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Variation in Tree Species Ability to Capture and Retain Airborne Fine Particulate Matter ($PM_{2.5}$)

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Human health risks caused by $PM_{2.5}$ raise awareness to the role of trees as bio-filters of urban air pollution, but not all species are equally capable of filtering the air. The objectives of this current study were: (1) to determine the foliar traits for effective $PM_{2.5}$-capture and (2) explore species-to-species differences in foliar $PM_{2.5}$-recapture capacity following a rain event. The study concluded that overall, the acicular needle shape made conifers more efficient with $PM_{2.5}$ accumulation and post-rainfall recapture than broadleaved species. The foliar shape and venation of broadleaved species did not appear to influence the $PM_{2.5}$ accumulation. However, the number of the grooves and trichomes of broadleaved species were positively related to foliar $PM_{2.5}$ accumulation, suggesting that they could be used as indicators for the effectiveness of tree $PM_{2.5}$ capture. Furthermore, the amount of $PM_{2.5}$ removal by rainfall was determined by the total foliar $PM_{2.5}$. Not all $PM_{2.5}$ remained on the foliage. In some species, $PM_{2.5}$ was resuspended during the growing season, and thus reduced the net particular accumulation for that species. These findings contribute to a better understanding of tree species potential for reducing $PM_{2.5}$ in urban environments.

$PM_{2.5}$ has raised severe public health concerns as particles easily penetrate the pulmonary alveoli[1] and pollution issues related to it have become increasingly severe as a result of global climate change. Periodic $PM_{2.5}$ pollution episodes in cities are more likely to develop in winter in the Northern Hemisphere because of increased air temperature as well as more frequent atmospheric inversions under the background of global climate change[2]. Moreover, summer episodes may also increase $PM_{2.5}$ concentrations due to an increased in the likelihood of stationary air masses[3], intense secondary aerosol formation[4–6] and forest fires[7–9]. Cessation of vehicular or industrial $PM_{2.5}$ emissions is not economically or functionally practical in highly urban areas such as Beijing. Therefore, cities will have to develop multiple measures to mitigate $PM_{2.5}$ concentrations. Tree planting (a.k.a. "greening") has been suggested as one method to reduce $PM_{2.5}$ in urban areas because these measures would effectively complement air pollution mitigation[10]. Foliage acts as a bio-filter of air pollution[11] and improve air quality[12] due to the leaves' rough texture and large contact area. Vegetated greenbelts (i.e., areas of natural or planted herbaceous and non-herbaceous vegetation) can effectively reduce the dust and filter the suspended particles that would otherwise impact urban areas[13]. Several previous studies have evaluated the amount of $PM_{2.5}$ removal from urban air by vegetation. For example, concentrations of $PM_{2.5}$ have been shown to decrease by 9% in woodlands immediately adjacent to urban areas[14]. On a larger scale, trees annually removed approximately 300 metric tons of air pollutants from Christchurch, New Zealand[15]. In Beijing, the trees removed 1,261 metric tons of pollutants, 772 metric tons of which was PM_{10}[16]. Studies conducted in the UK indicated that planting trees on one-fourth of the available urban area can reduce PM_{10} concentrations by 2 to 10%[17]. Overall, the findings suggest that urban vegetation have a direct and positive effect on human health by reducing $PM_{2.5}$. Thus, tree planting can be considered pollution mitigation measure in a variety of urban settings. However, open space suitable for tree greening programs is limited in cities. Therefore, if urban vegetation is to be employed as a measure for controlling the air pollution, the most efficient species and tree spacing should be used to maximize $PM_{2.5}$ uptake by vegetation. The ability to identify the most efficient vegetation attributes and species for capturing $PM_{2.5}$ is important because that will provide a basis for selecting plants to improve air quality in vulnerable areas.

Unfortunately, current studies on foliar $PM_{2.5}$ capture efficiency are inconclusive. Significant differences have been reported between and within modelling and experimental studies. A number of such studies were

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conducted to evaluate deposition velocity (V_d, cm s^{-1})[18], deposition amount (mg cm^{-2} or μg cm^{-2})[19, 20], magnetic deposition velocity (cm^{-1})[21], particle number (mm^{-2})[22], and particle cover area (%)[23]. For example, PM_{10} deposition velocities (V_d) on vegetation varied from ~0.01 to ~10 cm s^{-1}[24], but models were not very accurate at predicting V_d within a species. Simulated V_d of 1 μm diameter particles on *Picea abies* were calculated to be 0.02 cm s^{-1}[25], but measured rates of V_d were 0.55 cm s^{-1}[26], a 25-fold difference. Differences between modeled and measured V_d can be attributed to uncertainties associated different physical and chemical processes involved in tree-atmosphere interactions such as $PM_{2.5}$ capture model sensitivity to different plant boundary layer parameters, and to the importance of initial conditions[27]. Measured particle V_d are not only dependent on the measurement methods[17] but also on a number of other factors such as particle size and density[28], concentrations of other pollutant (e.g., ozone)[29], meteorological conditions (e.g. precipitation affecting particle removal from the leaves[30] and wind[31] affecting resuspension and boundary layer heights[32]) and the tree canopy morphology[30]. Although foliar $PM_{2.5}$ deposition varies with *in-situ* conditions, the foliar $PM_{2.5}$ accumulation is often just considered to be a species-specific[33]. An examination foliar $PM_{2.5}$ retention ability for different species under similar conditions is necessary to better rank tree species efficiency for capturing $PM_{2.5}$.

Given that the surface properties of objects are known to influence particle immobilization[34], plant species differ in their ability to scavenge dust-laden air[24, 34]. The dust-retention abilities of vegetation depend on several factors including canopy type, leaf and branch density, and leaf micromorphology (e.g., roughness, trichomes and wax)[18, 19, 31, 33, 35–39]. Conifers are considered to be more effective in PM_{10} capture than broadleaved species[34] and evergreen conifers have the potential to accumulate pollutants throughout the year. Within the broadleaved species, rough leaf surfaces are more efficient in capturing $PM_{2.5}$ than those with smooth leaf surfaces[31, 40]. Within species cultivar, leaf surface property variation can also impact $PM_{2.5}$ capture. Large-scale sampling must be conducted to quantify the relationships between species traits and $PM_{2.5}$ capturing capacity.

Although it is understood that the temporary retention of particles by urban trees can reduce atmospheric $PM_{2.5}$ concentrations, the effectiveness of vegetation as a long-term alternative to other measures is still under debate[24]. Most particles are retained on the plant surface and subsequently removed from the canopies by resuspension to the atmosphere through rainfall and leaves fallen to the ground[30, 41]. Particle fate is also impacted by other factors, including canopy characteristics, micrometeorological conditions, particle size and leaf morphology[18, 31, 42, 43]. However, to date, no study has systematically compared interspecies effectiveness in capturing and retaining $PM_{2.5}$. It is not applicable to deduce the foliar $PM_{2.5}$ deposition and resuspension from PM_{10} studies due to the weak correlations between the PM size fractions[44].

Therefore, the aim of this study is to (1) quantify the relationship between $PM_{2.5}$ accumulation and the leaf macromorphology and micromorphology using a large sampling population and (2) explore the differences in $PM_{2.5}$ retention under rainfall conditions by leaves of different species. To our knowledge, this is the first study to quantify the relationship between leaf macromorphology and foliar $PM_{2.5}$ capture. The findings from this study have practical implications for urban tree species selection targeting air pollution. Moreover, the results presented here will assist urban planners to evaluate the potential capacity of $PM_{2.5}$ removal from the long term, large scale perspective.

Results

Foliar $PM_{2.5}$ accumulation of tree species. Atmospheric $PM_{2.5}$ captured on the leaves of urban trees varied among species and seasons (Fig. 1, see also Supplementary Table S1). Coniferous species predominately contained the largest accumulation (>20 μg cm^{-2}) of $PM_{2.5}$ were predominantly coniferous species (Fig. 1b). Conversely, the four most efficient broadleaved $PM_{2.5}$-retention species were *Catalpa specieosa*, *Ulmus pumila*, *Amygdalus triloba and Broussonetta papyrifera*, all characterized by leaves covered with dense hairs, whereas the least effective species were*Tilia tuan*, *Armeniaca sibirica and Lonicera maackii*, which had smooth leave surfaces.

Tree morphological traits and leaf retention of $PM_{2.5}$. Acicular (needle-shaped) leaves showed the highest capacity to capture $PM_{2.5}$ (one-way ANOVA, $P = 0.01$, Fig. 2a), followed by lanceolate leaves. No significant differences ($P = 0.09$) in the $PM_{2.5}$ amount per unit leaf area were observed between the different venation patterns of the studied species, although trinervious veins exhibited favourable $PM_{2.5}$ capturing effect (Fig. 2b).

Digitized morphological features were extracted from leaf images (Fig. 3). The groove ratio (groove area/total leaf area) ranged from ~3 to 25% across all species. Trichomes are the fine outgrowths (including various types of hairs) or appendages on plants. Among the examined species, 53% had no trichomes, (i.e., trichome density ($LH_{ave} = 0$). The remaining species had trichomes with varying morphologies and densities including sparsely distributed trichomes ($LH_{ave} = 5.6 \pm 1.13$ mm^{-2}) or densely covered trichomes ($LH_{ave} = 19.9 \pm 5.20$ mm^{-2}). The stoma densities ranged between 40 and 140 mm^{-2} and diameters ranging between 10 and 25 μm.

Across species, the highest rates of $PM_{2.5}$ capture were observed on foliage with micromorphological structures that included dense grooves (Fig. 3a–h) and epicuticular trichomes (Fig. 3q–t). *Armeniaca sibirica* (Fig. 3n) and *Phyllostachys propinqua* (Fig. 3i) both had pointy, protrusive structures. However, *A. sibirica* had grooves around the protrusion and was able to capture more $PM_{2.5}$ than *P. propinqua* that lacked these grooves. Protrusions that were flattened were not as efficient at retaining $PM_{2.5}$ (Fig. 3i–p) compared to protrusions that were pointy. However, the presence of stoma did not necessarily lead to a larger capacity for $PM_{2.5}$ capture (Fig. 3u–x).

A correlation analysis indicated that the amount of captured $PM_{2.5}$ trapped particulate matter was positively correlated with the total epicuticular trichomes ($R = 0.69$, Fig. 4a) and the groove density (Fig. 4b). No relationship was found between foliar $PM_{2.5}$ accumulation and the stomatal density or diameter.

Removal of $PM_{2.5}$ from leaf surfaces by simulated rainfall. $PM_{2.5}$ removal by rainfall was correlated with the amount of the pollutant retained on the leaf before the rainfall simulation (Fig. 5a). This is in consistent with the fact that rainfall scavenging being a first-order process, and dependent on leaf particle concentration[45].

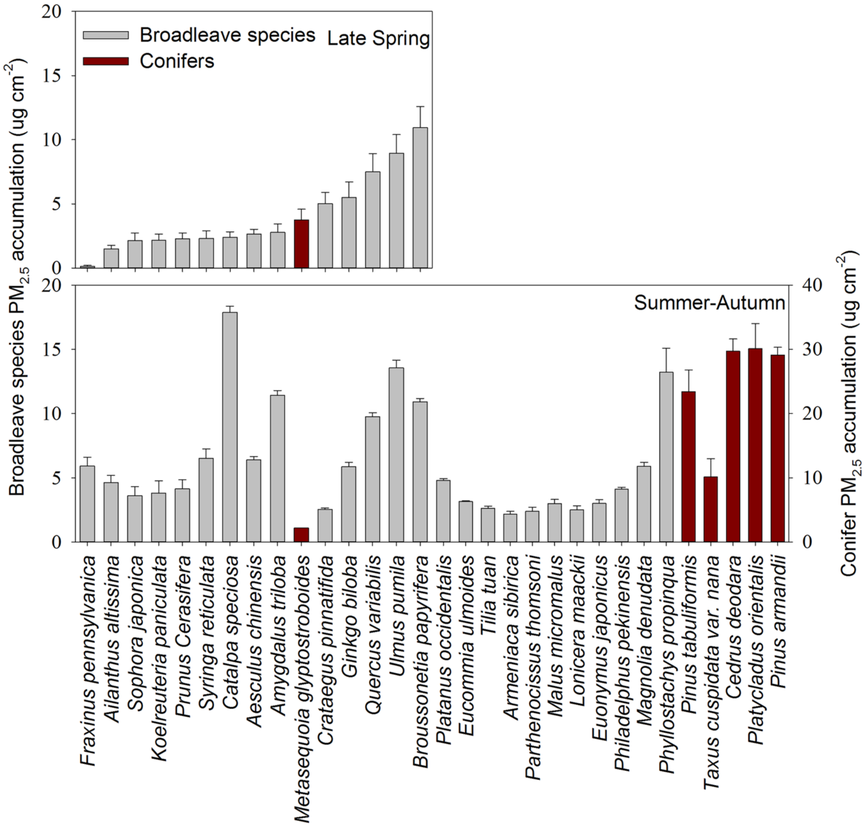

Figure 1. Comparison of foliar accumulation of atmospheric $PM_{2.5}$ among by different tree species (coniferous and broadleaved) measured in (**a**) late spring and (**b**) summer through autumn. The within-sample variability of $PM_{2.5}$ of each species presented as error bars.

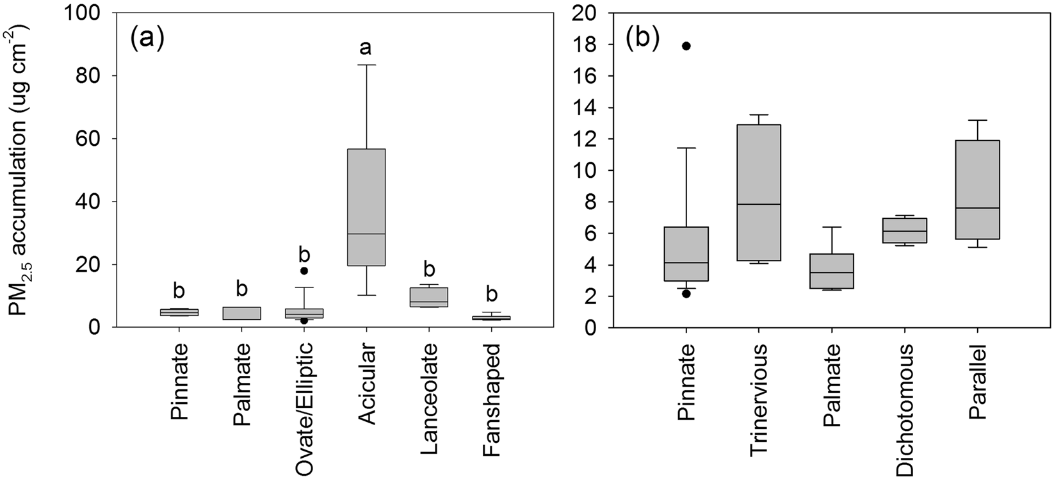

Figure 2. Differences in foliar $PM_{2.5}$ accumulation between different leaf shapes (**a**) and venation patterns (**b**) pooled from all species measured. Bars are means ± SE. Different letters above the error bars indicated significant differences between leaf shapes (Bonferroni test, $\alpha = 0.05$).

The $PM_{2.5}$ retention ability of different species varied significantly (one-way ANOVA, $P = 0.03$) (Fig. 5b). The average removal rate of the foliar $PM_{2.5}$ of the examined coniferous species was 60% (SE = 4%) while the removal rate for broadleaf species was 47% (SE = 3%). *Platyladus orientalis* and *Pinus armandii* lost up to 86% (SE = 5%) and 66% (SE = 4%) of their foliar $PM_{2.5}$, respectively. Only *Cedrus deodara* demonstrated a lower foliar $PM_{2.5}$ removal rate of 30%. Among the broadleaf species, *Eucommia ulmoides* and *Sophora japonica* demonstrated the highest average foliar $PM_{2.5}$ removal rate of 82% (SE = 4%) and 64% (SE = 4%), respectively. The removing

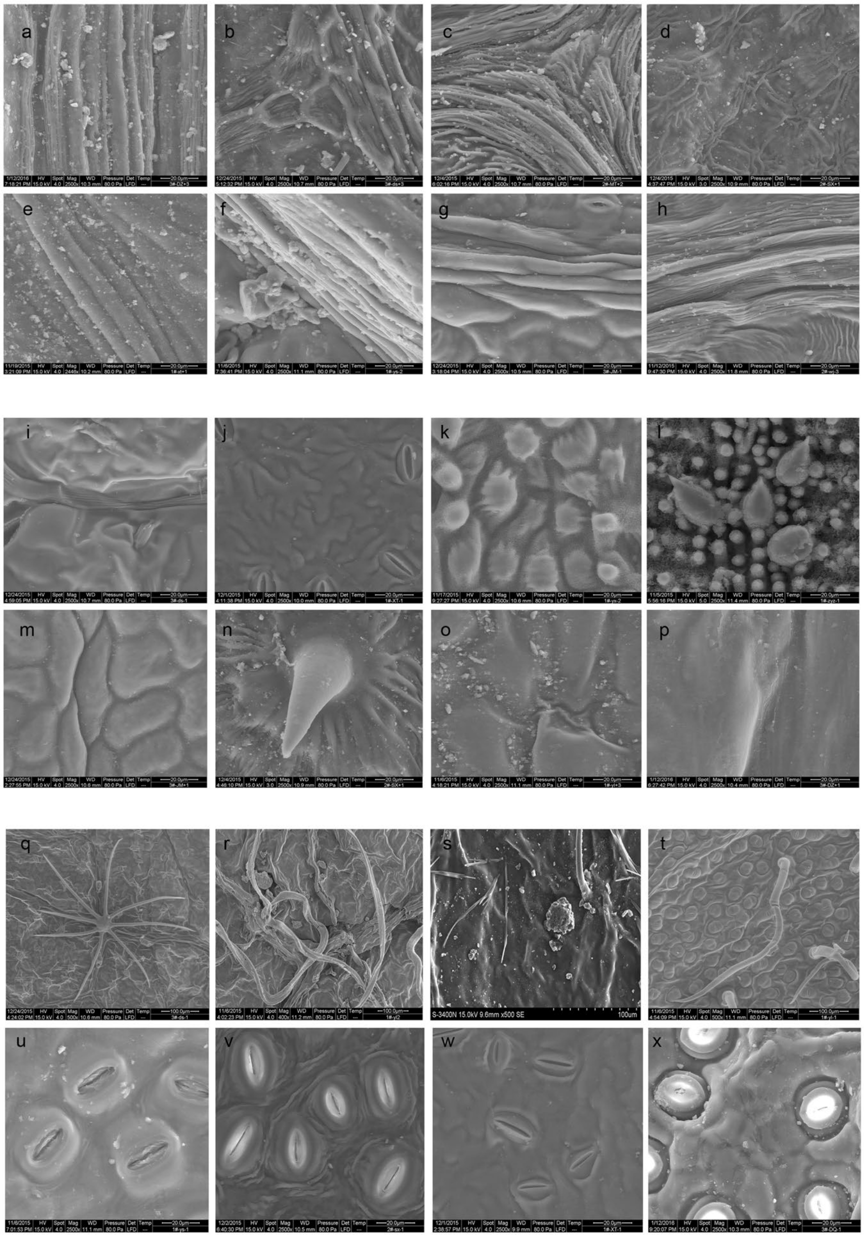

Figure 3. Images of leaf surface micromorphology and deposited particulate matters. The images corresponded to the samples of: (**a**) *Eucommia ulmoides*, (**b**) *Tilia tuan*, (**c**) *Platanus occidentalis*, (**d**) *Armeniaca sibirica*, (**e**) *Malus micromalus*, (**f**) *Ulmus pumila*, (**g**) *Lonicera maackii*, (**h**) *Parthenocissus thomsoni*, (**i**) *Tilia tuan*, (**j**) *Philadelphus pekinensis*, (**k**) *Ginkgo biloba*, (**l**) *Phyllostachys propinqua*, (**m**) *Lonicera maackii*, (**n**) *Armeniaca sibirica*, (**o**) *Magnolia denudate*, (**p**) *Eucommia ulmoides*, (**q**) *Tilia tuan*, (**r**) *Broussonetia papyrifera*, (**s**) *Sophora japonica*, (**t**) *Magnolia denudate*, (**u**) *Ulmus pumila*, (**v**) *Armeniaca sibirica*, (**w**) *Philadelphus pekinensis*, (**x**) *Ilex chinensis*.

process of $PM_{2.5}$ from the leaf surface by rainfall fluctuated with time (Fig. 6). It indicated that the removal process was species specific and subject to rainfall duration and species (Fig. 6).

Discussion

Species differences in accumulating $PM_{2.5}$.

The influence of morphological traits on foliar $PM_{2.5}$ accumulation was reflected by the changes in the sequence along the temporal progression (Fig. 1). For broadleaf species, large area of foliar ultrastructures, such as grooves, trichomes and glands, were exposed and captured the ambient $PM_{2.5}$ during the process of leaf expansion. The $PM_{2.5}$-capture capacity of such species is expected to increase as they grow mature. This has been observed in *Ginkgo biloba*, *Ulmus pumila*, and *Salix babylonica*[23]. Therefore, a collection of species with different phenology would maximize the $PM_{2.5}$ trapping effects. Thus, increased biodiversity would extend the period of leaf expansion and maximize $PM_{2.5}$ capture. On a large scale, conifers had higher rates $PM_{2.5}$ compared in broadleaved trees in urban environments[18]. In computer simulation

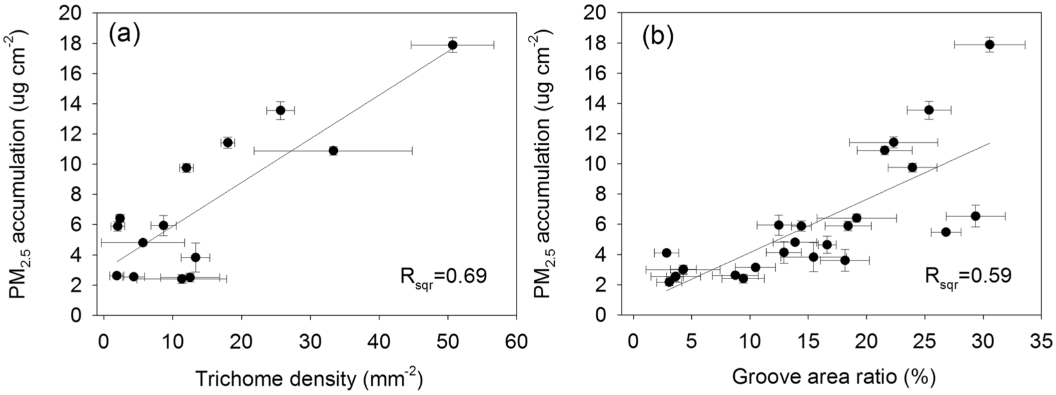

Figure 4. Relationship between foliar $PM_{2.5}$ accumulation and trichome density (**a**) and groove area ratio (**b**).

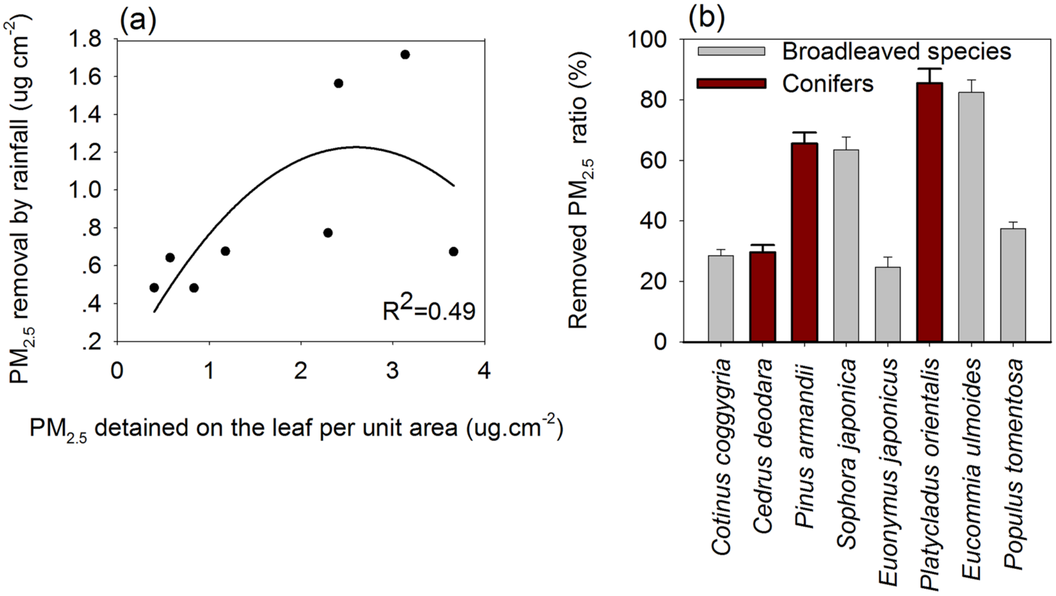

Figure 5. Foliar $PM_{2.5}$ removed by simulated rainfall (equivalent to 7.5 mm outdoor rainfall over 15 minutes) in relationship to total foliar $PM_{2.5}$ accumulation (**a**) and $PM_{2.5}$ removal percentage among species (**b**).

studies, conifers ranked highest in accumulated particulate matter on their foliage[46, 47]. The capture efficiency differences between the needles and the flat leaves can be expressed by the Stoke's number[31] which describes the relationship between the stopping distance of a particle and the characteristic dimension of an object (m)[48]. In short, narrow conifer needles have much larger Stoke's numbers and thus higher capture efficiency. For instance, in the study comparing species including both coniferous and broadleaved trees, the maximum Stoke's number for the coniferous species was 0.05 while the maximum Stoke's number for broadleaved species was 0.000012[31]. Additionally, in principle, more turbulent flow could occur across fine cylinders, like coniferous needles, than across large plates, like broadleaves, leading to the reduced boundary layer thickness of needle leaves. Small individual leaf area[49] of needles is another factor for developing thinner boundary layer in comparison with broad leaves. When the wind carrying $PM_{2.5}$ travels across the leaf, the boundary layer stays relatively stationary and forms a barrier between the surrounding air and the leave surface. Therefore, the thin boundary layer of long narrow needle leaves experienced more potential for $PM_{2.5}$ contact with the leaf surface. Conifer leaf morphology increased the potential for $PM_{2.5}$ capture but did not impact $PM_{2.5}$ release during a rain event as indicated by the high removal ratio (Fig. 5b) under rainfall. Studies on the self-cleaning of leaf surfaces have revealed that epicuticular wax ultrastructures are correlated with the hydrophobic properties of the leaf surfaces and leaf surface $PM_{2.5}$[50, 51]. Therefore, the pine species show greater $PM_{2.5}$ attenuation capabilities in urban areas, especially in winter when pollution concentrations are the highest and broadleaf tree species are leafless. However, pine species are not recommended to use in heavily polluted areas because they are susceptible to pollutant-induced injuries[34, 47] For example, ozone induces visible injury on pine needles[52]. Sulfur dioxide (SO_2) causes foliar necrosis in pine trees[53], and aluminium causes nutrient imbalance and structural changes in the pine needles[54]. Species response to pollutant loading is important for estimating total greening impacts on $PM_{2.5}$ reduction potential. For example,

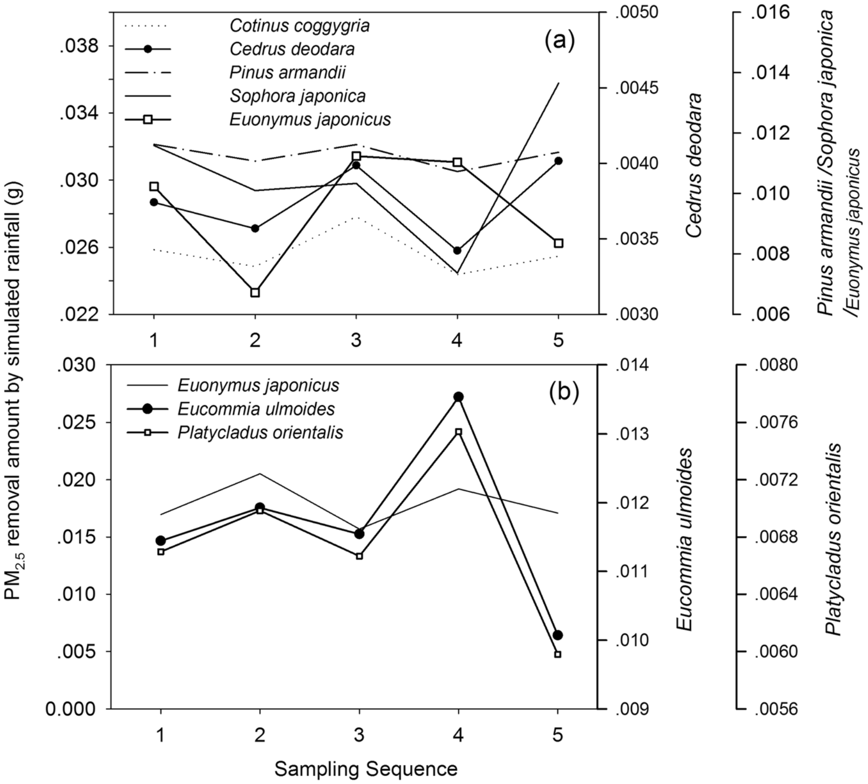

Figure 6. Progression pattern of foliar $PM_{2.5}$ removal by simulated rainfall (equivalent to 7.5 mm outdoor rainfall, over 15 minutes). The sampling time interval was 3 minutes. Sampling sequence from 1 to 5 represented samples taken at 3 minutes, 6 minutes, 9 minutes, 12 minutes and 15 minutes, respectively, from the start of simulated rainfall.

if a conifer has twice the $PM_{2.5}$ capture capacity per unit leaf area of a broadleaf tree, but only a third of the leaf area due to pollutant stress, then the broadleaf species would be more effective at capturing particulate matter at the stand level.

Influences of leaf micromorphology on $PM_{2.5}$ accumulation. For broadleaved species, leaf shape and venation did not have a significant influence on the $PM_{2.5}$ immobilization because the individual leaves cannot reflect the physical properties of canopy density. Canopy density influences the wind turbulence which has been proposed as a significant explanatory factor for the deposition of particulate matter[55], especially for the fine particles. Moreover, canopy density has significant influence over the air $PM_{2.5}$ concentration[56], and thus leads to different $PM_{2.5}$ deposition on leaves. However, the leaf micromorphology such as the groove area and trichomes (Fig. 3) also significantly influences the $PM_{2.5}$ deposition. Foliar surface morphology has been observed to have direct effects on the $PM_{2.5}$ capture by leaves. Specifically, leaf surfaces with grooves or trichomes have a higher capacity for $PM_{2.5}$ retention than smooth leaves (Fig. 4). This finding is consistent with those of previous studies where the degree of leaf roughness and the number of trichomes in upper and lower epidermis of a leaf determined the species dust retention capacity[57, 58]. Additionally, an increased roughness due to leaf hairs, scales, glands, furrows and veins, has been found to increase the particulate accumulation[33, 43, 59–61]. This study also found that dense leaf grooves provided an ideal condition for the deep retention of the $PM_{2.5}$. Foliar trichomes improved the $PM_{2.5}$ capture capacity of leaves. Species with densely haired leaves, such as *Catalpa speciosa*, *Ulmus pumila* and *Broussonetia papyrifera* were the most effective measured species for retaining $PM_{2.5}$ (Fig. 4) as indicated by the amount of $PM_{2.5}$ retention by the leaves after they were fully expanded. The effect of dense trichomes was also reported by other studies. Compared with the adaxial surface, the abaxial leaf surface is less efficient for the deposition of $PM_{2.5}$[61] due to the lighter micro-roughness of the surface. Therefore, species with abaxial indumentum (a covering of trichomes) were proved more effective in trapping $PM_{2.5}$[31]. For example, the hairy abaxial surface of *Platanus occidental* is reported to be more efficient at capturing $PM_{2.5}$ than the adaxial surface[40]. A previous study[44] ranked tree species $PM_{2.5}$ capture capacity based on visual observation of trichomes occurrence rather than density as was done in this study. The existence of trichomes would not necessarily correlate with the increased foliar $PM_{2.5}$ retention because sparse hairs had limited $PM_{2.5}$ retention ability. This was consistent with the SEM statistical results that indicated by pattern of $PM_{2.5}$ accumulation for species with a trichome density below 20 mm^{-2} (Fig. 4a). The limited ability of leaves with few trichomes was further evidenced in the SEM images, for species such as *Tilia tuan* (Fig. 3q). With improved trichome statistics from the detailed SEM images in this study, we were able to refine the previous analysis by developing a numerical relationship between trichomes and foliar $PM_{2.5}$ amount. Therefore, visual leaf macromorphological traits may not be sufficient for determining a species $PM_{2.5}$ capture capacity, leaf micromorphological examination may be necessary. This is also the case with the description of foliar roughness represented by groove area ratio.

Stomata size and stomatal density are considered important factors in controlling $PM_{2.5}$ accumulation[62]. For example, an indoor high-dose dust-spray experiment observed discernable particulate distribution around the stomas[35]. However, no significant influence of stomatal charateristics on foliar $PM_{2.5}$ amount trapping was observed in our study. The complex wind turbulence in the outdoor environment may create changing diffusion conditions leading to the lower exposure doses for the plants and disturbances for the deposition of particulate matters. Therefore, the foliar deposition of particulate matters in the controlled experiment would be more evident than in the outdoor natural environment. Moreover, this might be attributed to the lower stomatal density and stomata size in our samples than those in the studies that observed significant relationship between stomatal features and particles. Another study conducted in Beijing[63] observed that leaves with high stomatal density ($>189\,mm^{-2}$) demonstrated significant increase with stomatal density in trapping $PM_{2.5}$. However, this relationship was absent among species with lower stomatal density. Therefore, stomatal density appears to only increase particle capture when the stomatal density is high. This is proved by the comparison of another pair of studies. Stomatal density was related to foliar accumulation of air contaminant for species with high leaf stomatal density (ranging averagely from $237–757\,mm^{-2}$)[64]. By contrast, in another study where the leaf stomatal density ranged from $10.36–38.36\,mm^{-2}$ in average, no significant relationship was found between foliar particle accumulation and stomatal density[65]. Also, the stomatal size in our study was low, ranging from 10 to 25 μm. The study that observed similar stomatal size range (14.5–19.9 μm)[64] also failed to show consistent relationship between stomata size and foliar air pollutant accumulation. By contrast, this relationship was found in the study where the stomata size ranged from 20 to 192 μm[65]. The state of stomatal opening is another reason for the inconsistent relationship between stomatal density or stomata size and foliar $PM_{2.5}$ accumulation. The stomatal density and stomata size does not necessarily stand for the opening size of the stomas. Sensitive to ambient environmental factors, such as light and water status, the stomas can open to different extent, and thus lead to different rates of transpiration which in turn alters relative humidity. Given that relative humidity influences dry deposition velocity, foliar accumulation of $PM_{2.5}$ capture could be impacted[66]. Although the particles have been observed to enter the leaf through stomatal openings[67], the frequency to which this occurs is unknown. Blockage of stomata with PM could significantly decreased stomatal conductance and gas exchange, which may further influence the water regime, photosynthesis[67], and overall plant growth[68]. Therefore, we believe the entering of $PM_{2.5}$ through the stomatal openings should be an occasional observation.

Rainfall effect on $PM_{2.5}$ retention. The across-seasonal comparisons of the same species indicated that the foliar $PM_{2.5}$ accumulation did not necessarily increase with time (Fig. 1), which may have been caused when the maximum loading capacity of leaves was reached[3, 23, 58]. For example, plant leaves reached their maximum $PM_{2.5}$ loading capacities after 26 days of no rainfall in Guangzhou, China[69]. Therefore, the accumulation of $PM_{2.5}$ on leaves is not linearly related to exposure duration. Foliar $PM_{2.5}$ accumulation is dynamic. Leaves may capture $PM_{2.5}$ for some time before a wind event releases the material back into the air, or a rain event washes the material off of the leaf. Therefore, the amount of foliar $PM_{2.5}$ at the end of the examined period cannot be interpreted as a representation of the total mass of foliar $PM_{2.5}$ accumulation during the corresponding period (e.g., growing season).

This study examined the differences in rainfall removal of foliar $PM_{2.5}$ among species (Fig. 6b). Although the kinetic energy of rainfall is the predominant factor in the foliar washing process[70], leaf surfaces features like wax layer, trichomes and other protrusions can result in different contact angle between water droplet and different leaf surfaces[71]. These factors create different water-repellent performances between species[72]. Moreover, the hydraulic pressure change due to the impaction of raindrop can change the contact angle and thus the leaf wettability[73] which could also contribute to the $PM_{2.5}$ rainfall removal patterns. Measurements taken immediately after a rainfall indicated that once deposited, coarse and fine particles were not easily washed off of the leaves[18]. Therefore, modelling the associated processes throughout the entire season to obtain an accurate estimate of the amount of $PM_{2.5}$ immobilized by urban trees is necessary.

The amount of $PM_{2.5}$ accumulated on a leaf is therefore a combination of multiple factors, including species factors and meteorological condition[24]. The amount of washed-off $PM_{2.5}$ was not significant related to the leaf morphological traits but was significantly related to the total foliar $PM_{2.5}$ accumulation. This result suggests that mechanisms that determine rainfall loss of foliar retention of $PM_{2.5}$ may apply equally to broadleaf and coniferous species.

The present study demonstrates the $PM_{2.5}$ attenuation dynamics of different species and provides insights for species selection for $PM_{2.5}$ pollution mitigation in urban areas. Nevertheless, trees may also act as $PM_{2.5}$ sources by emitting biogenic volatile organic compounds (BVOCs)[74] and enhancing local $PM_{2.5}$ concentration under dense planting schemes[75]. BVOCs can react with nitrogen oxides (NO_x) to form O_3 and secondary organic aerosol (SOA)[76]. Therefore, the release of BVOCs from vegetation may pose a problem if the planted species are high emitters[77]. BVOC emissions can vary widely among tree species and even within species[78, 79], depending on physiological and environmental factors[76]. For instance, in Mediterranean areas, where summer is usually characterized by high temperature and little precipitations, the potential for BVOC and ozone formation is high[78]. Therefore, BVOC emissions should be considered during the design of urban green spaces and trees with high BVOC emissions should be avoided to achieve the improved net air pollution reduction benefits from the tree planting. Although $PM_{2.5}$ dispersion was not the focus in the present study, the release of deposited $PM_{2.5}$ from foliage should be considered. A simulation of tree and shrub effects on particle dispersion suggested that particulate matter concentrations would be highest on streets with a high density of trees[80]. Based on dispersion conditions, different planting configurations were showed to have varied abilities to mitigate airborne $PM_{2.5}$ concentrations[44]. Therefore, to optimize the benefits of trees in various urban settings, the pros and cons of different taxa in relation to $PM_{2.5}$ pollution must be considered[81].

In conclusion, needle-leaved coniferous species are more efficient at removing atmospheric $PM_{2.5}$ and have a higher potential than broadleaved species to recapture $PM_{2.5}$ after rain events. For broadleaved species, macromorphological traits, such as leaf shape and venation, do not have a significant influence on foliar $PM_{2.5}$ retention, whereas micromorphological traits, such as grooves and trichomes, are strongly correlated with foliar $PM_{2.5}$ accumulation and can be used as effective species selection criteria. The temporal process of foliar $PM_{2.5}$ wash-off is highly species-specific, which implies the influence of rainfall duration and intensity on the ability of leaves to accumulate $PM_{2.5}$. A cross-season comparison showed positive and negative increases in $PM_{2.5}$, which reflected the varying influence of resuspension on the amount of foliar $PM_{2.5}$ accumulation. These findings indicate that the accumulation-suspension cycle of urban trees must be further investigated to accurately evaluate accurately the long-term potential bio-filtration capacity of different trees.

Methods and Materials

Leaf-washing experiments for the species comparison. Samples for the leaf-washing experiments were collected on the campus of Beijing Forestry University (40°00′ N, 116°34′ E), Beijing, China, which offers abundant vegetation species within a radius of 500 m. Thus, it is reasonable to assume (for the purpose of comparison) that the vegetation is exposed to the same $PM_{2.5}$ concentrations. Thirty-one tree species (specific species are listed in the results) were tested for their capacity to accumulate $PM_{2.5}$ on the leaf surface. Leaves of 15 species were collected in April 2015 as late spring samples, and the leaves of 16 additional species were collected during September 2015 as the summer-autumn samples. The following criteria applied to leaf collection days: sunny and wind speed less than $5\,m\,s^{-1}$. Lower wind speeds ensured that the wind would not affect the particle deposition on the leaf[82]. A portable meteorological station (NK4500, Kestrel Co., Philadelphia, PA, USA) was used to measure the sampling conditions in the middle of an open green space at a height of 10 m. All the branches were firmly held and cut carefully from a height of 1.5–2.5 m to avoid losing particulate matter from the leaves. Each species included three sampled trees, and eight small peripheral branches at four azimuth angles were cut from one sampled tree. This sample size provided a leaf area range of 300 to 500 cm^2. During collection, samples were immediately closed and labeled in plastic bags to avoid contamination, and they were stored in the lab in a freezer (−18 °C).

$PM_{2.5}$ can be trapped both on the leaf surface and in the leaf wax. However, only $PM_{2.5}$ deposition on the leaf surface was assessed in this study because of the environmental concerns of using chloroform[81] and the relatively low ratio of in-wax $PM_{2.5}$[20]. Once the samples were removed from the freezer, 10 g (fresh weight) of leaves were washed with distilled water and brushed carefully so that the $PM_{2.5}$ was fully removed from the leaf surface. The solution was run through a metal sieve with a mesh diameter of 100 μm to obtain a suspension of liquid sample I. Ten percent of the liquid sample I was injected into pre-weighted PP plastic bag (W_1) and dried. The dried bag was weighted again (W_2), and the difference between W_2 and W_1 was the weight of 10% of the total suspended particulates (TSP) in the rinse water, which was translated to the TSP amount (W_{TSP}) in the original liquid sample by dividing by 0.1. The remaining 90% of water sample I was pumped through filters (PTFE membrane, Whatman, UK) of 10 μm and then of 2.5 μm to intercept particles with a diameter of 10–100 μm and 2.5–10 μm, respectively[81]. The filters used for the analysis were first soaked in distilled water for 2 hours and then dried at 105 °C in a drying chamber for 3 hours to remove soluble impurities, and they were then placed in a balancing chamber for 48 hours to stabilize the humidity change. Filters were weighed before and after filtration (XS105DU balance, Mettler-Toledo International Inc., Switzerland). The resulting weight of the $PM_{10\text{-}100}$ and $PM_{2.5\text{-}10}$ only account for 90% of the original rinsing liquid and therefore should be divided by 0.9 to obtain the total $PM_{10\text{-}100}$ ($W_{PM_{10\text{-}100}}$) and $PM_{2.5\text{-}10}$ ($W_{PM_{2.5\text{-}10}}$). The $PM_{2.5}$ mass was then calculated as the difference between the W_{TSP} and the sum of $W_{PM_{10\text{-}100}}$ and $W_{PM_{2.5\text{-}10}}$.

To facilitate the species comparison, leaf area-normalized $PM_{2.5}$ accumulation results (i.e. in the unit of $\mu g\,cm^{-2}$) are required. Therefore, the leaves were scanned, and the surface areas were obtained from digital images processed with Photoshop (version: Photoshop CS5, Adobe Systems Incorporated, San Jose, CA, U.S.). Branches were excluded from the $PM_{2.5}$ deposition quantification due to issues with accurately assessing branch area and the potentially low ratio in the total plant area index[83]. For needle leaves, we measured water displacement to determine leaf volume and converted the volume to the leaf area according to the following formula:

$$A_L = 2L\left(1 + \frac{\pi}{n}\right)\sqrt{\frac{nV}{\pi L}} \tag{1}$$

where A_L is the leaf area, V is the water displacement volume as a the substitute of the needle-leaf-volume, n is the number of needle leaves in a single bundle, and L is the average length of the needle leaves[84].

Microscopic observation of foliar morphology. Leaf samples were examined under an environmental scanning electron microscope (ESEM, Quanta 200 FEG, FEI, USA) operated in the low vacuum mode (15 kV, 80 Pa) to test for relationships between foliar micromorphological traits and $PM_{2.5}$ accumulation. These analyses were conducted on the same day to prevent desiccation and subsequent alteration of leaf surface micromorphology[3]. Two pieces ($1 \times 1\,cm^2$) were excised from the centre of the lamina of each leaf[22]. Then, two adaxial specimens and two abaxial specimens[23] were coated with a thin conductive film of platinum in order to increase electrical conductivity and improve optical transmission. The processed samples were mounted on the stubs for microscopic observations. Grooves, trichomes and stomata were included for further analysis after a preliminary visual screening of the scanning images to determine the effective micromorphological traits for $PM_{2.5}$ retention. The roughness of the leaves on the abaxial or adaxial side was quantified in terms of the groove area ratio (%), which represents the ratio of groove area to total leaf area of the specimen;

$$Groove\ area\ ratio = \frac{A_G}{A_L} \times 100\% \tag{2}$$

where A_G stands for groove area (μm^2) and A_L stands for total leaf area (μm^2). Due to the overlapping of trichomes, this trait could not be quantified in a measurable unit. Instead, the pubescent area (defined as the surface bearing trichomes) as calculated as a measurement of the trichomes density. The stomata were quantified in terms of stomatal density (SD, mm^{-2}) and stomatal size (evaluated in diameter, DS, μm).

Simulated rain wash experiment. A controlled rainfall wash experiment was conducted to evaluate the species differences in resisting rainfall removal of foliar $PM_{2.5}$. Branches with leaves of different species were fixed in a container with a hole at the bottom for sampling water, and they were then placed underneath an artificial rainfall system. The system was installed at the ceiling of a room with a base area of 400 m^2. Based on the statistical data provided by Haidian District Water Authority, Beijing, China (http://hdsw.bjhd.gov.cn/zxfw/bmcxfw/hdqyqcx/index_3.htm), the average daily rainfall during the sampling period was 8.4 mm, and individual rainfall event s varied between 2–10 mm most frequently in the region. Combined with the settings of the system, the samples were subjected to a simulated rain event of 7.5 mm that lasted for 15 minutes. The water containing the washed foliar $PM_{2.5}$ was sampled every 3 minutes to examine the change in the amount of $PM_{2.5}$ washed off the leaves. Each species had three repetitions. In addition, to exclude the influence of impurities from the water, a parallel experiment was conducted for the control group, and it included three containers with the same configurations as all of the others but with no branch sample. The final result of the washed-off $PM_{2.5}$ was calculated as the difference between the water samples from planting containers and that from the control group.

Statistical analysis. Significant differences among species were examined via one-way ANOVA in SPSS 18.0 (SPSS Inc., Chicago, IL, USA). The Bonferroni correction method was applied for the comparison statement because of the unequal sample size among groups. Curve fitting and plotting were conducted using Sigmaplot software 12.5 (Systat Software, San Jose, CA, USA).

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Author Contributions

L.X.C. contributed to the literature search, study design, data analysis and interpretation, and manuscript writing and revision. L.Z., C.M.L. and R.Z. contributed to study design, experiment performance and data processing. Z.Q.Z. led the study design and field experiments, and revised the draft manuscript.

Additional Information

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