an uncontaminated environment, Chiam et al., 2019), while other authors

sampled after rainfall to have a surface free from pre-existing particles, while others acclimated plants in greenhouses or did not mention any preparation procedure.

3.5.3. Declination of WT&CH method and parameters

Three different declinations were identified in the WT&CH method depending on the detection method used to quantify the surrogate PM. The most found declination was air tracer concentration (70%), followed by mass of the tracer (27%) and tracer particles counting (13%). It is worth noting that some papers reported more than one declination. Since the WT&CH method is defined by a specific condition, it always requires a combined method to quantify the vegetation effectiveness in air particulate pollution mitigation. Even if obtained in different ways depending on the declination, several parameters were provided. Removal efficiency (%) and deposition velocity (cm s⁻¹, m s⁻¹) were the most found, followed by mass of PM mainly expressed per unit leaf area (µg cm⁻² leaf area, µg m⁻² leaf area, mg cm⁻² leaf area, mg m⁻² leaf area, mg tree⁻¹, mg g⁻¹ leaf d. w., mg l⁻¹ wash off cm⁻² leaf area), number of particles per unit leaf area (n mm⁻² leaf area, n cm⁻² leaf area, n m⁻² leaf area), particle number concentration (n cm⁻³ air) or particle mass concentration (µg m⁻³ air) and resuspension rate (%). The particles investigated were mainly ultrafine, fine and coarse, ranging from 9.8 nm–10 µm.

3.5.3.1. Air tracer concentration. The 'air tracer concentration' declination measured particle concentration in the air (mass or number), employing the same instruments used in the AM method but in controlled conditions (e.g., aerosol spectrometers, particle counters and laser nephelometers). For this reason, we reported AM method as a combined method of this declination of WT&CH method.

Removal efficiency (Re, %), a parameter used to express the ability of plants in reducing air PM concentration, was calculated in this declination by comparing air PM concentration upwind and downwind to the vegetation samples, considering in some cases also the amount of PM retained by the device walls. The basic equation used to calculate this parameter was (Equation (1)):

$$Re = (C_{up} - C_{down}) / C_{up} \times 100 \quad \text{Eq. 1}$$

Where Re is the removal efficiency (%), C_{up} and C_{down} are the air PM concentration upstream and downstream of the vegetation sample (WT) or of the entire device (CH), respectively. In three recent papers on green walls studied in CH (Irga et al., 2017; Morgan et al., 2022; Paull et al., 2019), this parameter was calculated by comparing air PM concentration in CH with and without vegetation (SPRE, single pass removal efficiency). Dry deposition velocity (V_d) is a key parameter to directly measure the particle retention capacity of vegetation and it varies depending on surface roughness and surrounding environmental conditions. V_d is widely used in predictive models to evaluate plants effectiveness in air quality amelioration (Yin et al., 2019; Zhang et al., 2021a, 2021b). In the declination 'air tracer concentration', we found that V_d was obtained by one main equation (Equation (2)):

$$V_d = F / C \quad \text{Eq. 2}$$

Where V_d is the dry deposition velocity of PM to the plant surface (m hr⁻¹ or cm s⁻¹), F is the deposition flux insisting on the sample (µg m⁻² hr⁻¹) and C is the air PM concentration in the WT or CH environment (µg m⁻³). Based on this equation, many authors developed different optimized formulas to take into account the quantity of particles retained on the WT or CH walls, by considering the collection efficiency of empty WT or CH. Three recent papers reported an indirect method to determine V_d in CH (Yin et al., 2019; Zhang et al., 2021a, 2021b). This method used an exponential attenuation model to measure the decline in tracer concentration in a closed environment with or without plants inside (Yin et al., 2019). This model is based on volume of the chamber, leaf area of sample plants, attenuation rate constant of control curve

(empty chamber) and of test curve (chamber with plants) expressed as unit time (s^{-1}).

Other parameters provided by the air tracer concentration declination, although less frequently, were particle number concentration, mass of PM concentration, mass of PM or number of particles per unit leaf area and resuspension rate. It is worth noting that in one case (Yin et al., 2020), the air PM concentration (mass of surface PM and number of particles) was measured with an aerosol spectrometer in the chamber immediately after resuspension from the brushed leaves and plotted to the leaf surface; this procedure was named by the author 'sweep-resuspension'.

3.5.3.2. Mass of the tracer. In the declination 'mass of the tracer', the mass of the surrogate PM was detected on washing solution or mineralized leaves. Based on tracer nature different combined methods were found. When NaCl was used as a source of PM, the total mass of its water-soluble ions, Na^+ or Cl^- , could be detected. In the reviewed papers, Na^+ was detected with atomic absorption spectrophotometer on washing solution (Beckett et al., 2000a; Freer-Smith et al., 2004) or mineralized leaves (Blanusa et al., 2015), or electrical conductivity on washing solution (Zhang et al., 2021b), while Cl^- with ion chromatography on washing solution (Liang et al., 2016). Ion chromatography on washing solution was also used to detect the water-soluble ions NH_4^+ and SO_4^{2-} of the ammonium sulfate tracer (Chen et al., 2017). Finally, to quantify the mass incense smoke and engine exhaust gas, the gravimetric method (VF) on washing solution previously described was used as the combined method.

In this declination, parameters were expressed as mass of PM (mainly per unit leaf area). Vd was calculated in two cases (Chiam et al., 2019; Zhang et al., 2021b) according to Eq. (2), where F was obtained by gravimetric method (VF) or by electrical conductivity of Na^+ in washing solution, while C was measured by the aerosol monitor method, using an aerosol spectrometer. In other two cases (Beckett et al., 2000a; Freer-Smith et al., 2004), Vd and Re were calculated according to Equations (3) and (4):

$$Vd = N/XA \quad \text{Eq. 3}$$

$$Re = N/XUA \quad \text{Eq. 4}$$

Where N is the number of particles captured (derived from the mass), A is the total leaf area, X is the dose of particles in the air stream (calculated from the number of particles per m^3 of air multiplied by exposure duration) and U is the wind speed.

3.5.3.3. Tracer particles counting. In the declination 'tracer particle counting' the number of the surrogate particles was detected on washing solution or leaf surfaces. In this declination only insoluble PM surrogates were used. Laser particle counter on washing solution (Xie et al., 2018, 2019, 2022) or Digital image-analysis system (DIAS) connected to microscope on leaves (Blanusa et al., 2015) were used to detect SiO_2 and talcum respectively. In this declination, the returned parameter was the number of particles per unit leaf area ($n \text{ cm}^{-2}$ or mm^{-2} leaf area).

3.5.4. Considerations

The WT&CH method imparted and measured a surrogate PM in specific indoor controllable conditions containing vegetation samples, mainly providing removal efficiency and deposition velocity as parameters.

- *PM deposition and dispersion on vegetation can be clearly analyzed in the ideal environment of WT&CH method, which however does not reflect the complexity of the real environment*

Some authors stated that this method is a necessary and comparative method to study the influencing factors and mechanisms of PM deposition and dispersion on vegetation (Shen et al., 2022). On one hand, this

has the advantage of providing an ideal environment in which variables like wind speed and particle concentration are controllable. Moreover, the real vegetation organs can retain their biological properties in these experiments, rather than just being treated as a porous medium model in numerical simulation works, as highlighted by Shen et al. (2022). On the other hand, deposition and dispersion are much more complex in the real environment, since these processes are influenced by many factors, difficult to simulate, such as meteorological and pollution dynamics, landscape geometry, etc. (Shen et al., 2022; Zhang et al., 2021b).

- *On facility and set-up: WT is more suitable to study vegetation in a street canyon, CH for plants free-standing in open spaces; CH is cheaper than WT; in both, PM adsorption on the devices inner walls should be considered for an accurate quantification. Setting up higher flows than those in real environment should highlight the differences in PM capture among species.*

There is a substantial difference between WT and CH facilities. In WT, vegetation samples are exposed to a unidirectional flow, which might represent the canalized wind direction in a street canyon, while in CH samples are exposed to an omni-directional flow, thus representing plants free-standing in a more or less open space (Hwang et al., 2011; Yin et al., 2019). Thus, the choice between WT and CH should be driven by the type of the green infrastructure which the study would like to analyze. The choice might be also guided by costs and other factors. A WT is much more costly to purchase and maintain compared to a CH (Zhang et al., 2021b) and usually need larger space (Yin et al., 2019). Moreover, it requires calibrations with high precision and accuracy (to allow low turbulences and uniform velocity profiles in the test section) and a separate control room to command environmental conditions (since it usually does not contain instruments to control temperature, humidity and other variables) (Zhang et al., 2021b). CH, instead, are usually smaller than WT and cheaper to maintain. Temperature- and humidity-controlling instruments can also be installed inside them. Some limitations linked to both WT and CH facilities included the surrogate PM adsorption on the inner surface of the devices, which could affect the PM removal potential estimate. Authors used different ways to face this issue. Someone used glass made chambers, often completely sealed with stainless steel and adhesive, to minimize the electrostatic accumulation of particles on the equipment walls (Cao et al., 2019; Jeong et al., 2021; Ryu et al., 2019), someone else cleaned the inner device sides prior to the experiment, with dust-free papers and anti-static agents (Jang et al., 2021), pure nitrogen (Zhang et al., 2021b) or ethanol (Morgan et al., 2022). Some others developed optimized formulas, to estimate deposition velocity or other parameters, by considering the collection efficiency of empty WT or CH (e.g., Hwang et al., 2011; Shen et al., 2022).

Regarding the WT and CH set up, it is worth mentioning that some authors chose to impart environmental variables to mimic the natural environment, as in the case of Chiam et al. (2019), which selected a turbulent air flow, or of Zhou et al. (2020), which injected in the CH the engine exhaust gas to simulate the morning and evening rush hours. Other authors chose to impose low (e.g., Cho et al., 2021), intermediate (e.g., Blanusa et al., 2015) and/or high wind speed (e.g., Beckett et al., 2000a). Blanusa et al. (2015) pointed out that higher wind speeds correspond to higher particle inertia and thus to a more effective impaction. Thus, higher flows should highlight the differences in PM capture among species, which may not be much evident in calmer weather conditions. Similarly, some authors imposed surrogate PM concentrations higher than real ones, to ensure a sufficient and detectable deposition and to simulate the increasingly polluted conditions of urban areas (Chiam et al., 2019).

- *On the surrogate PM: even if suitable to maximize the differences in PM trapping among species, it does not reflect the real and complex urban PM.*

The surrogate PM should be carefully chosen depending on the aim of the study and on the selected detection method for quantification.

The surrogate PM was chosen depending on chemical stability, water solubility or insolubility and particle size, factors sometimes also strictly dependent from the detection method used. For example, the diamond powder was chosen as tracer for PM_{2.5} due to its high chemical stability (Yin et al., 2019), silica micro powder (SiO₂) was selected as PM_{2.5} and PM₁₀ tracer due to its high chemical stability and low solubility (Xie et al., 2018, 2022). Incense smoke was used due to its low hygroscopic growth, with particle diameters relatively consistent under varying levels of humidity (Chiam et al., 2019). This tracer includes both water soluble and insoluble particles and their relative presence is brand-specific, influenced by components such as color additives and fragrance oils (Chiam et al., 2019). Moreover, some authors underlined that, while the PM generated may not be similar in the chemical content or health risks associated with other pollutant sources, incense generally produce a wide variation in particle sizes (ranging from 0.1 to 10 µm), which make it comparable to traffic-derived PM (Chiam et al., 2019). Regarding the particle size, it is worth mentioning that some tracers were most suitable for the larger size of PM_{2.5} (e.g., diamond powder, which is mainly composed from 1.8 to 2.5 µm particles), while others for the smaller size of PM_{2.5} (e.g., NaCl, which ranged in particle diameter from 0.8 to 1.2 µm) (Yin et al., 2019) and that other ones were assumed as PM_{2.5} tracer even if only 80% of the particles were below 2.5 µm diameter (e.g., cigarette smoke) (Cao et al., 2019).

Another point to underline is that deposition dynamics are different depending on the tracer used and on its dimension. For example, although Chen et al. (2017) found clear differences among species using both, (NH₄)₂SO₄ and ambient air tracers, they found a negative effect of plants on PM concentration only when using ambient air as proxy. This result highlights that tracer material and aerodynamic effects occurring in the system, rather than plant filtering capacities, can influence the concentration variation of particles. Moreover, it should be considered that smaller particles are less frequently fixed on leaves, and the measured deposition was typically less than larger particles (Yin et al., 2019). Thus, if the aim of the work is to compare different species in their PM removal potential, larger particles should be used, to make species differences more evident. Deposition dynamics are also affected by the composition of the tracer, which is variable and more or less known for the different PM surrogates. For example, Arizona Dust commercial powder (A1 Ultrafine TestDust, Powder Technology Inc.) and incense smoke both contained SiO₂, Al₂O₃, and trace amounts of Fe₂O₃, Na₂O, CaO, MgO, TiO₂, and K₂O; in addition, incense smoke included many gaseous substances (such as CO, CO₂, NO₂, and SO₂, and volatile organic compounds) (Ryu et al., 2019).

Overall, it must be stated that an important limit of the WT&CH method is working with a proxy of PM. Whether it is water soluble (e.g., NaCl) or insoluble (e.g., SiO₂) or both (e.g., incense smoke), whether it is mono- (e.g., NaCl) or multi-components (e.g., incense smoke, commercial powder, exhaust gas), a PM surrogate is extremely simplified compared to the real, complex, multi-dimensional and multi-source PM of the urban environment. For this reason, outdoor ambient air (Chen et al., 2017), engine exhaust gas (Wang et al., 2019; Zhou et al., 2020) or even soot and tire wear particles (Cho et al., 2021) appear to us more suitable PM surrogates to study the vegetation effectiveness in PM removal in urban sites, as more similar to the real ones. However, it should be noted that tracers of known and well-defined composition, could maximize the potential differences among species, also allowing a better understanding of the functional traits involved in PM trapping.

- *On vegetation sample and preparation procedures: using entire plants, with a surface free from pre-existing particles, should be preferable.*

Some authors underlined the effect of vegetation sample on the measured parameters. Specifically, Zhang et al. (2021b) attributed the

Vd values found, smaller compared to the ones of relevant studies using branches or seedlings, to the employment of leaves and shoots, which do not reproduce well the complexity of the spatial distribution of the leaves, reducing the micro turbulence and thus the deposition. For this reason, from our point of view, using entire plants as samples should be preferable. Even an accurate sample preparation is important, to ensure a surface free from pre-existing particles and well study the deposition dynamics. In our opinion, sampling after rainfall or wash cleaning should be more recommendable than a dry brush cleaning, which could create damages to morphological traits involved in the PM trapping.

- *On WT&CH declinations: 'air tracer concentration' (especially the indirect sub-declination) in CH seems a less laborious and cheaper way to determine deposition velocity compared to 'mass of the tracer' or 'tracer particle counting' in WT*

Regarding the method declinations, the most used was the 'air tracer concentration', especially in CH, in which this method was prevalent (almost all the articles). An advantage of this declination is that it is applicable to all the tracer types, regardless of their water soluble or insoluble nature. Within the 'air tracer concentration', we identified that authors measured the changes in the tracer concentration based on space (measuring the tracer concentration upstream and downstream to the vegetation in WT or of the entire device in CH or in empty or filled with vegetation CH) and time (in the approach used in the indirect sub-declination in CH). Particularly, this indirect method is based on an exponential attenuation pattern of the tracer concentration when the chamber is empty (control curve) and when it includes plants (test curve), which rely on the relative attenuation rate constants expressed as unit time (s⁻¹). It was developed by Yin et al. (2019) and compared to the 'mass of the tracer' used in WT by Zhang et al. (2021b). They suggested this method applied in CH as a less expensive and easier way to determine deposition velocity (Vd) compared to the methods applied in WT. This not only for the high cost of the WT equipment but also for the demanding multi-step procedures required by some traditional methods applied in WT well described by Zhang et al. (2021b). For example, compared with 'mass of the tracer' applied in WT, in which the determinations of Na⁺ and the electrical conductivity (EC) both required to wash off the NaCl tracer and dissolve it in deionized water, the indirect method applied in CH could obtain the data without extra treatments. This reduces the possibility of accumulating experimental errors, which could result in uncertainty, leading ultimately to inaccurate Vd values. Moreover, in the 'mass of the tracer' in WT, once EC was measured, it must be converted in deposition flux, and this requires paying a lot of attention to strictly control the temperature, which can affect the solubility of NaCl and thus the EC and the deposition flux. Once again, in the equation for calculating Vd, the 'mass of the tracer' declination in WT requires the tracer concentration, while this is not valid for the indirect method in CH, thereby simplifying the calculation. Overall, since Zhang et al. (2021b) did not find significant differences between Vd values from the wind tunnel and indirect methods using NaCl as tracer, the indirect method in CH should be taken into consideration, since it can augment the choices for researchers to quantify the PM capture ability of different species in an easier and cheaper way compared to the 'mass of the tracer' in WT.

The 'mass of the tracer' declination, as anticipated, seems to us laborious, since it requires different steps, such as wash cleaning or leaves mineralization, before measuring the mass of the tracer or its indicators, such as EC; in addition, for calculating Vd it also requires other measurements and their integration in formulas. Also, 'tracer particle counting' declination, which was found only in WT, resulted laborious for us, generally for the same reasons.

- *On parameters: WT&CH method is an important way to determine deposition velocity (Vd), a parameter widely used in predictive models*

Regarding the parameters provided by the WT&CH method, removal efficiency (Re, %) and deposition velocity (Vd, cm s^{-1} , m s^{-1}) were the most found and there is a substantial difference between them. The first indicates the relative effectiveness of different species in capturing particles, while the second indicates the absolute conductance of the vegetation organ (Beckett et al., 2000a). Vd is a key parameter for quantifying the ability of a plant surface to intercept particles and it is widely used in predictive models. Since it is difficult to measure accurately due to its small size and due to the variety of influencing factors (Shen et al., 2022) in the field, the WT&CH method appears to be an important tool to determine it, due to the controlled conditions offered.

3.6. Saturation Isothermal Remanent Magnetization (SIRM)

Saturation Isothermal Remanent Magnetization (SIRM) is the magnetization retained by a sample after a short exposure to a large magnetic field, e.g., 0.3 or 1 T (Bertold et al., 2019; Hofman et al., 2013). This technique measures the ferromagnetic and magnetizable component of PM (especially rich in traffic derived PM; Hofman et al., 2014b) accumulated on plant surfaces. The parameter provided is used as a proxy of air PM concentration in biomonitoring studies but also as a means to evaluate vegetation effectiveness in PM removal (Bertold et al., 2019; Kardel et al., 2011; Muhammad et al., 2022). We selected for this review any paper which used this technique to quantify vegetation effectiveness in PM removal or which reported data related to this aspect while having biomonitoring as its main purpose.

We identified three types of remanent magnetization: i) Anhyseretic Remanent Magnetization (ARM), which is produced by applying a slowly varying, weak magnetic field, ii) Isothermal Remanent Magnetization (IRM), which is produced isothermally by applying a strong magnetic field and iii) Saturation Isothermal Remanent Magnetization (SIRM), which is the maximum IRM which can be produced, i.e., in response to a saturating magnetic field. The latter was the main found and was provided as a parameter in all the analyzed papers. It is worth noting that some other authors (e.g., Kardel et al., 2011) referred to SIRM even if the magnetization occurred at a magnetic field below 1 T, the conventionally used, reporting that IRM measured at 0.3 T and SIRM measured at 1 T revealed no significant difference and a 99.9% correlation.

3.6.1. Experimental conditions

All the analyzed studies were conducted under real conditions, on urban vegetation in open field, except for one work (Wang et al., 2019), which was carried out under controlled conditions, in a wind tunnel. All the studies carried out in the real environment measured the outdoor air quality, except for one study, which evaluated the impact of vegetation (even if placed outside) on indoor air quality (Maher, 2013).

3.6.2. Sampling and preparation procedures

Sample material included mostly leaves, while in few cases the SIRM signal was quantified on branches (Wuyts et al., 2018), screen swabs (Maher, 2013) or on filters (Mitchell et al., 2010; Muhammad et al., 2022). The filters were obtained by aerosol monitor or gravimetric methods. Some authors specified the number of non-rainy days after which sampling took place (here in the range 1–11 days). After the sample collection, some preparation procedures were performed before the magnetic analysis: samples were usually tightly packed together by cling film, avoiding the movement of any sample parts, and pressed into 10 cm^3 containers; in some cases, after drying at 45–50 °C for 5–6 days.

3.6.3. Declination of SIRM method

We identified two declinations in the SIRM method: magnetization (78%) and demagnetization coupled with magnetization (22%). To measure the SIRM signal all the authors magnetized the sample with a magnetic field in the range 0.3–1 T using a magnetizer and then measured the magnetic intensity with a magnetometer, generally

calibrated routinely (after 8–10 measurements) against a laboratory rock specimen. Few authors (Mitchell et al., 2010; Wang et al., 2019) demagnetized the sample before the magnetization, measuring in sequence ARM, IRM and SIRM, respectively.

3.6.4. Parameters

All the papers provided as the main parameter the SIRM normalized by leaf area, except for one study which reported SIRM normalized by branch area (Wuyts et al., 2018); both were expressed in the range 10^{-7} A to μA . In addition, Maher (2013) reported the SIRM measured on paper swabs taken from the television or computer monitor screens placed in houses to assess the effect of installing trees outside on the indoor air quality. One work also provided the magnetic deposition velocity (MVd, cm s^{-1} ; Mitchell et al., 2010). In two cases (Hofman et al., 2014b; Muhammad et al., 2022) SIRM was provided for different types of PM: total surface accumulated (SIRMU) and immobilized (SIRMW) PM, measured on unwashed and water washed leaves, respectively. Specifically, by washing the leaves, the authors assumed that the resulting leaf SIRM was determined by particles immobilized (SIRMW), i.e., strongly retained on the leaf surface (e.g., on trichomes) or accumulated inside the leaf (trapped in the epicuticular wax layer or entered in the leaf tissues through the stomata). Without washing the leaves, instead, they assumed that the resulting leaf SIRM represented the biomagnetic signal of all leaf-deposited particles (SIRMU): leaf-immobilized and washable fraction, with the latter representing the water insoluble PM accumulated on the leaf surface removable by water washing procedures.

Fractions of PM were identified only when a combined method was used, i.e., when SIRM was quantified on filters obtained by aerosol monitor or gravimetric analysis (Mitchell et al., 2010; Muhammad et al., 2022), and covered the fractions from fine to total suspended particles.

3.6.5. Considerations

The SIRM method quantifies a proxy of PM accumulation on vegetation, since it measures its ferromagnetic and magnetizable components, providing a magnetic signal as parameter.

- Unless SIRM is widely used in biomonitoring studies, it can be also applied to quantify PM capture by different plant species

Although not addressed in this review, it is worth mentioning that the use of SIRM in the biomonitoring field (which includes studies using plants as bio-indicators of air pollution) is increasing. Indeed, compared to the low spatial resolution data of monitoring stations, SIRM measured on the urban plant networks can provide data at unprecedented high spatial resolution and at pedestrian-relevant heights (Mitchell et al., 2010). Some authors reported that it offered a sensitive, reliable, rapid, and relatively cheap method to estimate air PM (Bertold et al., 2019; Hofman et al., 2013), particularly the iron-rich, ultrafine, combustion-derived PM (Maher, 2013), without requiring any power source or protection from vandalism (Mitchell et al., 2010). Despite its use in this field, since the magnetic signal is species-specific (Kardel et al., 2011), it is also applied to estimate the PM accumulation capacity of different plant species.

- On sampling: using leaves is less time consuming compared to other plant organs

Although leaves were the most used sample, some authors investigated other vegetation organs to quantify vegetation effectiveness in PM removal, e.g., branches, but also highlighted that in this case the sample preparation required more handling, thus being more time-consuming and prone to errors compared to leaves (Wuyts et al., 2018).

- On SIRM declinations: magnetization appears less time consuming than coupling demagnetization with magnetization

Regarding the SIRM method declinations, coupling demagnetization with magnetization (2-step procedure) appears to us more time-consuming and complicated than only magnetization (1-step procedure) and authors employing this procedure did not mention particular advantages of this technique compared to the other.

- *On parameters: SIRM is only a proxy of PM (specifically, iron-rich PM), thus not representing always a good predictor of PM. It can not provide different PM size fractions, unless with a combined method*

Unlike PM, the SIRM signal is not prone to change chemical and physical properties, but since it is not defined by its size distribution, it does not allow to identify different fractions of PM without a combined method (Bertold et al., 2019). It should not be forgotten that the SIRM is a measurement for the total concentration of magnetic grains which linearly relates to the presence of Cu, Pb, Zn and Fe concentrations. Thus it is only a proxy for PM pollution and its associated toxic heavy metals, typically related to combustion, metallic wear and abrasion processes (Bertold et al., 2019; Hofman et al., 2014b). Therefore, in addition to being only a proxy of PM, it must be underlined that the SIRM signal is a good proxy only for a specific type of PM: the traffic derived PM (Hofman et al., 2014b). Consistently, Muhammad et al. (2022) found that plant species which showed a high mass of water insoluble removable PM on their leaf surfaces did not necessarily show a high SIRM of water-insoluble removable PM. This demonstrated that the SIRM signal was not always a good predictor of PM and that, besides particle mass, the composition of leaf surface accumulated particles differed between plant species. Moreover, they underlined the importance of investigating the water-soluble PM, which could be a not negligible part of the total leaf PM (in the range 7–50 %; Xu et al., 2019a), when using water washing procedures to discriminate between total surface accumulated and immobilized PM. Indeed, they found that the sum between the SIRM of the removable PM in three size fractions (SIRMVC, SIRMC, SIRMF, measured on filters obtained by the gravimetric method) and the SIRM of immobilized PM (SIRMW, measured on water washed leaves) was systematically lower than the SIRM of surface accumulated PM (SIRMU, measured on unwashed leaves), attributing the loss of 33 % in SIRM signal to the water-soluble fraction of PM. In addition, they pointed out that the tedious and time-consuming process of leaf washing could be avoided, since SIRMU is a good indicator of SIRMW for most (90%) of the investigated plant species.

3.7. Simulated rainfall experiments

Although only few papers analyzed in this review (7%) investigated the rainfall effect in the PM cycle on vegetation, the use of simulated rainfall systems is promising for the characterization of PM wash off by rain. These experiments were performed mainly in controlled conditions (80%, in laboratories or sealed glasshouses), while in few cases (20%) in real conditions.

The simulated rainfall was applied through manually spraying (20%; Przybysz et al., 2014, 2020) or by using more complex experimental devices (80%), which ranged in dimensions from 0.09 to 64 m². Specifically, the latter could be composed by a cistern when the water was stored from the rainfall (Xie et al., 2019; Xu et al., 2017), a pump which propelled water through the pipes, nozzles which sprayed the simulated rainfall, and boxes to collect the runoff (e.g., Zhang et al., 2019). Leaves were the samples most used (40%), followed by branches, shoots and twigs and in some cases a sample support was used (e.g., a metallic wire, a platform, a tripod and other holders). The water employed to simulate rainfall was not only rainwater (30% of the papers), but also distilled water (40%) and tap water (10%), while some authors did not specify this aspect. The simulated rainfall intensity ranged from 4 to 80 mm h⁻¹ and the time of exposure from 5 min to 6 h, for a total rainfall amount ranging from 0.1 to 60 mm.

Most of the papers used the gravimetric method (60%) to analyze the

runoff water. This was collected by dripping directly in containers or by previously passing through pipes or other containers. The runoff water was then filtered using VF and IVF methods previously described. The parameters obtained were the mass of surface PM per unit leaf area ($\mu\text{g cm}^{-2}$ or g m^{-2}) washed off or retained after a simulated rainfall event and the rate of PM washed off (%).

Thirty percent of the papers did not collect the runoff water but directly analyzed the leaves before and after the simulated rainfall event through the SEM method, specifically using the image analysis tools declination previously depicted. In this case, parameters provided were the number of particles retained on a specific leaf area (number of particles) of washed and unwashed leaves, from which the rate of PM washed off (%) could be calculated.

The WT&CH method was used only by one work (10%; Xie et al., 2019) to expose to surrogate PM the branches samples, which were then moved in a rainfall chamber to perform the simulated rainfall experiment. Authors did not collect the runoff water but sampled the leaves after the rainfall event, washed them (cleaning) and analyzed the resultant solution with a laser particle counter. In this way, the number of particles retained on the unit leaf area after a rainfall event was obtained (n cm^{-2} leaf area).

3.7.1. Considerations

Simulated rainfall is a tool to manage the rainfall characteristics in a controlled environment, providing the PM washed off and/or retained by leaves.

- *Mode of application: simulated rainfall duration and intensity should be carefully set, even if the complexity of natural rainfall cannot be achieved*

The mode of application of the simulated rainfall is an important factor because, besides the different species and the plant organ macro- and micro-structures, also the rainfall characteristics influence the PM wash-off and retention by the vegetation, especially duration and intensity (Xu et al., 2017). For example, Przybysz et al. (2014, 2020) manually sprayed distilled water. This type of application allows to consider the quantity of rainfall but not its duration or intensity, providing limited information compared to the more complex experimental devices in which these parameters can be easily managed (Zhang et al., 2019). Moreover, manually spraying may not provide sufficient intensity and kinetic energy compared to direct rainfall to remove particles from leaf surfaces (Weerakkody et al., 2018c). Nevertheless, it is worth noting that in natural rainfall the intensity of precipitation is not constant within a given period. Although in simulated rainfall trials the effects of a rainfall event on vegetation PM capture capacity were quantitatively assessed, the intensity and duration set for the simulation could not adequately represent natural rainfall. However, to obtain more accurate results, some authors carried out multiple pre-tests before the beginning of the formal simulated rainfall experiments, in order to calibrate the rainfall intensity and uniformity (Zhou et al., 2020).

- *Type of water: distilled water can clearly identify PM wash off dynamics, even if it does not consider the wet deposition which can occur with natural precipitation*

The type of water used for the simulated rainfall trials is another aspect that should be considered. Many authors employed distilled water, with the advantage of revealing the dynamic changes of PM washed off by plant organs (Cai et al., 2019). On the other hand, using the water from natural precipitation for the simulation experiments would also allow to consider the wet deposition coming from dirty raindrops, which may affect the PM deposition process. Indeed, the wet deposition on plant organ surfaces is a pathway of additional PM input: the particulates in the natural rainfall may drip onto the plant surfaces which can adsorb them (Zhou et al., 2020). Future studies could focus on

combining results from natural precipitations and simulated rainfall, as suggested also by Cai et al. (2019).

- *The method used in simulated rainfall experiments can affect the type of PM detected (water-insoluble or -soluble) and the parameter provided*

Considering the composition of PM particles retained by the plants, the water-soluble fraction is dissolved in rainwater (Beckett et al., 2000b) and removed from the plant organ surfaces earlier than the water-insoluble particles. Thus, it turns out that the washing effect of the rainfall could be strongly affected by the water-soluble fractional composition of PM (Xu et al., 2019a) and, consequently, by the sensitivity of the method used to detect this type of PM. For instance, Xu et al. (2017) and Weerakkody et al. (2018c) employed a similar rainfall simulation approach with different methods for PM quantification. The former analyzed the PM washed off by simulated rainfall using gravimetric method (VF), considering mainly the water-insoluble fraction of PM; the latter performed particle number concentration on leaves surfaces with SEM before and after the rainfall simulation trial, ensuring the inclusion of water-soluble fraction of PM in the analysis. In general, by considering the different methods applied in the simulated rainfall experiment we found that: i) gravimetric method (VF and IVF) was used to analyze the wash-off water providing the amount of PM washed off and retained by leaves; ii) WT&CH method analyzed the leaf retained PM using a laser particle counter on washing solution at different stages of simulated rainfall; iii) SEM method analyzed the leaf retained PM before and after the simulated rainfall, with also the possibility to derive the wash-off rate.

- *The laboratory environment is a simplification of the real conditions, thus not considering some important factors, such as canopy structure and meteorological conditions (e.g., wind)*

Some authors highlighted persistent differences between laboratory and field studies. These were mainly linked to the use of samples, which do not represent the effect of the whole plant, and to the difficulty in reproducing the dynamic natural conditions. Indeed, in the simulated rainfall experiments different plant organ samples were used (e.g., leaves, shoots, branches), mounted on different support structures to obtain the PM washed off from the leaves surface. This limits the trials to the small scales and makes difficult the upscaling of the relative findings to the entire tree or shrub (Xu et al., 2017). Indeed, it is difficult to replicate in the laboratory the effect exerted by the complex canopy structures to the rainfall rate and PM wash-off. This effect includes the limitation of the intensity of the rainfall in the inner parts of the crown with a consequent reduction of raindrop energy and its wash-off effect, and the reduction of the rainfall volume on the leaf surfaces in the lower crown (Xu et al., 2019a). Thus, simulated rainfall applied in experimental devices wash-off particles from sample leaf surfaces more easily than natural precipitation on entire plants in real conditions.

Laboratory experiments are independent from the effective weather conditions, thus they can be carried out and scheduled regardless of the natural precipitation occurrence. Moreover, simulated rainfall can be set up based on the specific aim of the studies, thus its characteristics such as quantity, intensity and duration can be managed to obtain comparable results. However, the technological limits offered by experimental environments should be considered, such as the inability of experimental devices to reproduce the sudden and continuous change of rainfall intensity that occurs in real conditions (Xu et al., 2017), or to replicate the effect of rainfall at low intensity (lower than 10 mm h^{-1} , as observed by Wang et al., 2015). It must be pointed out that the laboratory environment is an approximation of the real conditions, that in many cases does not consider some important external factors which could be involved in the cleaning effect exerted by the rainfall. Wind is one of these factors; it may affect the air PM concentration, even if it is still unknown whether strong winds strengthen or weaken the effect of

rainfall on PM removal (Xu et al., 2017). Therefore, to fill the gap existing between the natural meteorological conditions and the simulated conditions, an improvement of the experimental environments is necessary, together with further exploration of the synergistic effect between dynamic meteorological factors and PM wash-off by rainfall (Zhou et al., 2020).

4. Pros and cons, accuracy and reliability, and sustainability of the different methods and standard proposal possibility

The suitability of the methods for specific aims, their pros and cons, accuracy and reliability, and their environmental, economic and social sustainability are summarized in Table 2.

Very few studies have compared different direct methods to assess air PM removal by urban vegetation. Sgrigna et al. (2020) compared VF to SEM-EDS. Specifically, they compared the PM load obtained by VF (expressed in $\mu\text{g cm}^{-2}$) to the one obtained by SEM-EDS (in this case, the PM load expressed in $\mu\text{g cm}^{-2}$ was obtained by summing the element quantities, considered as an estimation of the amount of leaf deposited PM per unit leaf area). Sgrigna et al. (2020) discovered consistent results between the two metrics for six of the twelve tested species; for the remaining species findings were rather different (or even opposite). They attributed such discrepancies to the honeydew and wax presence. For example, the honeydew (detected by SEM images) found on the leaf surface of *Robinia pseudoacacia*, *Populus nigra* and *Tilia cordata* caused an overestimation of the PM mass measured by VF, since honeydew residuals were probably weighted together with washed PM. Furthermore, honeydew can have caused an underestimation of the PM measured by the SEM-EDS technique, since it could have hindered particles from being detected. On the other hand, Sgrigna et al. (2020) found that *Platanus × acerifolia* expressed a low PM capture when analyzed by VF, but a high PM capture when analyzed by SEM-EDS. A possible explanation, as reported by the authors, could be that the electron beam (5 and 15 KeV) used in the SEM-EDS was probably able to penetrate the wax layer and measure all PM, while VF not coupled with specific solvents (chloroform) cannot measure these particles. Overall, Sgrigna et al. (2020) found that other relevant literature results, within the same species, were more similar to their findings from SEM-EDS rather than from VF. Thus, they concluded that SEM-EDS appears to be a more appropriate method than gravimetric (VF) for PM load analysis on leaves, since it allows to better identify either PM dimensional classes and its chemical components, and to better evaluate the overall PM amount. These findings highlight that results of PM abatement may differ according to the analytical method used (Wróblewska and Jeong, 2021), until being even opposite.

In accordance, Song et al. (2015), which employed VF and SEM techniques on the same five species, found that fine PM (less than $2.5 \mu\text{m}$) only accounted for or 2% of the total mass PM, but that the number of fine PM was large and accounted for 96% of the total PM number on the leaf surface, respectively. This underlines that no direct relationship between particle mass and particle density exists (Muhammad et al., 2022). Similarly, Muhammad et al. (2022) found that, among the six trees and shrubs species investigated with both VF and SIRM methods, species which showed a high mass of water-insoluble removable PM from their leaf surfaces did not show a high SIRM value. This discrepancy of results emphasizes, according to the authors, that besides PM mass (detectable with VF method) also its composition differs among species and that SIRM method is able to detect these differences. Also, Maher (2013) used two independent approaches to quantify trees PM capture efficacy, AM and SIRM, but in this case they agreed, both indicating a reduction greater than 50% in measured indoor PM concentrations.

Considering not only the method and the provided parameters but also the removal mechanism, it should be considered that, although most of the authors focused their studies on PM deposition rather than on PM dispersion in the last thirty years, there is a need for a

Table 2

Suitability of the five methods analyzed in this review for specific aims, their pros and cons, accuracy and reliability and environmental, economic and social sustainability.

Method	Suitable to	Pros (+) and Cons (–)	Accuracy and reliability	Sustainability
Gravimetric (G)	i) study PM deposition ii) quantify the differences of PM retention among species iii) provide PM mass per unit sample area as parameter iv) detect particles mainly $>0.2 \mu\text{m}$ (from fine to large PM)	PROS: (+) divides PM into specific diameter classes (Yan et al., 2016b) (+) quantitative results (Xu et al., 2020) (+) possibility to analyze different plant organs as samples CONS: (–) static snapshot of a single moment (–) impossibility to study the PM load directly on the samples (cleaning procedures are required) (–) quantification of the surface PM; the in-wax PM is detectable using specific solvents (chloroform, in VF and IVF methods) (–) the chloroform, the solvent used to quantify the PM embedded in waxes, can potentially dissolve some of the particles containing non-polar molecules (Abhijith & Kumar, 2020) (–) quantification mainly of the water insoluble fraction of the adsorbed PM (underestimation) (Yan et al., 2016b; Yin et al., 2020). To quantify the water-soluble fraction, generally dissolved into the washing solution, combined techniques are needed. The same is valid for the elemental characterization (Ristorini et al., 2020) (–) retention of small particles after saturation of the filter membrane (Yin et al., 2020) (over- and under-estimation of large and fine PM respectively, VF and IVF methods) (–) method based on the geometric diameter (VF and IVF) instead of actual or aerodynamic diameter of particles (Esposito et al., 2020) (–) limited possibility of upscaling in space and time (since it requires multiple measurements in different time periods and environmental conditions)	ACCURACY: (+) weighing the particles after rinsing (VF and IVF method) is an approach frequently used in research on dust retention, which can provide a relatively accurate data (Paul et al., 2020) (–) however, the quantification of the mass of particles instead of the size and amount of particles can slightly approximate the real risk for human health with respect to heart and lung diseases (Ottel� et al., 2011), since actual particles diameters are not measured RELIABILITY: (+) time-efficient and cost-effective (Corada et al., 2021) (+) simple procedure (Yan et al., 2016b)	ENVIRONMENTAL: (–) use of a lot of water for wash cleaning and/or filtration procedure (–) use of a lot of consumables (e. g., filters, solvents) (–) use of chloroform poses environmental concern ECONOMIC: (+) equipment and consumables relatively cheap and easy to find (+) does not require sophisticated instrumentation (–) requires a lot of electricity (e. g., freezer for sampling storage, drying systems for filters stabilization, vacuum pumps functioning) SOCIAL: (+) does not require high skilled labor
Scanning Electron Microscopy (SEM)	i) study PM deposition ii) quantify the differences of PM retention among species iii) identify the micro-morphological traits involved in PM trapping (visual assessment of the active trapping sites) (Corada et al., 2021) iv) characterize the elemental composition of retained PM (an indicator of PM source), when coupled with Energy-dispersive x-ray spectroscopy (EDS) (Lin et al., 2017) v) provide particle number per unit leaf area as parameter vi) detect particles mainly $>0.1 \mu\text{m}$ (from fine to large PM)	PROS: (+) quantitative and qualitative results (+) study of PM load directly on the leaves (+) quantification of the surface PM; the in-wax PM could be detectable using BSE (Sgrigna et al., 2020) (+) quantification of both water-insoluble and water-soluble PM (+) automatic identification of the number of particles retained using automatic approaches (Yan et al., 2016a) (+) particles actual diameters identification (Yan et al., 2016b; Lin et al., 2017) (+) identification of a high number of diameter classes (higher compared to G) (+) particles shape information (Yan et al., 2016b), which other methods cannot provide (+) capacity for great magnification, large depth of field (Yan et al., 2016b) (+) possibility to discriminate the PM deposition on ab- and ad-axial leaf surfaces (Abhijith & Kumar, 2020) CONS: (–) static snapshot of a single	ACCURACY: (+) the method can visually measure the number, size, shape and distribution of the retained particles. Measuring the actual diameters of the particles considers the risk for human health with respect to heart and lung diseases (Ottel� et al., 2011) (–) however, the data have relatively low accuracy (Shao et al., 2019) if a statistically significant number of micrographs is not analyzed RELIABILITY: (–) high cost of the SEM (–) time-consuming (Wang and Shi, 2021; Ristorini et al., 2020) (–) SEM only acquires a very limited area, limiting the amount of reliable data (Wang and Shi, 2021) (–) since the scanning area is much smaller than the leaf (or other organs) surface area, a statistically significant number of micrographs is required to have a representative sample of the overall PM deposition (Paul et al., 2020; Abhijith & Kumar 2020; Ristorini et al., 2020)	ENVIRONMENTAL: (+) does not require the use of water or other solvents (–) can require gold or carbon coating for sample preparation ECONOMIC: (–) requires sophisticated and expensive instrumentation (–) requires a lot of electricity (e. g., freezer for sampling storage, SEM functioning) SOCIAL: (–) requires high skilled labor

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Table 2 (continued)

Method	Suitable to	Pros (+) and Cons (-)	Accuracy and reliability	Sustainability
		moment (-) difficult PM recognition (specifically when the leaf structure is complex or when particles are aggregated) (Wang and Shi, 2021) (-) small field of view (high results randomness) (Yin et al., 2020) (-) since the whole leaf is analyzed, it is difficult to discriminate with EDS whether the detected elements are due to leaf deposited particles or to plant uptake from soil (Ristorini et al., 2020) (-) analysis seems to be restricted only to leaf samples, excluding other plant organs (-) method based on the actual diameter instead of the aerodynamic diameter of PM (-) limited possibility of upscaling in space and time (since it requires multiple measurements in different time periods and environmental conditions)		
Method	Suitable to	Pros (+) and Cons (-)	Accuracy and reliability	Sustainability
Aerosol Monitor (AM)	i) study PM deposition in relation to dispersion ii) quantify the differences in PM removal among different planting configurations at large scale iii) provide air PM concentration parameter vi) detect particles also <0.1 μm (from ultra-fine to large PM)	PROS: (+) dynamic survey: long term non-stop monitoring possibility (+) measuring PM in the inhalable air (net of retention, wash off and resuspension), not on vegetation organs (+) quantification of the overall PM (although without distinguish between surface and in-wax PM) (+) quantification of both water insoluble and water-soluble PM (+) quantify also ultra-fine PM (+) possibility to discriminate PM fractions based on the aerodynamic diameter, depending on the instrument used CONS: (-) not suitable to quantify the differences of PM retention among species (-) limited possibility of upscaling in space and time (since it requires prolonged measurements in different environmental conditions)	ACCURACY: (-) data accuracy can be affected by environmental factors interference, even if specific approaches can be used to handle its effects (Ranasinghe et al., 2019) RELIABILITY: (+) large variety of air PM sensors (accuracy and costs): wide range of possibilities to face with different type of experiments and budgets (-) but, at the same time, possible difficulties in the choice of the more appropriate sensor (Srbinska et al., 2021) (-) instruments limitations (e.g., power supply, poor reliability in specific weather conditions, vandalism) (Freer-Smith et al., 2005; Tong et al., 2015)	ENVIRONMENTAL: (+) does not require the use of water or other solvents (-) some measurements require vehicles as instrument supports (-) some instruments require batteries and/or consumables (e.g., filters) (-) relatively short-lived tools, not easy to be disposed ECONOMIC: (+) large variety of air PM sensors, from the cheapest to the most expensive (+) does not require a lot of electricity (e.g., no freezer for sampling storage) SOCIAL: (+) does not require high skilled labor
Wind tunnels and deposition chambers (WT&CH)	i) study PM deposition, also in relation to dispersion (Shen et al., 2022) ii) quantify the differences of PM retention among species iii) study single influencing factors (e.g., plant traits, wind effect) and mechanisms of PM deposition and dispersion on vegetation iv) provide deposition velocity and removal efficiency as parameters v) detect particles also <0.1 μm (from ultra-fine to large PM)	PROS: (+) dynamic survey (+) controlling of environmental variables (e.g., wind velocity, air pollutant concentration, PM diameter) (Hwang et al., 2011; Lin et al., 2012) (+) simulating particle retention with a precision that is hard to replicate in the field (Yan et al., 2016b) (+) possibility to analyze the whole plant (even if potted) rather than a single plant organ, allowing to study the effect of the crown (+) possibility to quantify the overall PM (although without distinguish between surface and in-wax PM) (+) possibility to quantify both water insoluble and water-soluble PM (+) quantify also ultra-fine PM (+) possibility to discriminate PM fractions based on the aerodynamic diameter, depending on the instrument used (+) possibility of upscaling in space and time (since it provides Vd, a parameter widely used in predictive models and hardly to accurately	ACCURACY: (+) accuracy can be high due to the controlled condition offered (-) which, however, are not representative of the complex real environment, leading to uncertainties of the findings (Abhijith et al., 2017) RELIABILITY: (-) high cost (purchasing and maintaining the equipment), especially for WT (Zhang et al., 2021b)	ENVIRONMENTAL: (-) use of a lot of consumables (e.g., air PM surrogates) (-) use of some tracers (e.g., engine exhaust gas) poses environmental concern ECONOMIC: (-) high costs for WT (+) but less cost for CH (-) requires a lot of electricity (e.g., WT&CH functioning) SOCIAL: (-) requires high skilled labor

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Table 2 (continued)

Method	Suitable to	Pros (+) and Cons (–)	Accuracy and reliability	Sustainability
Saturation Isothermal Remanent Magnetization (SIRM)	i) study PM deposition ii) quantify the differences of PM retention among species (Bertold et al., 2019) iii) assess the air quality by using plants as indicators of air PM concentration (biomonitoring) (Mitchell et al., 2010; Kardel et al., 2011) iv) get information about the ferro-magnetic composition of PM (an indicator of PM source) v) provide magnetic signal as parameter	measure in the field, Shen et al., 2022) CONS: (–) simplified condition compared to the real one (Zhang et al., 2021b; Shen et al., 2022) (–) applying and measuring a proxy of PM PROS: (+) quantification of both water insoluble and water-soluble PM (Corada et al., 2021) (+) study of PM load directly on the leaves CONS: (–) static snapshot of a single moment (–) quantification of the surface PM; the in-wax PM is detectable applying water washing procedures (Muhammad et al., 2022) (–) since the whole leaf is analyzed, it is difficult to discriminate with SIRM whether the detected elements are due to leaf deposited particles or to plant uptake from soil (Ristorini et al., 2020) (–) impossibility to discriminate different PM fractions without a combined method (Bertold et al., 2019; Muhammad et al., 2022) (–) limited possibility of upscaling in space and time (since it requires multiple measurements in different time periods and environmental conditions)	ACCURACY: (–) low accuracy: SIRM does not measure PM but a proxy of PM (i.e., its ferro-magnetic component) (Bertold et al., 2019; Hofman et al., 2014a, 2014b) RELIABILITY: (–) expensive and time-consuming (Corada et al., 2021)	ENVIRONMENTAL: (–) electromagnetic pollution ECONOMIC: (–) high costs of equipment (–) requires a lot of electricity (e.g., functioning of instrumentation) SOCIAL: (–) requires high skilled labor

simultaneous evaluation of these two removal mechanisms, to figure out the relative contribution of each in the total air pollution removal, as pointed out by Abhijith & Kumar (2020). They coupled the evaluation of PM deposition by SEM to the PM dispersion by AM, deriving the deposition velocity for a roadside hedgerow of *Fagus sylvatica* and providing insights into its PM removal potential. At the same time, it is worth noting that a lot of studies were carried out on PM retention, while few on wash-off and PM resuspension (Xu et al., 2020), processes which should be deepened in future studies. Moreover, in their experiment carried out on roadside vegetation, Poppek et al. (2022) observed with VF analysis that the first row of trees captured, per unit leaf area, a significantly lower quantity of PM compared to herbaceous plants in the same location (probably due to the shorter distance of the herbaceous species from the road). Despite this, the high concentration of PM in the air between the road and the first row of trees, found with AM method, revealed that the amount of PM deposited on trees should be higher. The authors attributed these findings to the re-suspension of PM temporarily accumulated on trees by wind and rain, which the gravimetric approach is not able to consider. Mori et al. (2018) compared the air PM deposition on filters of passive samplers (AM indirect method), an indicator of air PM concentration in the experimental area, to the leaf PM deposition (obtained with VF). Similarly, authors found different dynamics between the two methods, but in this case they were probably attributable to the experimental settings (PTFE membranes were renewed at each sampling while leaves were continuously exposed), to the structure of the passive samplers (which were differently influenced by the action of climatic factors compared to the leaves) and to the experimental material (PTFE membranes and leaves have different macro- and micro-characteristics).

This review has shown that there are a wide range of metrics used to quantify the air PM reduction of urban plants and that, within the same metric, a high variability exists (e.g., in sample size, interval time, preparation procedures, method declination and combined techniques, sample area measurement, parameters expression). We realized that depending on the specific purpose there are methods more appropriate than others. For example, SEM is a good method to study the

species-specific morpho-functional traits involved in PM trapping, WT&CH to study specific influencing factors or processes, G for PM deposition, AM for PM dispersion. SIRM is more suitable for air PM monitoring rather than for the purpose of vegetation effectiveness in air quality amelioration. However, a standard universal procedure, non-existent to date in our knowledge, is needed to accurately quantify the effectiveness of urban vegetation in air PM mitigation and to make possible the comparison across studies. This scientific demand was also recently pointed out by other authors (Corada et al., 2021; Paull et al., 2020; Xu et al., 2020).

The standard procedure, from our point of view, should i) quantify the PM net removal, i.e., the amount of PM removed from the air by vegetation in a specific time-frame, net of PM wash-off and resuspension (Muhammad et al., 2022; Xu et al., 2020), ii) consider both deposition and dispersion mechanisms, iii) provide deposition velocity as parameter, and iv) represent the best compromise among accuracy, reliability and sustainability (environmental, social and economic). Despite most of the studies focus on PM deposition, more attention should be given to PM dispersion, which is more related to the actual concentration of pollutants which citizens can experience. Thus, for the above-mentioned points, we believe that coupling gravimetric and aerosol monitor methods (G and AM) in real conditions, could be a possible route for future studies on the developing of a standard procedure. These methods are the best compromise among accuracy, reliability and sustainability, according to the findings emerged in this review. They are also able to provide deposition velocity (Vd), a key parameter which not only considers both deposition and dispersion mechanisms, but also can be used to model vegetation effectiveness in air quality amelioration. For example, once Vd is known for a species, it can be modelled for the same species in different contexts, with different air PM concentrations. Providing parameters implementable in models can give an important applicative value to the scientific research, since models can be used for planning purposes. Potential challenges linked to the standardization using methods in real conditions are connected to the complexity of plant-air pollution interactions existing in the real environment. However, in our opinion, it is more appropriate to build a standard procedure

based on experiments performed in real conditions. Indeed controlled conditions represent an ideal and simplified environment not able to reflect the complexity of the real environment. However, performing further comparative evaluations among the existing metrics, too few to date, is required to identify the right direction. In this way it will be possible to identify the more suitable metric or metrics combination for the development of a commonly recognized procedure.

We put a lot of effort into extricating the big melting pot existing on this challenging research subject. Hopefully, this integrative review will guide all the interested researchers in the study of air PM removal by vegetation, providing them an organic and clear tool to choose the more suitable metric for their purpose, time and budget, being aware of the different advantages, drawbacks, accuracy and reliability of the different approaches. Finally, we believe that this work can represent a scientific and critique perspective for future studies, a first step towards the creation of a standardized approach for the quantification of the urban vegetation effectiveness in air PM removal.

CRedit authorship contribution statement

Irene Vigevani: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Denise Corsini:** Data curation, Formal analysis, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Sébastien Comin:** Validation. **Alessio Fini:** Conceptualization, Supervision, Writing – review & editing. **Francesco Ferrini:** Writing – review & editing.

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.

Data availability

Research data are available in the supplemental material.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.aeoaa.2023.100233>.

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