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An investigation on the leaf accumulation-removal efficiency of atmospheric particulate matter for five urban plant species under different rainfall regimes

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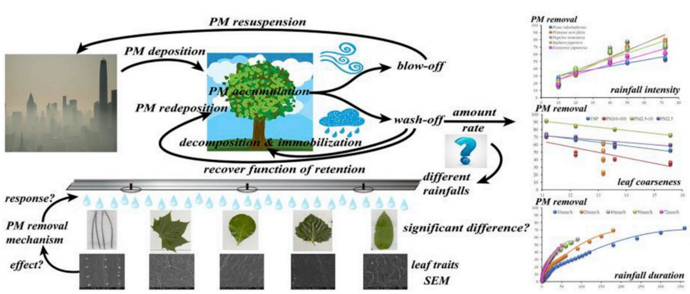
GRAPHICAL ABSTRACT



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ABSTRACT

Urban trees and forests are widely used as biological filters to combat the airborne particulate matter (PM). Precipitation washing PM off from plants is regarded as filter cleaning, which is a key factor for recovering the function of foliar PM filtering. However, it is uncertain on how much PM can be total filtered by urban trees due to lack of understanding about how PM deposition, removal, resuspension and redeposition interact with species and rainfall variability. For this reason, we developed a study to determine foliar PM removal amount and rate of different sizes for five plant species commonly used for urban greening by simulated different rainfall regimes. Our specific objectives were to: (1) explore the difference in PM removal between different plant species and different rainfall patterns; (2) understand the response of foliar PM removal as a function of rainfall characteristics; and (3) quantify the relationship between foliar PM removal rate and leaf coarseness. Results showed that significant differences ($P < 0.05$) in PM removal amount and rate were found not only between different species within the same rainfall pattern, but also between different rainfall patterns for the same species. PM removal rates from the leaf surface were significantly correlated with rainfall intensity ($P < 0.01$). Different size PM cumulative removal rate exhibited an exponential loss with rainfall duration ($P < 0.01$). For smooth leaf surfaces, long duration-low intensity rainfall could increase PM removal rate while for rough leaf surfaces, short duration-high intensity rainfall could achieve a larger removal rate using the same amount of total rainfall. Additionally, more PM was removed by rainfall than that by water washing. The findings from this study have implications for better estimating long-term air purification potential of urban plants, and for air phytoremediation planning in urban areas.

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1. Introduction

Airborne particulate matter (PM) has the potential to exert negative impacts on human health, and therefore has been an issue of concern from the global to urban scale (Chen et al., 2017; Dzierżanowski et al., 2011; Freer-Smith et al., 2004; Speak et al., 2012; Wang et al., 2015). Urban trees and forests are found to be significant sinks for gaseous, aerosol, particulate and rain-borne pollutants (Fowler et al., 1989; Freer-Smith et al., 2004). Urban trees and forests can reduce the emission of PM by covering the soil surface (Endalew et al., 2009; Escobedo et al., 2011), alleviate PM concentration and re-suspension by directly capturing particulates through their large leaf area (Beckett et al., 2000a,b; Freer-Smith et al., 2004; Terzaghi et al., 2013), and facilitate deposition of particulates by improving micrometeorological conditions (Beckett et al., 1998; Buccolieri et al., 2011, 2009; Chen et al., 2017; Freer-smith et al., 2005). Five mechanical processes control PM deposition onto leaves, including sedimentation under gravity; Brownian diffusion; interception; inertial impaction; and turbulent impaction (Petroff et al., 2008a). The large leaf area and more turbulent air movement caused by their structure make trees particularly effective for particulate removal (Beckett et al., 2000a,b; Fowler et al., 1989; Hofman et al., 2014; Speak et al., 2012). Therefore, urban trees and forests are widely used as biological filters to combat the airborne particulate matter and thus to provide healthier and cleaner air for urban residents (Nowak et al., 2006; Terzaghi et al., 2013). A quantitative and comprehensive assessment of PM retention on the leaves is very important for accurately assessing the air purification ability and efficiency of the urban forests (Liu et al., 2018). Some relevant studies have been conducted on vegetation retention capacity (Freer-Smith et al., 2004; Liu et al., 2015; Przybysz et al., 2014), species difference (Chen et al., 2016a,b; Sæbø et al., 2012), mechanism of deposition (Beckett et al., 1998; Freer-smith et al., 2005; Petroff et al., 2008a; Smith and Staskawlcz, 1977), factors of influencing foliar captured PM (Hofman et al., 2016; Räsänen et al., 2013) and purification ability of urban forests (Chen et al., 2016a,b; Escobedo et al., 2011; Litschke and Kuttler, 2008; Nowak et al., 2006; Speak et al., 2012) in recent years to assess how effective trees are at removing PM from the atmosphere (Beckett et al., 2000a,b; Chen et al., 2016a,b; Jeanjean et al., 2016).

A better understanding of which plant species and their micro- and macro structural traits are most effective in accumulating PM, may allow improvement to air quality through adaptive management (Leonard et al., 2016). Microstructural leaf features like rough surfaces, thick waxy epicuticles, low leaf wettability and low stomatal density along with macrostructural features such as increased plant height, whorled leaf arrangements, larger leaf area and shorter petiole length are all leaf traits that enhance PM accumulation (Leonard et al., 2016; Nowak et al., 2006; Räsänen et al., 2013; Speak et al., 2012). In general, broad-leaved species with rough leaf surfaces are more efficient in capturing PM than broad-leaved species with smooth leaf surfaces (Beckett et al., 2000a,b). Evergreen conifers are considered to be more effective in PM accumulation than broad-leaved species due to their thicker epicuticular wax layer and the potential for accumulating toxic pollutants throughout the year (Beckett et al., 1998). Additionally, it is impossible to update needles every year because most of these plants keep their needles for several years (Beckett et al., 1998; Chen et al., 2017; Dzierżanowski et al., 2011).

However, a plant's capacity to capture PM is also a function of environmental factors such as wind, temperature, and most importantly, precipitation. Some leaf captured particulates can be washed off from the leaves by precipitation and deposited onto the ground where the organic component of PM can be decomposed and inorganic PM components immobilized in the soil (Dzierżanowski et al., 2011). The PM wash-off is regarded as filter cleaning, leaving the leaves ready for more PM deposition. This process is beneficial in the planning of air phytoremediation (Wang et al., 2015). Therefore, precipitation washing PM off from the leaf is a crucial factor for recovering the function of foliar PM filtering. For this reason, analysis of the PM removal effects of precipitation are essential for estimating the amount of PM retained on leaves during a growing season or a year (Wang et al., 2015a). A few studies have attempted to estimate PM removal by rainfall and found that capacity varied by tree species and precipitation amount. Approximately 30%–40% of the PM on the leaves of *Pinus sylvestris* can be removed by a 20 mm rainfall (Przybysz et al., 2014). While another study indicated that 28% and 48% of accumulated PM were washed off from leaves of *Ligustrum lucidum* by two rainfall events of 10 mm and 32 mm, respectively (Wang et al., 2015a). A more recent study indicated that simulated rainfall removed 51%–70% of PM from four immature twigs (Xu et al., 2017).

However, no study has systematically examined whether there is a difference in foliar PM removal of different species or different rainfall patterns, how the PM removal process responds to various rainfall intensities, durations, and patterns and how the leaf traits of different species affect PM removal. Such information is necessary in accurately evaluating the total PM deposition during a season or year. Therefore, this study aims to determine foliar PM removal amount and rate of different sizes for five plant species commonly used for urban greening by simulated different rainfall regimes. Our specific objectives were to: (1) explore the difference in PM removal amount and rate between different plant species and different rainfall patterns; (2) understand the response of foliar PM removal process as a function of rainfall characteristics (i.e., rainfall intensity, rainfall duration and rainfall pattern); and (3) quantify the relationship between foliar PM removal rate and leaf coarseness. The findings from this study have important management implications for assessing the potential for urban plants remove airborne PM.

2. Materials and methods

2.1. Plant species selection and sample collection

Four tree species (*Pinus tabulaeformis*, *Platanus acerifolia*, *Populus tomentosa*, *Sophora japonica*) and one shrub (*Euonymus japonicus*) commonly used in urban greenspace and roadside greening in Beijing were

Table 1
Simulated rainfall and PM sampling setting.

Rainfall Intensity (mm h^{-1})	Rainfall Duration (h)	Rainfall Total (mm)	Sampling setting during the rainfall		
			Sampling Intervals (Sampling Times)		
10	6	60	3 min (10)	9 min (10)	60 min (4)
20	3	60	1.5 min (10)	4.5 min (10)	30 min (4)
40	1.5	60	45s (10)	2min15s (10)	15 min (4)
50	1.2	60	36 s (10)	108s (10)	12 min (4)
72	1	72	25 s (10)	75s (10)	8 min20s (4), 10min (1)

selected for this study (Table A1). Sampling was conducted on July 10, 2016 when there was no previous rainfall for more than five days. For each species, four sampled plants were selected and all plants had no sign of insect or disease damage. Five mature and healthy branches were cut from the traffic-exposed side (east side of the road) and subjected to five rainfall intensities. Sampling height varied from 0.6 to 4.0 m above ground level depending on plant structure. All the sampled branches were from the same location, same direction of the canopy exposed to the same atmospheric environment. Under such circumstance, only species specific features have the influence on the detention of PM. A total of 100 experimental branches were obtained (i.e., 5 species × 4 stems/species × 5 branches/stem). To obtain sufficient material to conduct the PM filtration and rainfall simulation experiment, the length and diameter of each sampled branch were about 0.45m and 0.25m to fit the holding boxes used for rainfall stimulation. Each branch was placed in a polyethylene bag, then closed, labeled, and transported to the laboratory. Leaves with a total surface area of 300–400 cm^2 were cut from five branches of each sampled plant for each species and used to determine the total PM accumulation on the surface. The remaining part of sampled branches were used to conduct the rainfall simulation experiment to determine foliar PM removal efficiency. One branch was removed from each species and used as an experimental control with all the leaves removed to exclude the influence of PM retained on the branchlets. As a result, each species has three sampled branches as repetitions and one control under each simulated rainfall trial.

2.2. Rainfall simulation system

Rainfall simulation experiment was carried out in an indoor condition to avoid the wind effect during the experiment. We used a trough rainfall simulator developed jointly by Beijing Normal University and Beijing Jiaotong University. The rainfall simulator simulated a wide range of rainfall intensities from 10 to 132 mm h^{-1}. One trough rainfall simulator was composed of five nozzles and the distance between the two nozzles was 1.1 m. A single nozzle had a rectangular rainfall area of 5.4 m^2. The combined multiple nozzles were needed to generate a uniform rainfall distribution. The rainfall simulator was positioned 2.5m above the ground and the distance between the two simulators was 1.5m. The diameter of the simulated raindrops was similar to natural raindrop distribution and size (Huo et al., 2015). The homogeneity coefficient of simulated rainfall intensity was more than 0.89 (Yun et al., 2008).

2.3. Experiment design

Rainfall intensities were set to 10, 20, 40, 50 and 72 mm h^{-1} and the duration of the water addition was set to 6, 3, 1.5, 1.2 and 1h correspondingly, representing a spectrum of rainfalls from long time-low intensity to short time-high intensity events. The rainfall simulation equipment was recalibrated after each rainfall trial. Distilled water was used for rainfall simulations to avoid the possible influence from physical properties and chemical matter in water on the particulates measurement.

Four boxes 30 cm long, 30 cm wide and 50 cm high were used to harvest the runoff and washed off particulates from plant branches for each simulated rainfall experiment. An outlet in the bottom of the box was equipped to collect the runoff. A 1.5 cm wide by 10 cm tall cylinder was welded inside the center of the box to support and fix the sampled branches. The runoff sampling structure was designed to capture the rainfall displaced PM. Runoff sampling time was listed in Table 1. Three repetitions (i.e., branches with leaves in the box) and a control (i.e., branches without leaves) for each species were carried out at the same time (Fig. 1).

2.4. Leaf washing and PM filtration

Leaves collected from sampled branches before the rainfall simulations were first washed in distilled water and filtered to determine the leaf amount of PM. A no-hair-loss brush and tweezers were used for washing the leaf surfaces. These measurements represent PM that can be washed off from the leaves by water (for simplicity, in this paper termed 'WPM' hereafter). Every solution was passed through a metal sieve with mesh diameter of 100 μm to get a suspension liquid Sample I. Ten percent of the liquid Sample I was injected into plastic bag that was pre-weighed as W1 and then dried in the oven with the temperature of 105 °C until the water evaporated completely. The dried bag was re-weighed as W2. Difference between W2 and W1 was the weight of 10% of the total suspended particulates (i.e., TSP) in the rinse water, which can be translated to the TSP amount in the original liquid sample divided by 0.1. The remaining 90% of water sample I was pumped through two types of micro porous membranes (PTFE membrane, Whatman, UK) with the pore size of 10 μm and then of 2.5 μm to intercept particulates with a diameter of 10–100 μm and 2.5–10 μm, respectively (Dzierżanowski et al., 2011). All filters used for the analysis were first soaked in distilled water for 2 h and then dried at 105 °C in a drying chamber for 3 h to remove soluble impurities. The filters were then put in a balancing chamber for 48 h to stabilize the humidity change. Every filter was weighed before and after filtration three times

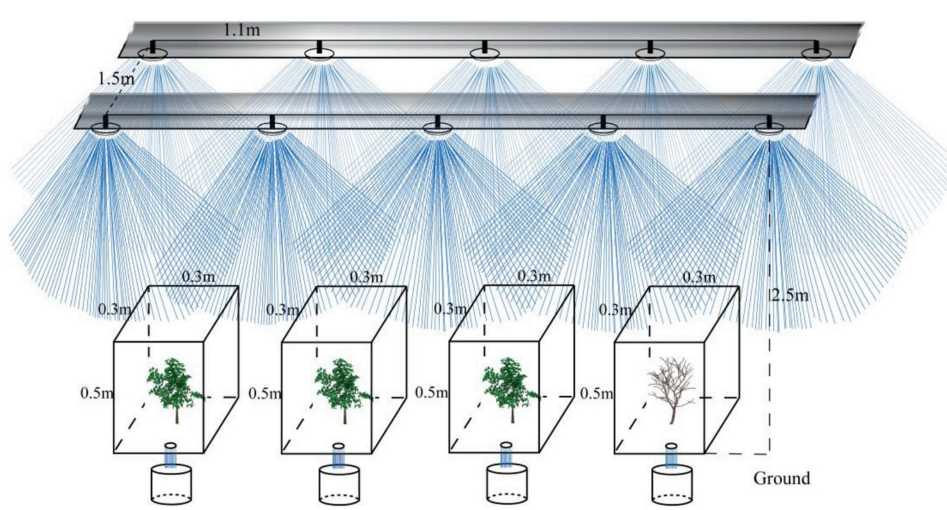

Fig. 1. The schematic diagram of rainfall experiment.

to reduce the potential errors using XS105DU balance (Mettler-Toledo International Inc., Switzerland). The filtered membranes were placed in a drying chamber at 105 °C for 3 h and then weighed as above. The resulting weight of the PM10–100 and PM2.5-10 only account for 90% of the original rinse liquid. Therefore, we divided the resulting sample weight by 0.9 to derive the total weight of PM10–100 and PM2.5-10. The PM2.5 mass was then calculated as the difference between the TSP and the sum of PM10–100 and PM2.5-10.

We used ‘PM removal rate’ to make comparison and correlation analysis to standardize the differences in original PM amount detained on the leaf surface. The water samples collected during the simulated rain event were handled according to the same method described above to filter PM. The PM that was removed from the leaves by the simulated precipitation was termed ‘RPM’. The leaves were collected after the simulated rainfall event ended, and were then washed by distilled water to calculate the weight of particulates left after rainfall was termed ‘LPM’. The PM removal rate by rainfall was calculated by Eq. (1):

$$\text{PM removel rate} = \frac{\text{RPM}}{\text{RPM} + \text{LPM}} \times 100\% \quad (1)$$

2.5. Leaf and branchlet surface area determination and calculation

We used per unit leaf area g m^{-2} to compare the PM accumulation and removal differences to standardize the leaf shape and leaf traits differences between different species. Broad leaves were dried at the ambient temperature after washing, and digitally image processed using Image J software (Version 1.48; National Institutes of Health, Bethesda, MD, USA) to measure leaf area. For needle leaves, we measured leaf volume water displacement and converted the volume to leaf area according to the following Eq. (2):

$$S = 2L\left(1 + \frac{\pi}{n}\right)\sqrt{\frac{nV}{\pi L}}. \quad (2)$$

where S is leaf area, V is water displacement volume as the substitute of needle-leaf-volume, n is the numbers of needle leaves in a single bundle and L is the average length of the needle leaves (Chen et al., 2017).

The surface area of the branches was calculated by treating each branchlet as a cylinder and measuring the diameter and length of each cylinder via a vernier scale. Then the total surface area of the whole branch was obtained by summing the area of each branchlet.

2.6. Calculating PM removal per unit leaf area

Removed foliar PM per unit of leaf area was calculated as the difference between the each of three sampled branch repetitions and that from the control group according to the following Eq. (3):

$$PM = \frac{TPM_i - \frac{TPM_c}{TBA_c} \times TBA_i}{TLA_i}. \quad (3)$$

where PM is the PM removal per unit leaf area, TPM is the total removed PM by rainfall, i are the replicates (i.e., i = 1,2,3), c is the control group, TBA is the total branch area, and TLA is the total leaf area.

2.7. Micro leaf traits scanning

Leaf samples were examined using an environmental scanning electron microscope (ESEM, Quanta 200 FEG, FEI, USA) operated in the low vacuum mode (15 kV, 80 Pa). Scanning was conducted after rainfall experiments were completed. Leaf samples were preserved by the method described below to prevent subsequent alteration of leaf surface micromorphology (Speak et al., 2012). Two samples (about

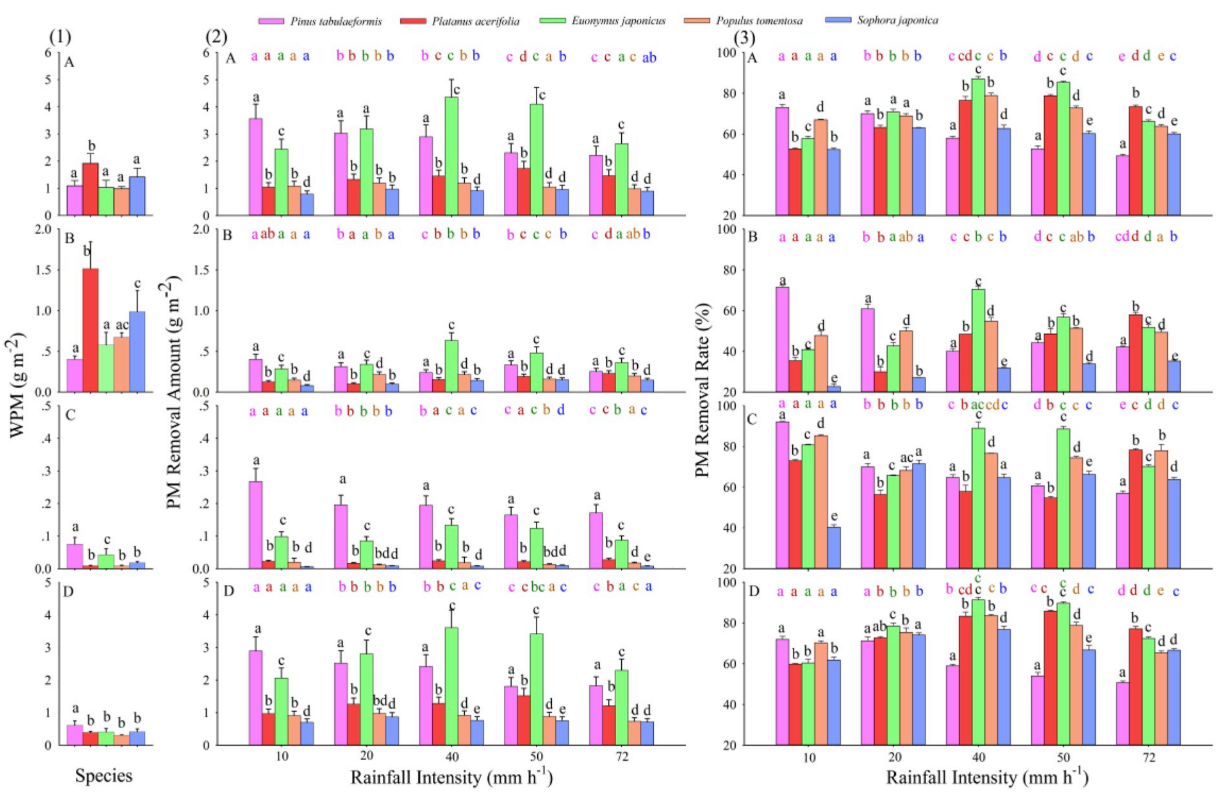

Fig. 2. Panel (1) represents WPM of different sizes. Panels (2) and (3) represent different size PM removal by the different rainfall patterns: (2) PM removal amount and (3) PM removal rate. A to D in each panel represent different PM sizes: (A) TSP, (B) PM10-100, (C) PM2.5-10, and (D) PM2.5. Data are presented as mean ± SE. The black letters indicate significant differences between different species, and the colored letters corresponding to each species indicate significant differences between different rainfall patterns.

5 mm × 5 mm) were excised from the center of the lamina of each leaf. Then, two adaxial specimens were coated with a thin conductive film of platinum to increase electrical conductivity and to improve optical transmission (Chen et al., 2017). The processed samples were then mounted for microscopic observation.

2.8. Leaf coarseness calculation

Leaf coarseness was quantified from the upper and lower epidermis of SEM images (magnified 500 times) using a Tamura textural feature extraction (Tamura et al., 1978).

2.9. Statistical analysis

Differences between species were examined using a one-way ANOVA in SPSS 18.0 (SPSS Inc., Chicago, IL, USA). The Pearson's correction method was applied for analysis correlation. Curve fitting and plotting were conducted using SigmaPlot software 12.5 (Systat Software, San Jose, CA, USA).

3. Results

3.1. Species difference in foliar PM accumulation

All five species captured PM on their leaves, but the captured amount varied in species and particulate size (Fig. 2 (1), $P < 0.05$). *Platanus acerifolia* was the most effective species in capturing TSP ($1.91\ g\ m^{-2}$) and PM10-100 ($1.52\ g\ m^{-2}$). These values are significantly higher than TSP and PM10-100 capture from the other species ($P < 0.05$) (Fig. 2 (1) A, (1) B). In addition, *Sophora japonica*, *Pinus tabulaeformis*, *Euonymus japonicus, and Populus tomentosa* captured 1.41, 1.09, 1.03 and $0.99\ g\ m^{-2}$, of TSP respectively. *Sophora japonica*, *Populus tomentosa, Euonymus japonicus* and *Pinus tabulaeformis* captured 0.99, 0.67, 0.58 and $0.40\ g\ m^{-2}$, of PM10-100 respectively. *Pinus tabulaeformis*, a coniferous species, was the most effective species in capturing PM2.5-10 ($0.075\ g\ m^{-2}$) and PM2.5 ($0.61\ g\ m^{-2}$) (Fig. 2 (1) C, (1) D). *Euonymus japonicus*, a small shrub with dense leaves, also had the significant larger accumulation of PM2.5-10 ($0.043\ g\ m^{-2}$) than other three species. There was no significant difference between other four species in capturing PM2.5. *Populus tomentosa* was the least effective in PM capture (TSP $0.99\ g\ m^{-2}$, PM2.5-10 $0.009\ g\ m^{-2}$ and PM2.5 $0.31\ g\ m^{-2}$). PM10-100 was the major particulates captured on the leaf surfaces accounting for 62 ± 16% of deposited TSP mass, while PM2.5 accounted for 35 ± 13% of the TSP mass averaged across all species.

3.2. Leaf coarseness of different species

The coarseness of the upper epidermis of *Pinus tabulaeformis* was significantly finer than other species due to regular and shallow striping (Table 2, Fig. A1 (a1)). However, the coarseness of upper epidermis of *Platanus acerifolia* was significantly more than the other four species due to intricately corrugated leaf surface (Table 2, Fig. A1 (b1)). *Sophora japonica* was the only species that had hairs on its upper and lower epidermis (Fig. A1 (e1), (e2)). The *Populus tomentosa* lower epidermis was significantly coarser than the other species due to the density of surface hairs (Table 2, Fig. A1 (d2)).

3.3. Foliar PM removal by rainfall

3.3.1. Species differences in PM removal amount

The species differences in different size PM removal amount by different rainfall intensities under total precipitation of 60 mm were shown in Fig. 2 (2). The amount of PM removed from *Pinus tabulaeformis* and *Euonymus japonicus* was significantly higher than that from the other species under all rainfall intensities ($P < 0.05$). TSP removal amounts ranged from $0.78 \pm 0.11\ g\ m^{-2}$ for *Sophora japonica* with a rainfall intensity of $10\ mm\ h^{-1}$, to $4.35 \pm 0.65\ g\ m^{-2}$ for *Euonymus japonicus* with a rainfall intensity of $40\ mm\ h^{-1}$ (Fig. 2 (2) A). PM10-100 removal amounts ranged from $0.08 \pm 0.01\ g\ m^{-2}$ for *Sophora japonica* with a rainfall intensity of $10\ mm\ h^{-1}$, to $0.63 \pm 0.09\ g\ m^{-2}$ for *Euonymus japonicus* with a rainfall intensity of $40\ mm\ h^{-1}$ (Fig. 2 (2) B). PM2.5-10 removal amounts ranged from $0.0056 \pm 0.0008\ g\ m^{-2}$ for *Sophora japonica* with a rainfall intensity of $10\ mm\ h^{-1}$, to $0.26 \pm 0.04\ g\ m^{-2}$ for *Pinus tabulaeformis* with a rainfall intensity of $10\ mm\ h^{-1}$ (Fig. 2 (2) C). PM2.5 removal amounts ranged from $0.26 \pm 0.10\ g\ m^{-2}$ for *Sophora japonica* with a rainfall intensity of $72\ mm\ h^{-1}$, to $3.61 \pm 0.54\ g\ m^{-2}$ for *Euonymus japonicus* with a rainfall intensity of $40\ mm\ h^{-1}$ (Fig. 2 (2) D).

More TSP was removed by rainfall (RPM) than by water washing (WPM) for *Pinus tabulaeformis*, *Euonymus japonicas* and *Populus tomentosa* (Fig. 2 (1) A and Fig. 2 (2) A). More of the PM2.5-10 mass was removed by rainfall than by the water washing except *Sophora japonica* (Fig. 2 (1) C and Fig. 2 (2) C). The mass of PM2.5 removed by all intensities of rainfall was more than the amount of PM2.5 mass removed by water washing across all species (Fig. 2 (1) D and Fig. 2 (2) D). However, the amount of PM10-100 mass removed by rainfall, at any intensity, was less than the PM10-100 removed by water washing for all species (Fig. 2 (1) B and Fig. 2 (2) B). In addition, the PM10-100 removed by all forms of rainfall accounted for 13 ± 3% of removed TSP mass, and PM2.5 removed accounted for 84 ± 5% of total TSP removed when averaged across all species.

3.3.2. Species differences in PM removal rate

The species differences in different size PM removal rate by different rainfall intensities under total precipitation of 60 mm were shown in Fig. 2 (3). The TSP removal rate ranged from 49.3 ± 0.4% for *Pinus tabulaeformis* with a rainfall intensity of $72\ mm\ h^{-1}$, to 87.1 ± 1.2% for *Euonymus japonicus* with a rainfall intensity of $40\ mm\ h^{-1}$ (Fig. 2 (3) A). The PM10-100 removal rate ranged from 22.8 ± 1.4% for *Sophora japonica* with a rainfall intensity of $10\ mm\ h^{-1}$, to 71.5 ± 0.8% for *Pinus tabulaeformis* with a rainfall intensity of $10\ mm\ h^{-1}$ (Fig. 2 (3) B). The PM2.5-10 removal rate ranged from 40.4 ± 1.1% for *Sophora japonica* with a rainfall intensity of $10\ mm\ h^{-1}$, to 92.0 ± 0.3% for *Pinus tabulaeformis* with a rainfall intensity of $10\ mm\ h^{-1}$ (Fig. 2 (3) C). The PM2.5 removal rate ranged from 50.7 ± 0.8% for *Pinus tabulaeformis* with a rainfall intensity of $72\ mm\ h^{-1}$, to 91.3 ± 1.3% for *Euonymus japonicus* with a rainfall intensity of $40\ mm\ h^{-1}$ (Fig. 2 (3) D). The PM could not be completely removed from any of the species leaf surfaces that received a total rainfall of 60 mm when it was applied over either a long duration-low intensity pattern or a short duration-high intensity pattern. *Pinus tabulaeformis* had a significantly ($P < 0.05$) larger PM removal rate for the lowest rainfall intensity (i.e., $10\ mm\ h^{-1}$) compared to other species. The PM removal rate of *Euonymus japonicus* was significantly ($P < 0.05$) higher than the other four species for the moderate rainfall intensity of 40 and $50\ mm\ h^{-1}$. The PM removal rate of *Platanus acerifolia* was significantly ($P < 0.05$) higher than the other four species for the high rainfall intensity of $72\ mm\ h^{-1}$.

Table 2
Leaf coarseness of textural feature extraction of different species.

Tree species	Coarseness (Mean ± SE, N = 3)	
	Upper epidermis	Lower epidermis
Pinus tabuliformis	$11.14^{a} \pm 0.55$	$12.42^{a} \pm 0.80$
Platanus acerifolia	$15.56^{b} \pm 0.45$	$13.41^{b} \pm 0.25$
Euonymus japonicus	$13.59^{c} \pm 0.21$	$14.91^{c} \pm 0.64$
Populus tomentosa	$12.19^{d} \pm 0.35$	$16.38^{d} \pm 0.31$
Sophora japonica	$13.17^{c} \pm 0.65$	$12.87^{ab} \pm 0.42$

Different letters indicate significant differences ($P < 0.05$) between species mean values.

3.3.3. Patterns differences in PM removal rates for same species

Significant differences ($P < 0.05$) in PM removal amount and rate across different rainfall patterns for same species were showed in Fig. 2 (2)–(3). PM removal amount and rate for *Pinus tabulaeformis* by rainfall pattern of long duration-low intensity (i.e., 10 mm h^{-1} and 6h) were significantly ($P < 0.05$) more than that by short duration-high intensity rainfall (i.e.,72 mm h^{-1} and 5/6h). PM removal amount and rate for *Platanus acerifolia* by rainfall pattern of short duration-high intensity (i.e., 50 mm h^{-1} and 1.2h) were significantly ($P < 0.05$) more than that by long duration-low intensity rainfall (i.e.,10 mm h^{-1} and 6h). The PM removal amount and removal rate for *Euonymus japonicus* were largest with the rainfall pattern of 40 mm h^{-1} for 1.5h ($P < 0.05$).

3.4. Foliar PM removal rate and rainfall characteristics

3.4.1. Rainfall intensity

A correlation analysis showed that the PM removal rate was significantly related to rainfall intensity ($P < 0.01$) across all plant species. A significantly ($P < 0.01$) linear relationship between multiple size PM removal rate and rainfall intensity was observed by *Pinus tabulaeformis*, *Platanus acerifolia* and *Sophora japonica* (Fig. 3 (1), (2), (5) and Fig. 3 (4) C). For *Euonymus japonicus* and *Populus tomentosa*, PM removal rate first increased and then leveled off with rainfall intensity ($P < 0.01$) for all experiments except PM2.5-10 (Fig. 3 (3) and Fig. 3 (4) A, B, D).

3.4.2. Rainfall duration

Different size PM cumulative removal rate all exhibited an exponential loss with rainfall duration regardless of rainfall events ($P < 0.01$) (Fig. 4 and Fig. A2). The PM removal rate increased rapidly at the beginning of the simulated rainfall by all species regardless of rainfall intensities. However, there was a difference in the saturation time of PM cumulative removal rate across the different species. The low intensity rainfall needed more time to reach removal saturation compared to the high intensity rainfall.

3.4.3. Rainfall pattern

A correlation analysis showed that the PM removal rate was significantly related to rainfall pattern across all plant species (Fig. 5, $P < 0.05$). A significantly ($P < 0.0001$) linear decreasing relationship between different size PM removal rate and rainfall pattern was observed by *Pinus tabulaeformis* (Fig. 5 (1)). The PM removal rate increased first and then leveled off with rainfall pattern for the other species ($P < 0.05$) with a few exceptions (Fig. 5 (2)–(5)).

3.5. Foliar PM and leaf coarseness

3.5.1. Foliar PM accumulation (WPM) and leaf coarseness

The correlation analysis showed that foliar PM accumulation of different species was significantly ($P < 0.05$) related to leaf coarseness except for PM2.5 (Fig. 6 (1)). Among the five species, foliage accumulated TSP and PM10-100 on the upper epidermis were significantly and linearly correlated with increasing leaf coarseness. However, across species, there was not a significant correlation between PM accumulation and the lower epidermis coarseness except for PM2.5.

3.5.2. Foliar PM removal rate and leaf coarseness

The correlation analysis indicated that PM removal rate was correlated with the coarseness of upper epidermis ($P < 0.05$, Fig. 6 (2)–(6)). The PM removal rate corresponding to low rainfall intensities (10 mm h^{-1} and 20 mm h^{-1}) had a significant ($P < 0.05$) decreasing trend with the increasing coarseness when total precipitation of 60 mm (Fig. 6 (2), (3)). In contrast, the PM removal rate under high rainfall intensity (72 mm h^{-1}) had a significant ($P < 0.05$) increasing trend against the increasing coarseness (Fig. 6 (6)). There was a significant

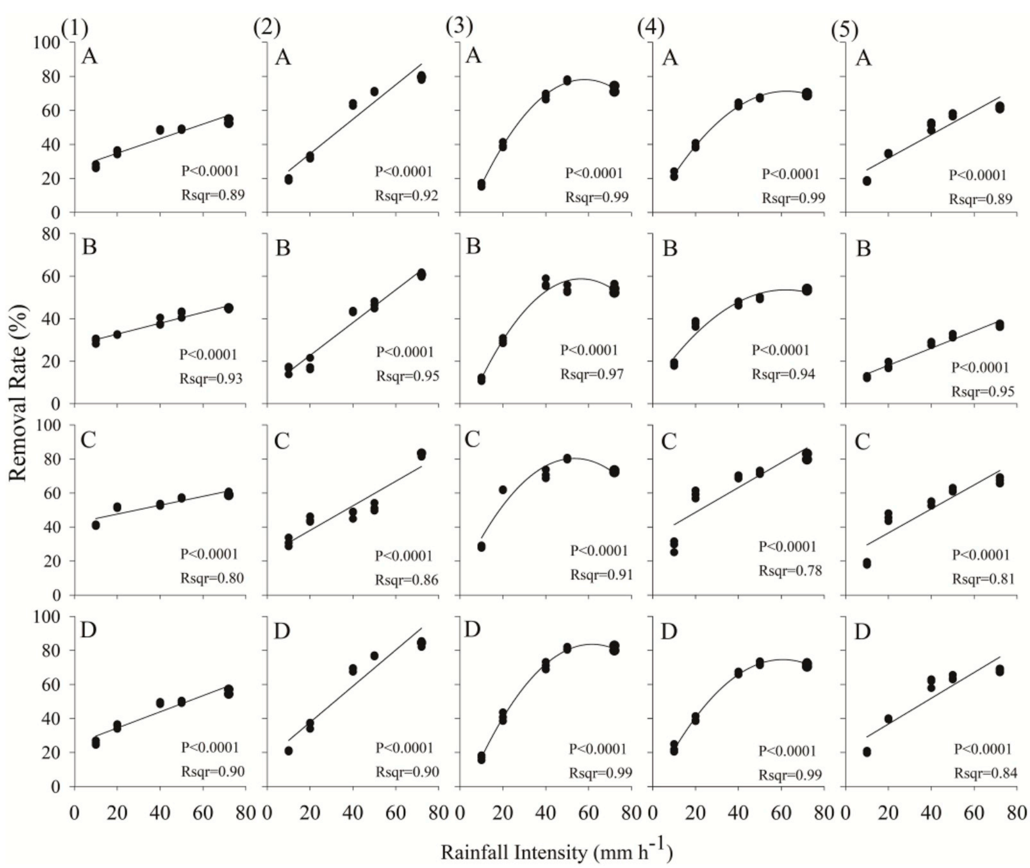

Fig. 3. Relationship between PM removal rate and rainfall intensity. Panels (1) to (5) represent different species: (1) *Pinus tabuliformis*, (2) *Platanus acerifolia*, (3) *Euonymus japonicus*, (4) *Populus tomentosa*, and (5) *Sophora japonica.* A to D in each panel represent different PM sizes: (A) TSP, (B) PM10-100, (C) PM2.5-10, and (D) PM2.5. Each intensity has three points representing the replicate samples.

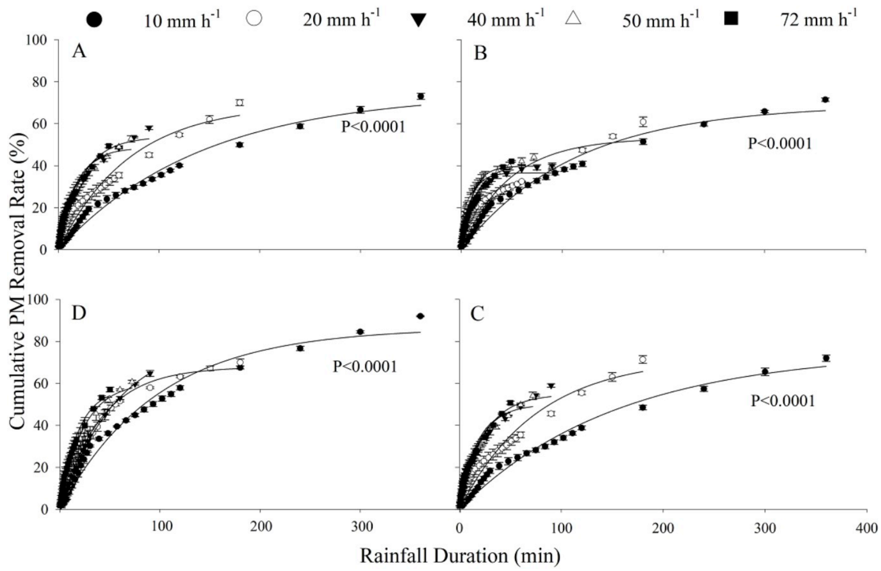

Fig. 4. Relationship between PM cumulative removal rate and rainfall duration for *Pinus tabuliformis* under different rainfall events. A to D represent different PM sizes: (A) TSP, (B) PM10-100, (C) PM2.5-10, and (D) PM2.5.

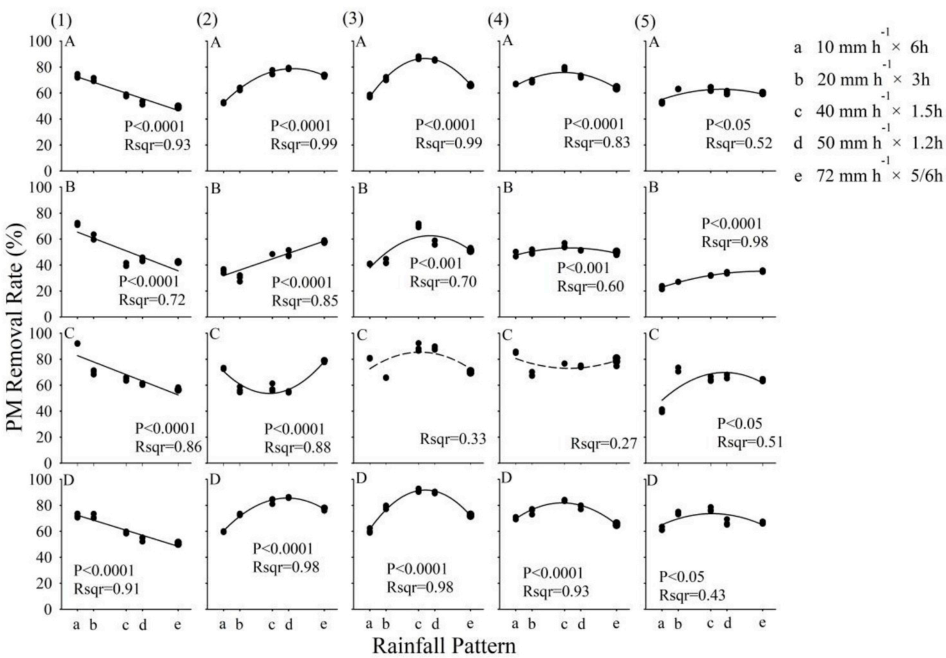

Fig. 5. Relationship between PM removal rate and rainfall pattern. Panels (1) to (5) represent different species: (1) *Pinus tabuliformis*, (2) *Platanus acerifolia*, (3) *Euonymus japonicus*, (4) *Populus tomentosa*, and (5) *Sophora japonica*. A to D in each panel represent different PM sizes: (A) TSP, (B) PM10-100, (C) PM2.5-10, and (D) PM2.5. Each intensity has three points representing the replicate samples.

($P < 0.05$) increase in the PM removal rate until the leaf coarseness reached 13 and then PM removal decreased under the rainfall intensities of 40 and 50 mm h^{-1} (Fig. 6 (4), (5)). The PM removal rate of *Sophora japonica* was lower than that of *Euonymus japonicus* (Fig. 6 (2)–(6): red plots) even though upper epidermis coarseness values of the two were similar (Table 2). No relationship was found between the foliar PM removal rates and the lower epidermis coarseness.

4. Discussion

4.1. RPM and WPM

Rainfall and water washing could not completely remove the PM from the leaf surfaces. However, more foliar TSP was removed by rainfall (RPM) than that by water washing (WPM), which indicated conventional washing method cannot accurately assess the PM retention capacity of plants. On average, 29%–46% of the PM remained on the leaves when leaves were water and brush cleaned (Liu et al., 2018). More PM can be washed off by rainfall because the continuous rainfall can lead to a longer water/PM contact time (compared to WPM), and the high kinetic energy of raindrops may remove more particulates by splashing/impacting the leaf surface (Neinhuis and Barthlott, 1998; Ouyang et al., 2015; H. Wang et al., 2015a). Additionally, the removal ratio of PM10–100 and PM2.5-10 to TSP was different than the ratio retained on the leaf surfaces, which suggested that rainfall had changed the PM morphology as soluble constituents within the PM were dissolved during the rainfall event. Aqueous insoluble particulates represent only a small proportion of the ultra-fine PM (Beckett et al., 2000a,b). Much of the ultra-fine fraction exists as an aerosol, containing soluble components, such as sulfate, nitrate, and chloride, that decrease in size as they dissolve in water (Freer-smith et al., 2005). Additionally, large particulates may be disaggregated into

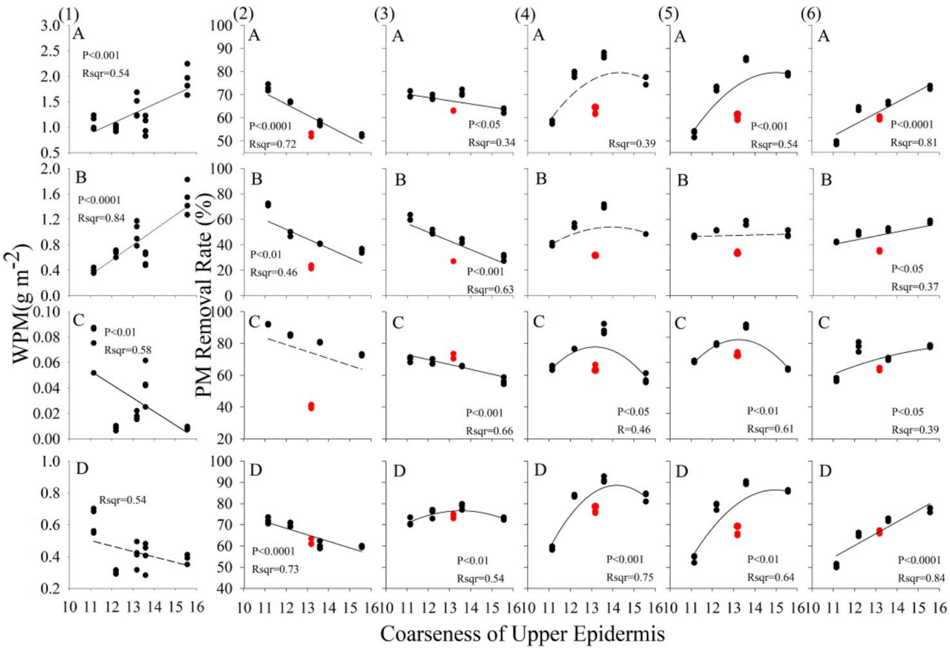

Fig. 6. Relationship between foliar PM and leaf coarseness. Panels (1) WPM, (2)–(6) representing PM removal rate for rainfall intensities of: (2) 10 mm h^{-1}, (3) 20 mm h^{-1}, (4) 40 mm h^{-1}, (5) 50 mm h^{-1} and (6) 72 mm h^{-1}. A to D in each panel represent different PM sizes: (A) TSP, (B) PM10-100, (C) PM2.5-10, and (D) PM2.5. Each leaf coarseness has three points representing replicate samples and the curve was fitted using only black plots (representing the other four plants besides *Sophora japonica*).

some smaller particulates by rainfall. Previous studies revealed that some fine particulates, associated with attractive and reactive properties, can agglomerate into larger particulates in the air, which can fall onto leaf surfaces via sedimentation (Beckett et al., 1998; Hofman et al., 2016; Petroff et al., 2008b; Speak et al., 2012; Terzaghi et al., 2013). These two processes can lead to a variation in PM size before and after the precipitation.

4.2. Response of PM removal rate to the rainfall characteristics

4.2.1. Rainfall intensity

A significant increasing trend of PM removal rate with increased rainfall intensity (Fig. 3) indicated that the increasing diameter and kinetic energy of the simulated raindrops lead to larger PM removal rate (Chen et al., 2017; Xu et al., 2017). The kinetic energy of rainfall is the predominant factor in the washing process (Ayub et al., 2012). However, the varying PM removal rate as a function of rainfall intensity between different species (Fig. 3) could be due to the different leaf microstructural traits described in 4.3.2. Additionally, leaf macrostructural traits such as leaf area, leaf shape and petiole length affecting leaf movement when different raindrop intensities striking the leaf surface are also important factors influencing the PM removal rate (Leonard et al., 2016).

4.2.2. Rainfall duration

The relationship between the PM cumulative removal rate and rainfall duration (Fig. 4, Fig. A2) indicated that PM removal rates were only partially controlled by rainfall duration (Xu et al., 2017). The amount of PM released by foliage during a rainfall event was largely a function of the amount of PM retained by the leaf. The number of days of accumulation before the rainfall and maximum PM retention capacity of the plants were strongly dependent on the plants growing condition and environment (Dzierżanowski et al., 2011).

4.2.3. Rainfall patterns

The leaves had significant ($P < 0.05$) differences in PM removal amount and rate for different rainfall patterns. The response of the PM removal rate to rainfall pattern was related with leaf coarseness (3.5.2). The intensity of the rainfall to the extent that it is resistant to leaf coarseness, coupled with a longer rainfall duration, may result in greater removal rate.

4.3. Leaf traits and foliar PM

4.3.1. Leaf traits and foliar PM accumulation (WPM)

Some species-specific features of leaves can enhance PM air filtration processes (Perini et al., 2017; Smith and Staskawlcz, 1977). Differences in the micro morphology of the leaf surfaces indicated by SEM images and leaf coarseness (Fig. A1 and Table 2) can explain the species differences in PM accumulation. Among the five species, *Platanus acerifolia* retained the largest amount of TSP and PM10-100 (Fig. 2 (1) A, B) due to the high leaf coarseness (Table 2), indicating that complex leaf microstructures can lead to capture of more particulates on leaf surfaces (Fig. 6 (1) A, B). Surface roughness can influence the transition from a laminar to a turbulent airflow (Vadlamani et al., 2017). Therefore, the deposition velocity can be an order of magnitude larger across a rough surface compared to a smooth surface (Fowler et al., 1989). *Sophora japonica* ranked second in TSP, PM10–100 and PM2.5 accumulations (Fig. 2 (1) A, B, D), potentially due to the presence of plants leaf hairs (Fig. A1 (e1), (e2)). This leaf effect would be consistent with previous studies showing that PM accumulation is greater on leaves with hairs because leaf hairs not only increase the surface area that can intercept more PM, but the hairs also make PM less likely to be dislodged when leaves are moving (Kardel et al., 2011; Mitchell et al., 2010; Tallis et al., 2011). *Pinus tabulaeformis* had the most accumulated PM2.5–10 and PM2.5 (Fig. 2 (1) C, D) but had the least coarse surface among the five species (Table 2). Coniferous leaves not only have a thicker layer of epicuticular wax (Beckett et al., 2000a,b), but also they also have many shallow stripes less than 10 μm (Fig. A1 (a1) (a2)) that can capture and store more small particulates. The ridges (at a scale of 1–2 μm) on the leaf surfaces were efficient in accumulating PM, particularly PM2.5 (Wang et al., 2015). The parallel grooves on the leaves can trap particulates and prevent their resuspension (Speak et al., 2012). In addition, smaller leaves and more complex shoot structures have larger Stokes number and thus higher inertial impaction efficiency (Beckett et al., 2000a,b; Chen et al., 2017; Freer-smith et al., 2005; Price et al., 2017). For these reasons, conifers capture more fine PM compared to broadleaved trees. However, there was not a significant relationship between the lower epidermis coarseness and PM capture, which indicated that upper epidermis is the major surface to capture PM (Wild et al., 2006).

4.3.2. Leaf traits and foliar PM removal rate

For smooth surfaces, long duration-low intensity rainfall created conditions for the highest rates of PM removal (Fig. 6 (2), (3)), while for rough surfaces, short duration-high intensity rainfall created conditions for the highest rates of PM removal under the same amount of total precipitation (Fig. 6 (6)). Differences in leaf coarseness associated with microstructure can lead to difference in leaf-wettability. Increased microstructural complexity can significantly enlarge the critical pressure (i.e., the maximum pressure for sustainable Cassie-Baxter wetting state) (Zheng et al., 2005). The Cassie-Baxter wetting mode may collapse when the hydraulic pressure (i.e., water-air interfacial tension) is lower than the critical pressure, if the microstructural is relatively complex (Zheng et al., 2005). Therefore, leaf microstructural features like wax layer, hairs and other protrusions can create different contact angles (θ) between a water droplet and different leaf surfaces (Wang et al., 2015b). These factors create different water-repellent characteristics between species (Bussonnière et al., 2017). As a consequence, rough leaf surfaces have wetting property, which can increase the contact time between rainwater and the leaf surface to remove PM from leaves when the rainfall intensity is high enough (Bussonnière et al., 2017) (Fig. 6 (6)). However, for smooth surfaces, raindrops with low intensity impact can quickly move across the water-repellent leaf surface and take away particulates (Neinhuis and Barthlott, 1998) and have a long time to repeatedly wash the leaf (Fig. 6 (2), (3)). Under conditions of moderate rainfall intensity, moderately coarse leaves were the most effective in removing PM from the leaf surface (Fig. 6 (4), (5)). It is worth noting that most PM removal rates of *Sophora japonica* were lower than those of *Euonymus japonicus* even though the two species had approximately the same leaf coarseness (Fig. 6 (2)–(6)). This difference may be that there are hairs on *Sophora japonica* upper epidermis but not on *Euonymus japonicas* (Fig. A1 (e1), (e2)). Other studies also found that the leaves with hairs were more water-repellent, especially where hair density was greater than 25 mm^{-2} (Leonard et al., 2016; Wang et al., 2015b).

5. Conclusion

Understanding the process of precipitation washing PM off from the leaf is necessary to accurately evaluate the long-term potential air purification ability and efficiency of the urban trees and forests. Foliar PM removal amount and rate by rainfall are governed by multiple biological (e.g., species and leaf micromorphology), environmental (e.g., rainfall intensity, duration, and patterns), physical factors (e.g., leaf wettability, rainfall kinetic energy) and by pre-rain PM accumulation (e.g., saturated and unsaturated). We found that significant differences ($P < 0.05$) in PM removal amounts and rates not only between different species within the same rainfall pattern, but also between different rainfall patterns for same species. PM removal rates were significantly correlated with rainfall intensity ($P < 0.01$) and was exponentially decay related to rainfall duration ($P < 0.01$). For smooth leaf surfaces, long duration-low intensity rainfall could increase the PM removal rate while for rough leaf surfaces, short duration-high intensity rainfall could achieve a larger removal rate for the same amount of rainfall. Additionally, more PM was removed by rainfall than that by water washing. The findings from this study have implications related to planting decisions for urban air phytoremediation.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.04.010.

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