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A comparative study of foliar particulate matter wash-off from plants under natural and simulated rain conditions

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Urban greening can reduce concentrations of particulate matter (PM). However, most of the PM is deposited temporarily on foliage and can be removed by precipitation. Due to the unpredictability and randomness of natural rain and the complexity of environmental conditions, the effect of rainfall on PM removal from foliage is usually examined using simulated rain. The aim of this study was to compare the effects of natural and simulated rain on washing off PM deposited on the foliage of seventeen plant species (evergreen/deciduous trees and shrubs, herbaceous plants). Regardless of the PM fraction studied, simulated rain washed off a larger fraction of the PM than natural rain. A novel finding is the differential role played by various plant characteristics in determining the effectiveness of PM wash off under natural and simulated rain conditions, with their influence being more pronounced under natural rainfall. The type of rainfall and PM size fraction influenced the effectiveness of the PM wash-off from different plant groups. Natural rain was more effective at removing PM from evergreen trees, while simulated rainfall was more effective at removing PM from deciduous shrubs and herbaceous plants. The results obtained from this study suggest that simulated rain does not reflect the effects of natural precipitation on the processes removing PM from leaves, and therefore should not be used to explain phenomena that occur in realistic conditions.

Keywords Air phytoremediation, Air pollutants, Precipitation, Natural rain, Simulated rain

Particulate matter (PM, solid and liquid particles with a diameter between 0.001 and 100 μm) has become one of the world's most pressing environmental issues[1]. PM pollution has adverse effects on the economy, and people's health and living comfort[2]. Despite making tremendous progress in recent years, China still has some of the most polluted air in the world[3]. The air in Wuhan is among the most polluted of Chinese cities, both in terms of high concentrations and the number of days exceeding air quality standards[4–6].

Air biofiltration efficiency depends on the plant community structure and individual plant characteristics, especially in relation to leaves[7–10]. Unfortunately, as a result of long, uninterrupted accumulation, the leaf may become saturated with PM[10–12]. Once saturation is reached, typically after a few weeks, with a significant decrease in PM accumulation after 10 days, the positive impact of plants on air quality is smaller[10–12]. PM accumulated on plants includes easily removable, difficult to remove and completely non-removable particles[13]. A small fraction of the deposited PM remains on the leaf surface/wax permanently or migrates inside the leaf[7,14]. Most of the PM accumulates on leaves temporarily and can be removed by environmental factors, mainly precipitation[12,15–21] and/or wind[12,16]. As much as 76% of previously accumulated PM may be resuspended[22].

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PM removal from foliage restores the PM accumulation capacity and prepares the plant for further air biofiltration[7,16,17,20,23]. PM washed off from the plant canopy and transported in throughfall can be re-neutralised by plants growing underneath, or by the soil if it is not covered by a paved surface[12,22]. Soil deposition of PM, together with PM permanently retained in the plant, is the net PM removal[22–24]. Therefore, it is important to understand the dynamics of PM accumulation, retention, wash-off and resuspension from foliage. These processes occur simultaneously and determine the effectiveness of the greenery in air biofiltration[11].

The amount of PM washed off leaves by precipitation depends on the characteristics of the rain, the morphometric characteristics of the plant, and the morphology of the leaf, similar to those that shape the ability of plants to accumulate PM from the air[11,12,21]. It is generally accepted that PM deposited on plants that have a high density of leaves of complex morphology is more difficult to wash off[9,10,12,16–19,23,25–27]. Variations in leaf surface microstructures not only affect foliage roughness, but can also lead to different contact angles between water droplets and leaf surfaces, thus affecting leaf water repellency[12,16,23–25]. Compared with smooth surfaces, rough surfaces reduce raindrop kinetic energy[12,17]. These differences result in the foliage having different hydrophobic properties, which is one of the main factors that increase or decrease the resistance of PM to being washed off[7,12,20,23,24,28].

Rainfall may also contribute to increased PM deposition on plants[12,16,22,29,30]. Precipitation washes pollutants from the air, and PM carried by raindrops can lead to the wet deposition on foliage[27,31–33]. Wet deposition may be responsible for the deposition of more than 50% of PM on plants[31,34]. Increased accumulation of PM by foliage may also be caused by higher air and leaf humidity, which are typical phenomena after rainfall and increase the stickiness of the foliage[33,35].

The literature is replete with studies evaluating the phenomenon of PM wash-off from plants by rain. Due to the unpredictability and randomness of natural rainfall, and the complexity of the effects of environmental conditions, the majority of these studies are laboratory experiments in which rainfall is simulated. The results of such experiments are then used as a basis for explaining processes in realistic conditions. Surprisingly, there has been no comparison of the effects of natural and simulated precipitation. The aim of this study was to compare the effects of natural and simulated precipitation on the washing-off of PM deposited on the foliage of several species (evergreen trees and shrubs, deciduous trees and shrubs, herbaceous plants). The study sought to answer the following questions: (i) Is the amount of PM washed off by natural and simulated rainfall the same? (ii) Are the morphometric and morphological characteristics of plants and leaves that influence the amount of PM washed off by natural and simulated precipitation similar? (iii) Is the amount of PM washed off by natural and simulated precipitation the same from plants in the same group and the same species?

Results

Morphometric and morphogenic assessment of the plants

The plants differed in their morphometric parameters (Table 1). The trees were tallest (*M. glyptostroboides*, *P. acerifolia*, *P. stenoptera*) and had the largest canopy diameter (*P. acerifolia*, *K. paniculata*). *Metasequoia glyptostroboides*, *K. paniculata* and *P. stenoptera* had the greatest canopy height. The largest canopy density (91–92%) was found for *L. chinense*, *M. grandiflora*, and *O. fragrans*, and the lowest for *M. glyptostroboides* (73%) and *P. stenoptera* (77%). *Metasequoia glyptostroboides* and *P. stenoptera* were also characterised by the lowest LAI. The highest LAI values were recorded for *M. grandiflora*, *O. fragrans*, *P. serrulata*, *K. paniculata* and *P. acerifolia*. *Magnolia grandiflora* and *P. acerifolia* had the largest leaves, while *M. glyptostroboides*, *L. chinense*, *C. dactylon* and *O. japonicus* had the smallest leaves. The leaves of *L. formosana* and *P. acerifolia* had the longest petioles, while the shortest petioles were found in *K. paniculata*, *P. stenoptera*, *L. chinense*, *R. simsii* and *L. indica* (Table 1).

Acer palmatum, *L. formosana* and *K. paniculata* were characterised by the highest stomatal density, *C. camphora*, *L. lucidum*, and *P. serrulata* by the longest stomata, and *L. formosana* and *M. glyptostroboides* by the widest midrib. A high density of hairs on the foliage was found in *M. grandiflora*, while long hairs were recorded in *R. simsii* and *L. chinense* (Fig. 1; Table 1).

Comparison of PM wash-off by natural and simulated rain

Regardless of the PM fraction (PM_{10}, $PM_{2.5\text{-}10}$, $PM_{2.5}$), simulated rain washed off a larger amount of the PM than natural precipitation (Table 2). Simulated precipitation washed off more $PM_{2.5\text{-}10}$ than $PM_{2.5}$. The variances were significantly different for all the PM fraction and were higher for natural rain (Table 2).

Plant traits affecting PM wash-off by natural and simulated rain

The morphometric and morphological characteristics of the plants and leaves had a greater influence on the amount of PM washed off by natural precipitation, but this varied depending on the PM size fraction (Table 3). The wash-off of PM_{10} and $PM_{2.5\text{-}10}$ was significantly positively correlated with plant height, LAI, leaf area, petiole length and leatheriness (leathery leaves, unlike those with a more papery structure, accumulated PM more efficiently), but negatively correlated with leaf texture (rough leaves structure, unlike the smooth one, did not facilitate more efficient PM accumulation). $PM_{2.5\text{-}10}$ was also positively correlated with canopy diameter. A negative correlation with $PM_{2.5\text{-}10}$ was found for crown height and leaf serrations. $PM_{2.5}$ wash-off was only correlated with foliar microstructures: positively with stomatal density and hair density and length, and negatively with midrib width. In the case of simulated precipitation, significant correlations were only found for $PM_{2.5\text{-}10}$ and petiole length (positive) and $PM_{2.5}$ and leaf area (negative) (Table 3).

Differences between plants in PM wash-off by natural and simulated rain

The natural rainfall facilitates the most efficient removal of PM from evergreen trees (PM_{10}, $PM_{2.5\text{-}10}$) and evergreen shrubs ($PM_{2.5}$), although this effect was not significant for $PM_{2.5}$ (Fig. 2). Analysis revealed an increase in PM deposition after natural rain on herbaceous plants and deciduous shrubs, except for $PM_{2.5}$ on deciduous

Species	Type of plant	Height (m)	Canopy diameter (m)	Canopy height (m)	Canopy density (%)	LAI (m^2m^2)	Leaf area (cm^2)	Petiole length (mm)	Midrib width (µm)	Stomata density ($number.mm^2$)	Stomata length (µm)	Hair density ($number.mm^2$)	Hair length (µm)	Wax ($µg.cm^2$)	Leaf texture	Leaf leatheriness	Serrated edges	Leaf shape
Cinnamomum camphora	Evergreen trees	7.45 EFG* (±1.79)	6.70 DE (±2.24)	5.05 AB (±1.45)	89 CD (±0.02)	2.01 D (±0.21)	16.1 BC (±2.51)	21.1 DE (±7.90)	23.7 A (±3.27)	254.1 CD (±27.8)	22.0 L (±0.90)	-	-	125.5 DE (±43.5)	smooth	leather	no	oval
Ligustrum lucidum		8.00 FG (±0.00)	7.50 E (±0.00)	8.00 BC (±0.00)	82 B (±0.02)	1.63 BC (±0.03)	25.8 C (±6.16)	14.3 CD (±1.25)	15.2 A (±4.09)	246.8 CD (±16.8)	18.9 K (±0.63)	-	-	137.7 DE (±35.1)	smooth	leather	no	oval
Magnolia grandiflora		9.50 G (±1.68)	6.95 E (±0.42)	6.65 AB (±1.90)	91 D (±0.07)	2.48 E (±0.35)	141.0 E (±16.1)	26.4 E (±4.57)	6.25 A (±0.88)	213.0 BC (±33.4)	6.58 B (±0.27)	357.0 C (±47.8)	321.5 B (±113.5)	32.3 A (±9.75)	smooth	leather	no	oval
Osmanthus fragrans		4.90 CD (±1.12)	4.91 CD (±1.25)	4.80 A (±1.02)	92 D (±0.02)	2.37 E (±0.16)	27.0 C (±4.39)	5.29 AB (±1.31)	9.69 A (±0.81)	515.4 H (±83.4)	7.19 BC (±0.81)	-	-	126.0 DE (±25.2)	smooth	leather	no	oval
Photinia serrulata		6.80 DEF (±0.89)	5.90 CDE (±1.14)	6.02 AB (±0.80)	89 CD (±0.03)	2.37 E (±0.25)	51.6 D (±6.17)	23.8 E (±1.73)	14.1 A (±2.63)	261.3 DEF (±47.4)	18.8 K (±0.18)	-	-	141.6 E (±42.3)	smooth	leather	yes	oval
Liquidambar formosana	Deciduous trees	7.25 DEFG (±0.96)	7.10 E (±1.60)	4.67 A (±1.20)	88 CD (±0.02)	1.97 D (±0.22)	50.5 D (±5.90)	60.4 G (±5.83)	165.6 D (±9.00)	833.3 I (±86.1)	7.66 BCD (±0.88)	-	-	51.1 AB (±15.1)	medium	medium	no	palmate
Koelreuteria paniculata		14.0 H (±2.71)	11.3 G (±1.59)	12.0 D (±3.37)	86 BC (±0.04)	2.29 E (±0.14)	23.7 C (±4.00)	1.79 A (±0.63)	7.03 A (±1.93)	820.2 I (±49.6)	7.19 BC (±1.09)	-	-	108.6 DE (±27.4)	medium	paper	yes	oval
Metasequoia glyptostroboides		18.5 I (±4.36)	7.87 EF (±1.94)	15.8 E (±4.74)	73 A (±0.02)	1.24 A (±0.19)	0.22 A (±0.02)	-	114.2 C (±24.3)	158.3 B (±31.9)	17.3 J (±0.65)	-	-	138.5 E (±40.2)	medium	paper	no	linear
Platanus acerifolia		14.7 H (±1.25)	13.4 H (±2.92)	10.9 CD (±0.17)	90 CD (±0.01)	2.29 E (±0.14)	140.1 E (±31.4)	64.0 G (±15.8)	13.1 A (±2.16)	333.9 FG (±37.5)	8.05 CDE (±1.31)	-	-	100.5 CD (±21.2)	rough	paper	no	palmate
Pterocarya stenoptera		16.0 HI (±3.61)	9.81 FG (±1.60)	12.5 D (±2.50)	77 A (±0.03)	1.41 AB (±0.17)	29.0 C (±5.43)	3.92 A (±3.85)	62.5 B (±23.3)	241.7 CD (±56.9)	8.75 DEF (±0.43)	-	-	104.7 CDE (±22.1)	rough	medium	yes	oval
Loropetalum chinense	Evergreen shrubs	1.35 AB (±0.38)	1.62 A (±0.33)	-	91 D (±0.04)	1.97 D (±0.13)	2.86 A (±0.24)	2.71 A (±0.34)	17.7 A (±5.01)	515.4 H (±64.4)	9.41 FG (±0.39)	6.39 A (±4.08)	467.2 B (±97.5)	52.9 AB (±9.46)	rough	medium	yes	oval
Pittosporum tobira		1.60 AB (±0.41)	2.00 A (±0.57)	-	90 CD (±0.02)	1.91 D (±0.14)	16.2 BC (±2.87)	12.9 BC (±1.67)	14.1 A (±2.63)	312.1 EFG (±43.5)	13.3 HI (±0.69)	87.1 B (±41.1)	60.3 A (±5.58)	57.3 AB (±13.7)	smooth	leather	no	oval
Rhododendron simsii		1.17 AB (±0.05)	1.50 A (±0.00)	-	87 BCD (±0.02)	1.97 D (±0.15)	3.68 AB (±0.83)	5.04 A (±0.91)	6.56 A (±1.30)	355.7 G (±36.5)	9.11 EFG (±0.41)	1.74 A (±0.67)	750.0 C (±135.0)	200.6 F (±32.2)	medium	medium	no	oval
Acer palmatum	Deciduous shrubs	2.92 BC (±0.74)	4.15 BC (±0.62)	-	86 BCD (±0.07)	1.60 BC (±0.17)	8.30 AB (±2.37)	45.0 F (±4.41)	19.1 A (±6.95)	1125.0 J (±95.7)	4.83 A (±1.95)	-	-	51.5 AB (±16.7)	rough	paper	yes	palmate
Lagerstroemia indica		5.25 CDE (±0.29)	2.30 AB (±0.36)	-	85 BC (±0.03)	1.77 CD (±0.13)	8.34 AB (±1.44)	1.00 A (±0.00)	9.53 A (±1.18)	283.1 DEFG (±27.8)	10.1 G (±1.70)	-	-	69.3 ABC (±24.8)	medium	paper	no	oval
Cynodon dactylon	Grasses	0.20 A (±0.00)	-	-	-	-	0.54 A (±0.04)	-	21.2 A (±4.65)	159.7 B (±37.5)	13.1 H (±0.24)	-	-	55.3 AB (±15.9)	medium	paper	no	linear
Ophiopogon japonicus		0.20 A (±0.00)	-	-	-	-	1.42 A (±0.21)	-	93.9 C (±49.5)	72.59 A (±50.3)	14.5 I (±0.65)	-	-	69.9 BC (±13.2)	medium	leather	no	linear

Table 1. Morphometric and morphological characteristics of the examined species. Data are means ± SE, $n = 4$. * Different uppercase letters indicate a significant difference between examined morphometric and morphological characteristics at $P < 0.05$ by t-test.

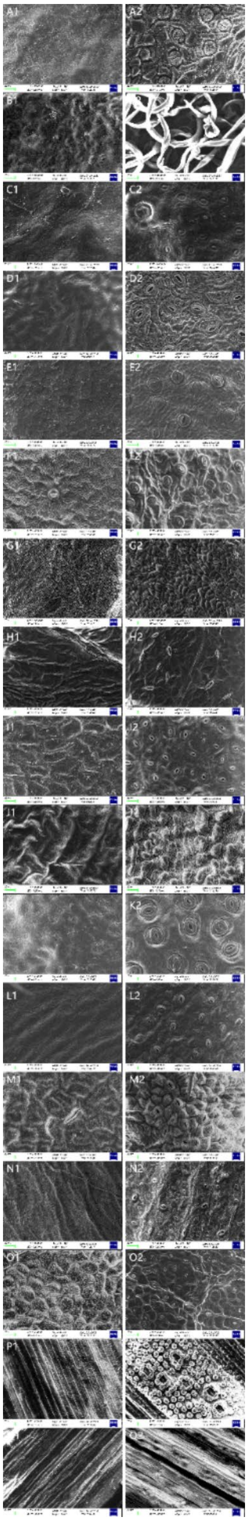

Fig. 1. Scanning electron microscope images of the leaf microstructure surface (**A** – *C. camphora*, **B** – *M. grandiflora*, **C** – *O. fragrans*, **D** – *L. lucidum*, **E** – *P. serrulata*, **F** – *P. acerifolia*, **G** – *K. paniculata*, **H** – *P. stenoptera*, **I** – *L. formosana*, **J** – *M. glyptostroboides*, **K** – *P. tobira*, **L** – *R. simsii*, **M** – *L. chinense*; **N** – *L. indica*, **O** – *A. palmatum*, **P** – *O. japonicus*, **R** – *C. dactylon*; 1 – adaxis, 2 - abaxis).

shrubs. Simulated precipitation yielded the highest, but insignificant, removal rates of PM_{10} and $PM_{2.5\text{-}10}$ from evergreen trees and, in a contrasting pattern to natural rainfall, from deciduous shrubs and herbaceous plants. $PM_{2.5}$ was washed off most efficiently by simulated rainfall, but not significantly so, from both shrub groups and herbaceous plants. In the case of simulated precipitation, there was no increase in PM on the foliage as

Pollutant	Percentage washed off					
	Average			Standard deviations		
	Natural rain	Simulated rain	*p*	Natural rain	Simulated rain	*p*
PM_{10}	19.93	24.11	0.35	35.81	6.67	< 0.001
$PM_{2.5-10}$	16.27 A*	26.20 B	0.10	48.93	8.01	< 0.001
$PM_{2.5}$	14.76 A	18.07 A	0.56	46.06	9.16	< 0.001

Table 2. Average and standard deviation values for the percentage of PM washed off by natural and simulated rain and *p*-values of t and Levene's tests at significance level α = 0.05. Data are means, *n* = 68. * Different uppercase letters indicate significant differences between different PM fractions or trace elements washed off by natural or simulated rain at *P* < 0.05 by t-test.

				Parameter	Particulate matter					
					PM_{10}		$PM_{2.5-10}$		$PM_{2.5}$	
					NR	SR	NR	SR	NR	SR
-				Height	**0.31**	-	**0.42**	-	-0.09	[illegible]
-				Canopy diameter*	0.17	-	0.11	-	[illegible]	[illegible]
[illegible]				Canopy density*	0.16	[illegible]	-	[illegible]	[illegible]	[illegible]
0.05	-			Crown height#		-0.28	-	**-0.31**	-	[illegible]
-	0.05	-		LAI*			**0.29**	-	**0.32**	
0.15	-0.14	**-0.28**		Leaf area			**0.26**	0.06	**0.40**	
0.27	-0.10	-0.07		Petiole length			**0.27**	0.24	**0.40**	
-0.09	**-0.26**	0.10		Midrib width			-0.22	-0.07	-0.13	
0.10	**0.37**	0.09		Stomata density			0.08	0.07	0.05	
-0.23	-0.23	0.19		Stomata length			-0.08	-0.17	-0.01	
-0.16	**0.29**	0.04		Hair density			0.17	-0.10	0.08	
-0.18	**0.32**	0.01		Hair length			0.09	-0.12	-0.03	
-0.06	-0.05	-0.20		Wax			0.05	-0.12	0.11	
0.07	0.03	0.00		Leaf texture			**0.50**	0.10	**0.55**	
0.00	-0.03	-0.15		Leaf leatheriness			**0.25**	-0.01	[illegible]	
0.10	0.20	0.09		Serrated edges			0.14	-0.12	[illegible]	
-0.07	-0.16	-0.08	0.11	Leaf shape			-0.06	-0.11		

Table 3. Spearman's correlation coefficients between the characteristics of the studied plants and the percentage of PM washed off by natural (NR) and simulated rain (SR). All significant correlations are in bold. Negative correlations are indicated by blue cells and positive ones by red cells. *only for shrubs and trees. #only for trees.

its depositions were consistently decreased. Comparative analyses between natural and simulated rainfall showed that simulated rain was significantly more effective at removing PM_{10} and $PM_{2.5-10}$ from deciduous shrubs and herbaceous plants, while natural precipitation washed off more PM from evergreen trees. Large, but not significant, differences were also found for evergreen shrubs where simulated precipitation was more effective at PM wash-off. The differences in the removal of $PM_{2.5}$ between natural and simulated precipitation were not statistically significant; notable variations were observed among deciduous trees, evergreen shrubs and herbaceous plants (Fig. 2).

The studied species exhibited significant variability in the removal of PM_{10} and $PM_{2.5-10}$ through natural precipitation (Table 4). *Metasequoia grandiflora* and *O. fragrans* demonstrated the greatest efficacy at washing off PM_{10} due to natural rainfall, with removal rates of 65.8% and 51.0% respectively. Conversely, *P. stenoptera* showed an increase in PM_{10} accumulation after natural rainfall quantified at 28.7%. When considering $PM_{2.5-10}$, *M. grandiflora* again led in removal efficiency at 77.0%, while *P. stenoptera* exhibited a 47.0% increase in $PM_{2.5-10}$ deposition. There were no significant differences between species in the amount of $PM_{2.5}$ washed off by natural rainfall. Simulated rainfall showed a uniform effect across all PM size fractions for each species (18.4–32.2% for PM_{10}; 18.8–34.6% for $PM_{2.5-10}$; 9.1–27.5% for $PM_{2.5}$). Significant differences in the amounts of PM washed off by natural and simulated precipitation were found for the majority of species: *C. camphora* (higher PM_{10} and $PM_{2.5-10}$ wash off after natural rain), *M. grandiflora* (higher PM_{10} and $PM_{2.5-10}$ wash off after natural rain), *O. fragrans* (higher $PM_{2.5-10}$ wash off after natural rain), *L. formosana* (higher PM_{10} and $PM_{2.5-10}$ wash off after natural rain), *K. paniculata* (higher $PM_{2.5}$ wash off after natural rain), *M. glyptostroboides* (higher PM_{10} and $PM_{2.5-10}$ wash off after natural rain), *L. chinense* (higher $PM_{2.5}$ wash off after natural rain), *R. simsii* (higher $PM_{2.5}$ wash off after natural rain), *A. palmatum* (higher $PM_{2.5-10}$ wash off after simulated rain) and *O. japonicus* (higher PM_{10} and $PM_{2.5-10}$ wash off after simulated rain) (Table 4). Actual values of PM (μg.cm^2) deposition on plant foliage and the amount of pollutants washed off by rain (natural/simulated) are presented in supplementary materials (Table S1).

Figure 3 illustrates the impact of the type of plants on the percentage of pollutants removed by natural and simulated rain. PC1 and PC2 collectively explained approximately 73% of the total variance in the dataset. Variables such as SR_$PM_{2.5-10}$ and SR_PM_{10} were correlated and had high values for *A. palmatum* and *C. dactylon* plants. Additionally, NR_PM_{10}, SR_$PM_{2.5-10}$ and SR_$PM_{2.5}$ demonstrated a correlation, with elevated

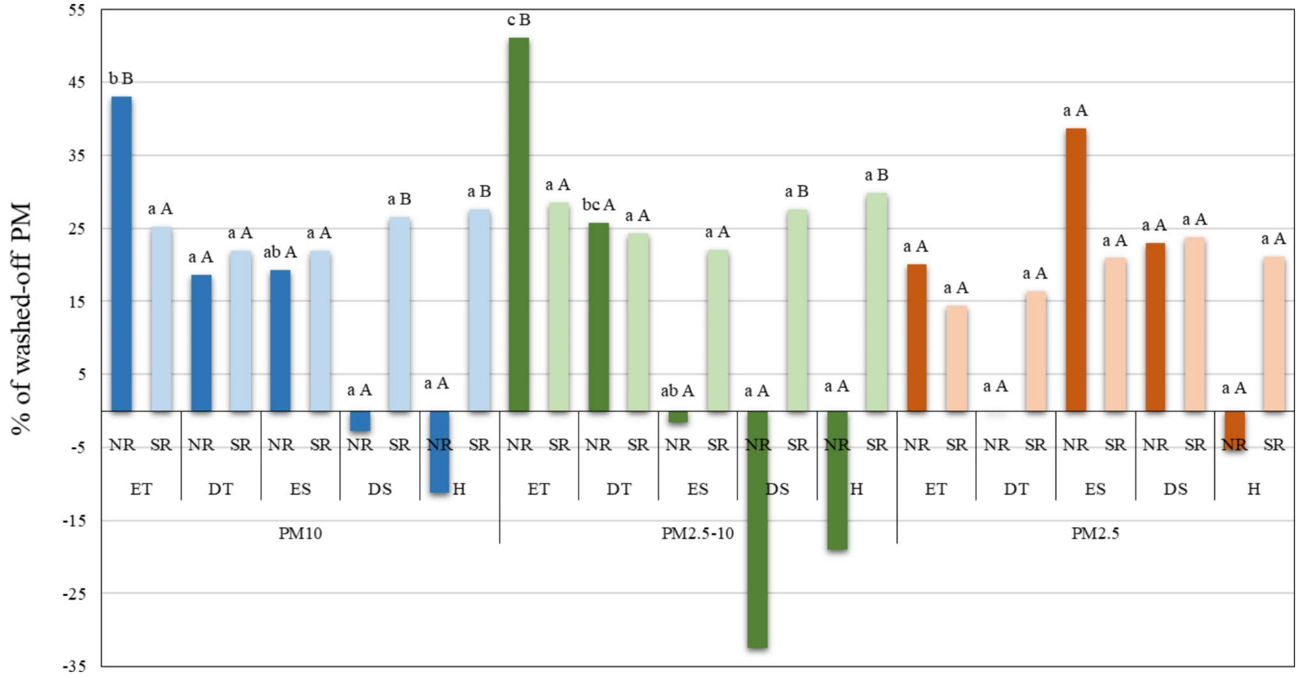

Fig. 2. Means for the percentage of washed-off PM attributed to the different types of plants studied (ET – evergreen trees; DT – deciduous trees; ES - evergreen shrubs; DS – deciduous shrubs; H – herbaceous plants), as well as natural and simulated rain (NR and SR, respectively). The same small letters for PM size fractions and TE indicate homogeneous groups of plant types, separately for NR and SR. The same capital letters indicate homogeneous groups for each plant type between NR and SR. Both were determined at $\alpha = 0.05$. Data are means, $n = 8$ (herbaceous plants and deciduous shrubs), 12 (evergreen shrubs), 20 (evergreen and deciduous trees).

levels consistently recorded for evergreen trees (*C. camphora*, *M. grandiflora*, *O. fragrans*, *P. serrulata*). The percentage of PM_{10} washed off by natural rain (NR_PM_{10}) was strongly positively correlated with the percentage of $PM_{2.5\text{-}10}$ ($NR\_PM_{2.5\text{-}10}$) and $PM_{2.5}$ ($NR\_PM_{2.5}$). Conversely, the percentage of PM_{10} washed off by simulated rain (SR_PM_{10}) was strongly positively correlated with $SR\_PM_{2.5\text{-}10}$ only. There was no significant correlation between $SR\_PM_{2.5\text{-}10}$ and $SR\_PM_{2.5}$. Most evergreen trees (*C. camphora*, *M. grandiflora*, *O. fragrans*, *P. serrulata*) exhibited a high removal percentage for $PM_{2.5\text{-}10}$ and $PM_{2.5}$ pollutants in natural rain conditions (high values of $NR\_PM_{2.5\text{-}10}$ and $NR\_PM_{2.5}$). In contrast, a high percentage of $SR\_PM_{2.5}$ was recorded for the deciduous tree *P. acerifolia*, whereas high values of SR_PM_{10} were observed for the deciduous shrub *A. palmatum*, herbaceous plant *C. dactylon* and evergreen tree *L. lucidum*. Points representing plants that are close to each other indicate similar values of the percentage of washed-off pollutants. Points representing evergreen shrub *L. chinense*, deciduous tree *P. stenoptera*, deciduous shrub *L. indica*, and herbaceous species *O. japonicus* are close to each other on the plot, which indicates similar patterns of PM removal by precipitation (NR_PM_{10}, $NR\_PM_{2.5\text{-}10}$, and $NR\_PM_{2.5}$, as well as SR_PM_{10} and $SR\_PM_{2.5\text{-}10}$) (Fig. 3).

Discussion

This study demonstrated that the proportion of PM, removed by natural versus simulated rainfall differs. The key observations are (i) simulated rainfall is more effective at removing PM from foliage than natural precipitation; (ii) different morphometric and morphological characteristics of plants and leaves affect PM wash-off by natural and simulated precipitation; and (iii) the impact of natural and simulated rainfall on PM removal varied across different plant groups and species.

PM wash-off by natural and simulated rain

Simulated precipitation serves as a method for evaluating the effects of rainfall on PM deposited on foliage[18,23]. Simulated rainfall can dislodge between 25 and 70% of the PM deposited on leaves prior to precipitation[7,15,17,18,20,30,36]. Natural rain has been shown to wash off 28–48% of PM deposited on foliage[16], and 69–79% in heavy rain events[37]. The effectiveness of PM removal is generally heightened by the increased intensity and duration of precipitation[16,18,20,37,38]. These values are in line with the results reported in this study. However, the comparative analysis of natural and simulated rain challenges the prevailing assumption of their equivalency in PM removal from foliage. Simulated rain appears to be more effective and stable at removing PM than natural rain. The efficiency of natural rain varied significantly within a species, indicating that the process is influenced by a complex interplay of plant and environmental factors and cannot easily be simulated in a laboratory. Moreover, it has been observed that in certain species, the quantity of PM on foliage increases after natural rainfall. This phenomenon has already been reported[12,16,22,29,30] and could be attributed to wet deposition

Species	Particulate matter					
	PM_{10}		$PM_{2.5-10}$		$PM_{2.5}$	
	NR	SR	NR	SR	NR	SR
Cinnamomum camphora	42.7 bc B (±10.9)	22.9 a A (±3.8)	54.1 bc B (±14.6)	25.7 a A (±3.9)	15.4 a A (±18.1)	14.1 a A (±3.0)
Ligustrum lucidum	19.9 abc A (±20.4)	28.8 a A (±9.7)	21.6 abc A (±29.0)	31.0 a A (±10.0)	4.0 a A (±41.2)	22.1 a A (±12.2)
Magnolia grandiflora	65.8 c B (±9.4)	24.6 a A (±10.3)	77.0 c B (±3.9)	28.7 a A (±13.3)	32.8 a A (±35.3)	11.5 a A (±5.2)
Osmanthus fragrans	51.0 c A (±26.3)	26.6 a A (±5.1)	59.4 bc B (±19.0)	32.2 a A (±6.2)	28.1 a A (±46.0)	9.1 a A (±3.1)
Photinia serrulata	36.1 abc A (±17.2)	23.3 a A (±8.4)	43.6 abc A (±17.6)	25.4 a A (±9.8)	20.3 a A (±27.6)	15.5 a A (±12.0)
Liquidambar formosana	39.7 abc B (±9.0)	24.6 a A (±3.9)	47.6 bc B (±4.4)	26.3 a A (±5.8)	25.8 a A (±20.2)	20.3 a A (±7.6)
Koelreuteria paniculata	36.2 abc A (±12.2)	22.5 a A (±4.6)	22.6 abc A (±23.6)	24.8 a A (±5.0)	47.6 a B (±12.8)	17.0 a A (±4.3)
Metasequoia glyptostroboides	46.3 bc B (±20.0)	18.4 a A (±3.4)	54.0 bc B (±19.7)	18.8 a A (±7.0)	26.6 a A (±29.3)	19.2 a A (±3.8)
Platanus acerifolia	-0.3 abc A (±46.8)	24.7 a A (±6.6)	28.7 abc A (±31.1)	29.5 a A (±7.8)	-46.8 a A (±87.4)	12.9 a A (±3.3)
Pterocarya stenoptera	-28.7 a A (±46.4)	19.4 a A (±5.3)	-24.2 ab A (±54.5)	21.9 a A (±4.3)	-54.1 a A (±75.1)	12.8 a A (±9.9)
Loropetalum chinense	5.7 abc A (±23.4)	19.3 a A (±6.9)	-33.5 ab A (±53.6)	19.4 a A (±5.7)	42.9 a B (±10.9)	19.3 a A (±10.6)
Pittosporum tobira	30.5 abc A (±13.1)	23.5 a A (±3.8)	32.9 abc A (±14.9)	21.9 a A (±4.8)	25.6 a A (±9.2)	27.8 a A (±2.1)
Rhododendron simsii	21.8 abc A (±17.8)	23.0 a A (±5.3)	-4.2 abc A (±24.0)	25.0 a A (±9.3)	47.5 a B (±11.7)	15.7 a A (±9.7)
Acer palmatum	15.8 abc A (±21.3)	30.7 a A (±9.2)	-17.9 ab A (±39.8)	33.6 a B (±6.5)	48.0 a A (±17.8)	21.8 a A (±19.1)
Lagerstroemia indica	-21.2 abc A (±40.6)	22.5 a A (±6.3)	-47.0 a A (±75.6)	21.6 a A (±5.8)	-2.0 a A (±28.4)	25.8 a A (±9.1)
Cynodon dactylon	-0.9 abc A (±49.1)	32.2 a A (±4.8)	-10.9 abc A (±71.0)	34.6 a A (±7.9)	9.0 a A (±29.4)	24.8 a A (±5.0)
Ophiopogon japonicus	-21.5 ab A (±16.9)	23.0 a B (±4.8)	-27.0 ab A (±14.4)	25.0 a B (±7.2)	-19.6 a A (±63.8)	17.3 a A (±7.3)

Table 4. Mean and standard deviations for the percentage of removed PM attributed to plant species, PM fraction, and natural (NR) and simulated rain (SR). For each variable, the same lowercase letters in columns indicate homogenous groups of treatments, as determined by Tukey's test at $\alpha = 0.05$. Different uppercase letters indicate significant differences between PM fractions/trace elements washed off by natural or simulated rain by t-test at $P < 0.05$.

of PM[16,31,34]. However, in contrast to the finding in this study, this increase in PM after natural rainfall is not immediate and is observed after a delay of several days[36]. It is notable after low-intensity rain, which might be insufficient to overcome the leaf's water retention capacity[11,12,17,30].

Differences between the effects of natural and simulated precipitation can be explained by the rain characteristics (duration, intensity) and the environmental factors (wind, humidity, PM emission). Distilled water, which has different chemical properties (pH) from natural rain, is used in simulated rainfall. Rainwater contains PM that have been washed from the air[27,32,36]. These PM can accumulate on foliage during the wet deposition[17,29,34]. Natural rainfall usually lasts longer than simulated rainfall, is of lower intensity, is not uniform, its intensity and direction may change within a short period and it is usually interspersed with rainless intervals[27,30]. The distance of a raindrop prior to its contact with a foliage plays an important role in the wash-off, particularly when simulated rainfall is generated from a minimal elevation[39]. The kinetic energy inherent in raindrops influences the mechanics of PM removal. Natural raindrops possessing higher kinetic energy have the capacity to dislodge PM through mechanisms involving direct impact, splashing upon foliage, and inducing physical movements of the leaves[17,18,40]. PM starts to be removed when the rain exceeds the maximum interception amount of leaves[11,12,17,19]. Replicating specific moisture and wetting conditions on leaf surfaces is readily achievable in controlled environments with simulated rainfall. Under natural conditions, part of foliage experiences limited interaction with rainwater[30]. Additionally, low rainfall results in an incremental increase in the moisture content of leaf surfaces, which subsequently enhances their adhesive properties[12]. In contrast, in simulated precipitation experiments, the rates of PM wash-off escalate during the initial 10-minute interval of rainfall[12,17,20,24,27,37]. This is because simulated precipitation experiments are typically conducted on individual leaves or shoots, rather than on whole plants. Simulated rain is precisely applied to a static leaf/shoot, ensuring that the resulting runoff is isolated from contact with any adjacent foliage. In contrast, natural precipitation follows a vertical trajectory from the upper to the lower canopy layers, thereby redistributing PM previously

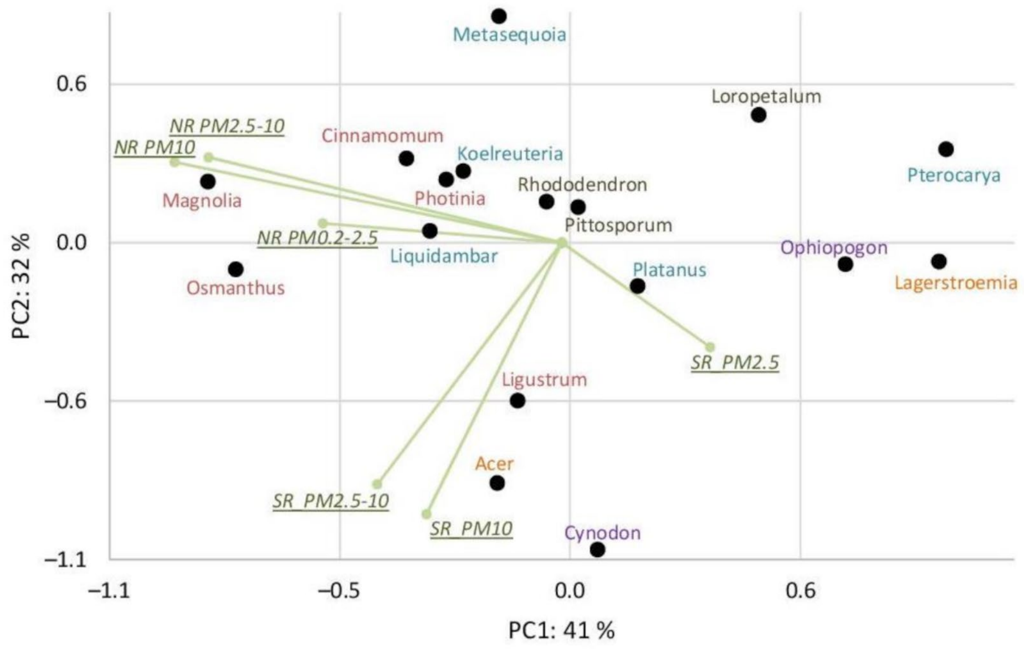

Fig. 3. Visual representation of the types of plants and the percentage of PM washed off by natural (NR) and simulated (SR) rain in the reduced-dimensional PCA space defined by PC1 and PC2. Different colours of points mean different plant types.

accumulated on plants[22,39]. Observations indicate that PM are more concentrated in throughfall than in direct rainfall, suggesting that droplets traversing down the plant surfaces act as vectors for PM[12]. Thus the effect of natural rain on PM wash off may vary with plant height and between different layers of greenery. Additionally, during rainfall, plants are influenced by wind, which induces plant movement and alters the pattern of leaf-rain interaction[18,41]. Furthermore, wind transports pollutants that may adhere more to moist and sticky foliage[35].

Precipitation facilitates the partial removal of PM from plant surfaces across all size fractions, but the efficiency of this process varies[37]. Simulated rainfall washes off effectively PM_{10-100} and $PM_{2.5-10}$, especially those with regular/angular shapes[12,16,18,23,27,30,37,38]. In comparison, $PM_{2.5}$ adheres more strongly to the foliage[12,18], mostly by attachment and adhesion to trichomes and irregularities in the leaf microstructure[16,27,38,40]. In this experiment, simulated rain washed off $PM_{2.5-10}$ more easily than $PM_{2.5}$, while under natural rain, the size of the PM did not affect the ease of removal. However, analysis of the PM size fraction in throughfall can be misleading because rainfall can disaggregate or solubilise PM, changing the particle morphology and leading to a variation in PM size before and after the rainfall[12,20].

Plant traits affecting PM wash-off by natural and simulated rain

The efficacy of PM wash-off from the leaf surface is significantly influenced by leaf morphology/microstructures and its orientation and placement on the plant[7,17,20,24,28]. Various microstructures impact PM of different size fractions. Grooves facilitate the retention of $PM_{2.5}$, while trichomes are better at retaining coarser PM[10]. The same microstructures, such as grooves of different widths may have diverse impacts on the removal of PM varying in size[42]. Moreover, epicuticular wax can have varying forms, each affecting PM retention differently[16]. This study also notes that the effect of rain on PM wash-off from foliage is contingent on the plant's morphometry and leaf morphology. A novel finding is the differentiated role of various plant characteristics determining the effectiveness of these processes under natural and simulated precipitation, with their influence being notably more pronounced under natural rainfall.

Natural rain washed off $PM_{2.5-10}$ most easily from large leaves with smooth surfaces and long petioles. The effective removal of PM from trees was also determined by plant height, LAI, and canopy diameter and height. Rainwater flows through tall, wide and dense trees, easily washing off larger PM particles. Similar observations have been made by Cai et al.[12] who demonstrated that taller plants with larger surface areas and a complex canopy structure intercept rainfall more efficiently, resulting in greater PM wash-off. In contrast, Chen et al.[25] and Popek et al.[26] report a negative correlation between plant density and the efficiency of PM wash-off. This study suggests that plant density increases PM wash-off from foliage, particularly under conditions of prolonged and intense rainfall. Long petioles contribute to the dynamic positioning of leaves under the influence of rain and wind, thereby exposing different leaf surfaces to rainwater[21,28,30]. Broader canopy enhances the distribution of rainwater during precipitation[43]. Interestingly, the presence or absence of microstructures and the amount of wax on leaves had a negligible effect on the retention of $PM_{2.5-10}$.

Conversely, $PM_{0.2-2.5}$ wash-off was influenced by leaf microstructures, and not by plant and leaf morphometry. $PM_{0.2-2.5}$ was most easily washed off by natural rain from leaves with a narrow midrib, dense stomata, and dense and long hairs. All these microstructures have been reported to influence PM retention in laboratory experiments[7–10,44]. Microstructures not only make it easier to capture $PM_{0.2\text{-}2.5}$ from the air, but also improve the leaf's self-cleaning ability, most probably by extending the contact time of raindrops with the leaf surface. Contrary, Xu et al.[28] note that while leaf morphology affects PM accumulation, there is no significant correlation with the PM wash-off rate in natural rain, which is associated with a large aspect ratio because leaves that are prone to disturbance from raindrop impacts tend to lose more PM. The important effect of the presence of microstructures is the creation of different water repellences that result in different wetting properties of foliage[7,20,24,28]. Leaf microstructural features can create different contact angles between a water droplet and leaf surfaces[16] and affect leaf wettability[24,25]. As a consequence, rough leaf surfaces have wetting properties that can increase the contact time between rainwater and the foliage, which provides plants with a greater self-cleaning effect when the rainfall intensity is sufficiently high[7,20,25,45]. The wash-off rate has been found to increase with the water interception efficiency and decrease with the water storage capacity of the leaves[19]. Xu et al.[28] showed that when a leaf has high wettability, rainwater slowly forms a film on the leaf surface, requiring sufficient rain to wet the entire blade and form raindrops that can wash off the leaf surface PM. Leaves with low wettability readily form raindrops that are propelled from the blade by external forces, effectively removing the leaf surface PM. Therefore, PM deposited on hydrophobic leaves is more easily washed off than that deposited on hydrophilic leaves[28]. Important is interaction between the leaf surface and rainfall intensity and duration[20]. For smooth leaf surfaces, long duration-low intensity rainfall could increase the PM removal rate, whereas for rough leaf surfaces short duration-high intensity rainfall could achieve a greater removal rate with the same amount of total rainfall[20]. This work shows that the increased duration of rain is crucial for washing off PM from leaves with complex surfaces, rather than smooth surfaces, as the former take longer to hydrate.

The influence of leaf morphometry and morphology on PM wash-off by simulated precipitation was insignificant. Exceptions to this trend were leaf size and petiole length. Specifically, increases in leaf size and petiole length correspond to a reduction in the quantity of $PM_{2.5\text{-}10}$ and $PM_{2.5}$ being washed off. In contrast, literature data suggest that leaf morphology has a large impact on the PM wash-off from foliage treated with simulated rain, i.e., trichomes and grooves hinder, while smooth and/or water-repellent leaves enhance the wash off[8–10,13,18,23,30,38,41,45,46]. Under simulated rain, the wetting ability of trichomes and rough leaf surfaces promotes contact with water droplets and removes more PM than from smooth surfaces, i.e. thus reveal phenomena similar to those in the present study in the case of fine PM and natural rain[7]. The lack of influence of leaf characteristics on the effectiveness of simulated precipitation can be explained by its precise application, which resulted in rapid hydration of the foliage and easy removal of PM regardless of the leaf characteristics.

Differences between plants in PM wash-off by natural and simulated rain

Natural and simulated rainfall result in the removal of varying quantities of PM from different plants. This variance was not only evident across different plant categories (evergreen shrubs and trees, deciduous shrubs and trees, herbaceous plants), but also within the same group and even the same species. The substantial impact of plant groups and species on PM removal efficiency was corroborated by the PCA, alongside corroborative findings from other investigations[12,18,19,30,38]. Literature suggests that the PM removal rate is greater for broadleaf trees than for conifers[36] and greater for herbaceous plants than for trees[38]. Contrary, Chen et al.[25] showed that the average removal rate of $PM_{2.5}$ from coniferous species was 60% while for broadleaf species it was 47%. This study found that the effectiveness of washing off PM from various groups of plants depends on the precipitation (natural or simulated) and the PM size. Cai et al.[12] demonstrated that large PM is washed off effectively from both trees and shrubs, while fine PM is washed off from trees more easily. In the present study, $PM_{2.5\text{-}10}$ was washed off evergreen trees more effectively by natural rain than by simulated precipitation, which was more efficient at removing PM from all types of shrubs and herbaceous plants, although the difference in evergreens was marginal. In contrast, the effect of rain type on $PM_{2.5}$ removal was not significant, although certain patterns emerged. Natural rain was more effective at cleansing evergreen shrubs of $PM_{2.5}$, while simulated rainfall showed greater efficiency at eliminating $PM_{2.5}$ from deciduous trees and herbaceous plants. Under realistic conditions, urban greenery is often layered, with lower plants growing under trees. Rainfall washes off PM from the higher plants and transports it to the lower layers. This applies especially to coarse PM, which is less strongly bound to the leaf surface. Furthermore, herbaceous plants can accumulate PM from soil particles splashed by rain.

Another interesting finding is that PM was washed off evergreen trees more easily by natural rainfall. Since they retain their leaves throughout the year, evergreen plants accumulate more PM than deciduous plants[25,30]. Winter PM accumulation in Wuhan is not interrupted by rainfall, which allows the PM to bind more tightly. Therefore, effective washing of PM from evergreen plants is possible with a relatively long period of rainfall. Spring rains in Wuhan are often long and intense, the rainfall examined in this work had an intensity of 30 mm and lasted for a day. In contrast, an intense but short simulated rainfall easily washes off PM loosely attached to the foliage, but is not long enough to deal with PM associated with leaf microstructures. The opposite was true for deciduous trees, there was no effect of rainfall type on washing off $PM_{2.5\text{-}10}$, but intense simulated rainfall was more effective at washing off $PM_{2.5}$.

There were differences in washed-off PM not only between different groups of plants, but also within a group of plants. From the majority of evergreen trees (*C. camphora*, *M. grandiflora*, *O. fragrans*, *P. serrulata*), PM was more easily washed off by natural rain. The exception was *L. lucidum*, from which more PM was removed by simulated precipitation. The only morphometric/morphological parameters that distinguish *L. lucidum* from the other trees studied are a smaller crown density and LAI. This confirms the previous hypothesis that in trees the underestimated factor influencing the ability of rain to remove PM is the shape and structure of the plant, and not just the morphology of the leaves. Even greater inter-specific differences were found in the deciduous trees. Natural rain washed off more $PM_{2.5\text{-}10}$ from *L. formosana* and *M. glyptostroboides*, while it increased the deposition of this PM in *P. stenoptera*. In the case of $PM_{2.5}$, natural rain easily washed off PM from *K.*

paniculata and *M. glyptostroboides*, while greater deposition of $PM_{2.5}$ was found on *P. acerifolia* and *P. stenoptera*. An interesting case is *P. stenoptera*, which does not differ significantly in any way from other deciduous trees. *Pterocarya stenoptera* has a low, but not the lowest, canopy density and LAI. *M. glyptostroboides* has lower values of both parameters than *P. stenoptera*. Compared with *P. stenoptera*, *M. glyptostroboides* is a coniferous plant, but it sheds its needles in autumn. *Pterocarya stenoptera*, like *K. paniculata*, also has relatively small leaves and short petioles. This suggests that it is difficult to identify a single feature of a plant that determines the efficiency with which PM is washed off by rainfall, especially natural precipitation. It is a combination of several traits. Within individual species, the greatest differences in efficiency of PM wash-off between natural and simulated rain were found in *L. indica* and *A. palmatum* (deciduous shrubs) and in *O. japonicus* and *C. dactylon* (herbaceous plants). Simulated rain washed off large amounts of $PM_{2.5\text{-}10}$ from these plants, while natural rain increased the deposition of $PM_{2.5\text{-}10}$. The influence of rainfall type on the amount of $PM_{2.5}$ on the foliage of deciduous shrubs and herbaceous plants was not significant, but in each case large differences were found between natural and simulated precipitation.

Conclusions

The results suggest that simulated rain does not reflect the impact of natural precipitation on the processes of PM wash-off from leaves and, therefore, should not be used to explain phenomena that occur in realistic conditions. Simulated rain washes off different amounts of PM to similar natural rainfall. Various morphological features in leaves influence the effectiveness of PM wash-off by natural and simulated precipitation. The role of plant traits in the removal of PM from plants represents a multifaceted process that necessitates both laboratory research and field experiments in order to gain a comprehensive understanding. This dual approach is essential in order to decipher accurately the interactions between plants, PM accumulation on foliage, and precipitation. In studies involving simulated rain, a notable limitation is the failure to incorporate environmental variables. Moreover, a critical oversight in these experiments is the absence of consideration given to the whole plant structure, which emerges as a key factor influencing the efficacy of PM removal. Finally, differences in the response to natural and simulated rainfall were found between different plant groups, but also within the same plant groups and even within species.

Methods

Location and plant material

Wuhan is located in the middle of China (E113°41′ – 115°05′, N29°58′ – 31°22′) (Fig. 4). The climate is humid subtropical with four distinctive seasons. The urban greenery cover is 43%. The average annual temperature ranges from 15.8 to 17.5 °C (3.0 °C in January, 29.3 °C in July), and average annual precipitation is 1,205 mm with a peak in the summer. Figure 5 shows weather conditions and the $PM_{2.5}$ and PM_{10} concentrations during the experimental period.

Jiefang Parkwas selected as the research object. Seventeen species were investigated: evergreen trees (*Cinnamomum camphora* Ness et Eberm., *Ligustrum lucidum* Aiton., *Magnolia grandiflora* L., *Osmanthus fragrans* Loureiro, *Photinia serrulata* (Desf.) Kalkman), deciduous trees (*Liquidambar formosana* Hance, *Koelreuteria paniculata* Laxm., *Metasequoia glyptostroboides* Hu and W. C. Cheng, *Platanus* × *acerifolia* (Aiton) Willd., *Pterocarya stenoptera* D.DC.), evergreen shrubs (*Loropetalum chinense* (R.Br.) Oliv., *Pittosporum tobira* (Thunb.) Aiton, *Rhododendron simsii* Planch.), deciduous shrubs (*Acer palmatum* Thunb., *Lagerstroemia indica* L.) and grasses (*Cynodon dactylon* (L.) Pers., *Ophiopogon japonicus* (Thunb.) Ker Gawl.).

The plant material was harvested twice in the spring of 2023 (12 and 14 April). The date of the first harvest was set before a rainfall event (31 mm), which took place on 13 April. The rain (pH: 6.38, EC: 27.3 $\mu S.cm^{-1}$) lasted for 20 h (3 a.m. − 6 a.m.: 0.8 mm, 8 a.m. − 11 a.m.: 7.8 mm, 12 a.m. − 2 p.m.: 1.9 mm, 10 p.m. − 12 p.m.: 20.8 mm). Wind on the day of precipitation was 1.6–3.3 $m.s^{-1}$. Harvesting was preceded by six days of no precipitation and strong winds ensuring a sufficient amount of PM deposited on leaves. The plant material harvested on 12 April was used to determine the amount of PM deposited on the leaves before natural rainfall, and used in experiments with simulated precipitation. The second harvest was conducted on 14 April after natural rainfall, just after the leaves had dried. Plant material from the second harvest was used to determine the amount of PM deposited on foliage after natural precipitation. For each species, plant material was harvested near the road traffic. To ensure data comparability, leaf samples (1000 cm^2 and 500 cm^2 from the first and second harvest respectively) with petioles were collected from the entire circumference of the plant and branches 1.5–2.0 m above the ground. Leaf samples were close in age. For herbaceous plants, the sample consisted of plant material taken from an area of 625 cm^2 and cut at a height of 5 cm above the ground. Plant material from each species was harvested from four trees/shrubs/herbaceous plots.

Morphometric and morphogenic assessment

Plant height and canopy height and diameter were assessed using standard methods. Canopy density (the ratio of the total projected area (canopy width) of the tree canopy on the ground in direct sunlight to the total forest area (stand), which reflects the density of the stand) was determined by processing the images using HemiView 2.1 (Delta-T Devices, UK) and data calculation. Measurements were taken from four biological replicates.

Hemispherical photographs were taken using a Canon EOS 50D digital SLR camera (Canon Inc., Tokyo, Japan) equipped with a 4.5 mm Sigma EX DC hemispherical lens (Sigma Corporation, Japan). HemiView 2.1 (Delta-T Devices, UK) software was used to process the images for Leaf Area Index (LAI) calculation. LAI was measured on the maximum number of individuals of a given species.

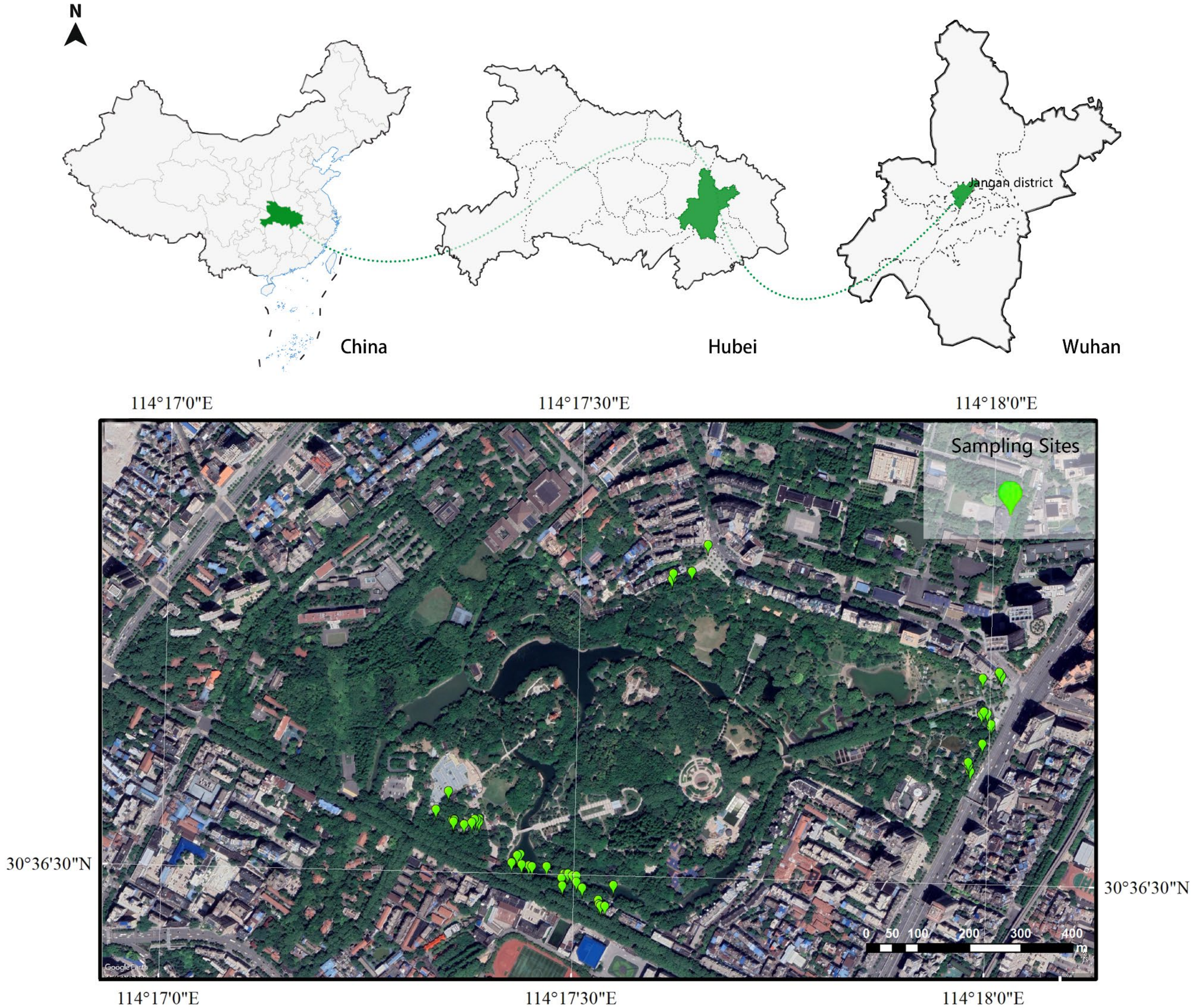

Fig. 4. Study location and the distribution of sampling sites in Jiefang Park, Wuhan, China[47].

Leaf area was measured using the Image Analysis System (Skye Instruments Ltd. UK) and Skye-Leaf software. Petiole length was assessed using a ruler. Measurements of leaf area and petiole length were taken from four biological replicates (single plant or plot with herbaceous plants) and 20 technical replicates (single leaves).

A scanning electron microscope (SEM) (ZEISS SmartSEM EVO-10, Carl Zeiss AG. Germany) was used to study the leaf microstructure. Five randomly collected leaves from each species were analysed. Prior to the analysis, the deposited PM was removed from the foliage by blowing. Two samples (adaxis and abaxis) of 5 mm width and 5 mm length were cut with a blade, pasted onto the sample table and subjected to gold spraying for 300 s. SEM images were taken from five randomly selected locations in each leaf sample, resulting in a total of 50 images of the leaf surface microstructure of each species. Stomatal density and length, midrib width, and hair density and length were measured and counted using the image processing software Adobe Photoshop CC 2018 (Adobe Inc., San Jose, CA, USA).

Categorical factors, leaf texture (0 – smooth: leaves with a smooth and regular texture, without convex structures and roughness; 1 – medium: leaves with a texture between smooth and rough; 2 – rough: leaves with an irregular and rough texture with numerous convex structures), leatheriness (0 – paper: leaves with thin and soft textures, similar to paper; 1 – medium: leaves with leatheriness between leather and paper; 2 – leather: tough and thick leaves) and shape (0 – palmate: leaves with maple-like notches; 1 – oval: leaves of oval or rounded shape; 2 – linear: elongated leaves with a linear shape similar to grasses), and serrated edges (0 – no; 1 – yes) were determined based on organoleptic evaluation of the leaves.

Evaluation of simulated rainfall effects

Leaves were placed on grids located on glass crystallisers (diameter: 23 cm, area: 415.5 cm^2) and sprayed with distilled water (pH: 5.97, EC: 1.5 $\mu S.cm^{-1}$) using plastic spray nozzles, simulating a 30 mm/h rain event lasting 15 min. The duration of the simulated rain was set at 15 min because PM is intensively washed off the plant

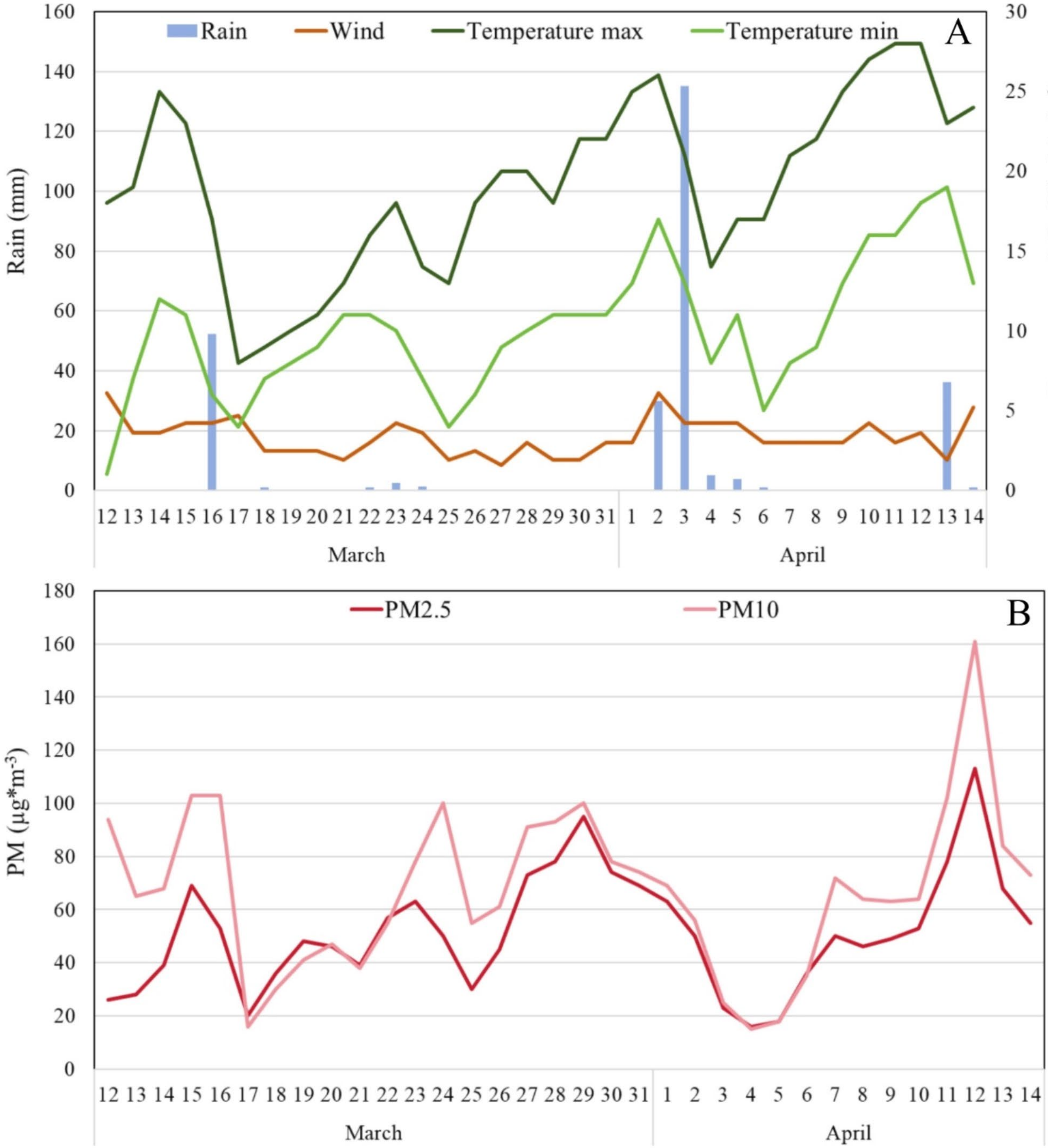

Fig. 5. Daily cumulative rainfall, temperature (min. and max.) and wind speed (**A**) and $PM_{2.5}$ and PM_{10} concentrations (**B**) at the study sites during the experimental period. Data were obtained from www.tianqi.com and www.tianqishi.com (rain), www.tianqi24.com (temperature), www.tianqishi.com (wind) and Wuhan Ecological Environment Bureau (hbj.wuhan.gov.cn, PM concentrations).

surfaces during the first 10 min of rainfall[25]. Plastic spray nozzles were held 50 cm above the leaves. The diameter of the drop was similar to a small raindrop. The leaf samples were then left for one hour to allow the runoff to be collected in the glass crystallisers. Prior to laboratory processing, the samples were stored at 4 °C before the runoff solution was examined for the amount of PM washed off from foliage. Plant material was analysed for residual PM.

Quantitative assessment of PM and leaf wax content

The content of water-insoluble PM deposited on foliage was assessed with the method by Dzierżanowski et al.[48]. The plant material (150 cm^2) was first washed for 60 s with 200 mL distilled water (surface PM) and then for 45 s with 100 mL chloroform (PM retained in wax). The wash solutions were filtered through a 10-µm paper filter (Whatman, UK, type 91), followed by a 2.5-µm paper filter (Whatman, UK, type 42), and finally a 0.2-µm PTFE membrane filter (Whatman, UK). Filtration was performed using a filtration set equipped with a 47-mm glass filter funnel (PALL Corp. USA) connected to a vacuum pump. Two fractions of PM were collected: 0.2–2.5 µm (fine; $PM_{2.5}$) and 2.5–10 µm (coarse; $PM_{2.5\text{-}10}$). The sum of both PM fractions was designated as PM_{10} (total; 0.2–10 µm). The filters were dried at 60 °C for 45 min, stabilised in the weighing room for 45 min, and weighed before and after filtration (balance XS105DU, Mettler-Toledo International Inc., deioniser gate, HAUG, Switzerland). The amount of wax dissolved in chloroform was determined for each plant sample in pre-weighed beakers after chloroform evaporation. The area of samples was measured (Image Analysis System, Skye Instruments Ltd. UK, Skye-Leaf software).

Statistical analysis

To evaluate the effect of rain (natural and simulated) on PM wash off and whether there are any differences between evergreen and deciduous trees and shrubs and herbaceous plants, nested ANOVA (analysis of variance), a statistical method used to compare the means of three or more groups where there is a hierarchical relationship between the groups, was used. Factor A was a plant (evergreen tree or shrub, deciduous tree or shrub, herbaceous plant), factor B was a plant species, and the outcome was the percentage of pollution removed. The null hypothesis stated that there is no difference in the average percentage of removed PM between types of plants. The ANOVA model was as follows:

$$\mathrm{Yijk} = \mu + \alpha \mathrm{i} + \beta \mathrm{j} + \alpha \mathrm{ij} + \epsilon \mathrm{ijk}$$

where Yijk is the observation at the i-th level of factor A, j-th level of factor B within A, and k-th observation within B at the i-th level of A, μ is the overall mean, αi is the effect of the i-th level of factor A, βj is the effect of the j-th level of factor B within A, αij is the interaction effect between factor A and factor B, and ϵijk is the residual error term.

Tukey's honestly significant difference (HSD) test was used to compare means in ANOVA. The analysis was performed at a significance level of 0.05. An unequal variance t-test was used to compare the means of two independent groups when the population variances were not assumed to be equal. This test is often used when the samples are small or the population distributions are unknown. Levene's test was used to assess the equality of variances for rain (natural and simulated). Spearman correlation, a non-parametric measure of association, was used to assess the relationship between plant characteristics and the amount of pollution washed off by natural and simulated rain.

For dimensionality reduction, Principal Component Analysis (PCA) was used, which transforms the original variables into a new set of uncorrelated variables, called principal components, which capture the maximum variance in the data. The data used for the PCA were the means of each variable (PM) for each plant type. Calculations were performed using STATISTICA 13 software (Statsoft, Tulsa, OK, USA).

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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Author contributions

Conceptualization – BJZ, CYZ, AP; Data curation – AP; Formal analysis – BJZ, EWG, AP; Funding acquisition – CYZ, AP; Investigation – BJZ, YZ, ZCF, HL, CYZ, MPismanik, ŁW, MMP, AP; Methodology – BJZ, YZ, CYZ, EWG, AP; Supervision – CYZ, AP; Visualization – BJZ, MMP, AP; Writing of original draft – AP; Review and editing – BJZ, CYZ, MP, MMP, HM, AP.

Declarations

Competing interests

The authors declare no competing interests.

Plant material statement

The experiment (including field studies and harvest of plant material) complied with relevant institutional, national, and international guidelines and legislation.

Additional information

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