Title: Understanding Particulate Matter Retention and Wash-Off during Rainfall in Relation to Leaf Traits of Urban Forest Tree Species

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Supplementary Materials:

Supplementary figures

Figure S1. Percentage changes of PM retention (+) and wash-off (−) in PM mass on both leaf surfaces and in waxes under three rainfall intensities. (**a**–**e**) Pie chart showing a detailed breakdown of the average percent allocation to different rainfall intensities of light (0.2 mm/h for 3 h, green shading), moderate (6.0 mm/h for 3 h, blue shading), and heavy rain (19.5 mm/h for 3 h, gray shading): (**a**) *P. yedoensis*, (**b**) *Q. acutissima*, (**c**) *M. glyptostroboides*, (**d**) *E. japonicus*, and (**e**) *P. densiflora*. Note: Percentage changes between measurements before and after each rainfall intensity are displayed as positive numbers for PM retention and negative numbers for PM wash-off. We calculated the percentage change in leaf-surface and in-wax PM mass after each rainfall intensity on the basis of the initial background value before each natural rainfall event. Data are mean, n = 10. Asterisks represent significant differences between background and estimated values before and after each rainfall event within each tree species (Mann–Whitney U-tests, ** $p < 0.01$; *** $p < 0.001$).

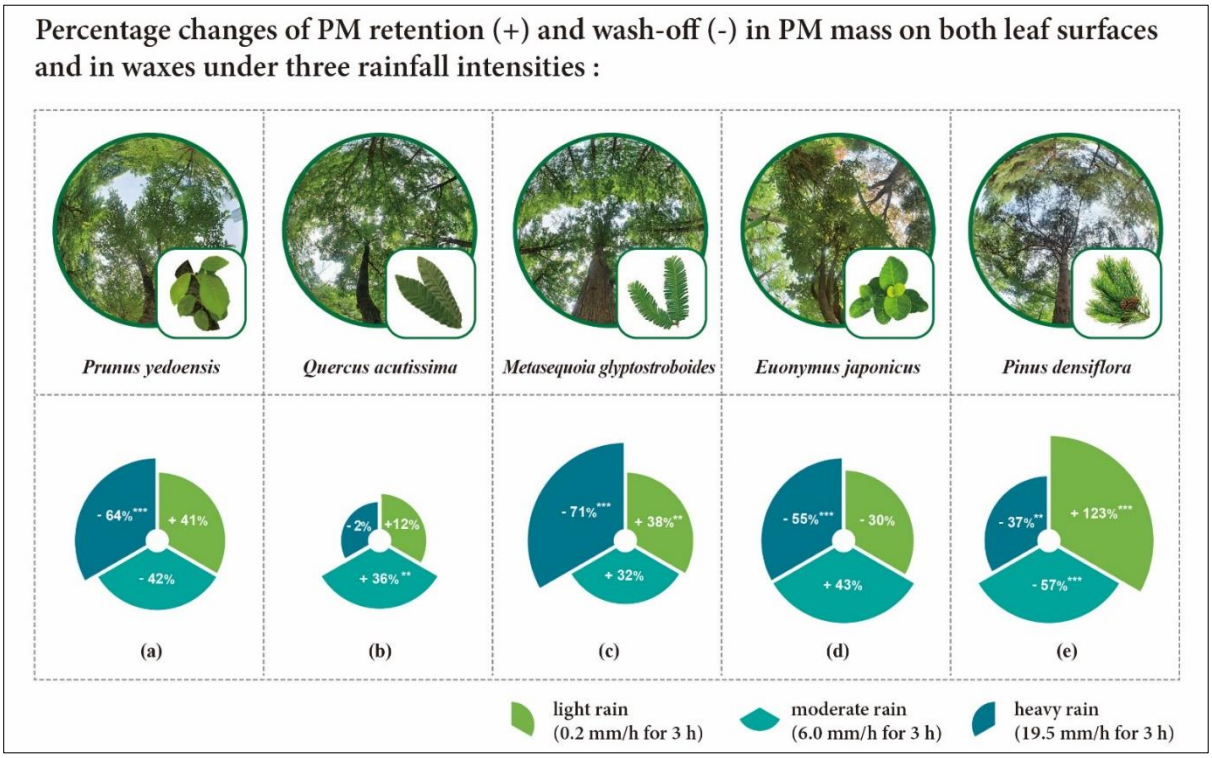


Figure S2. Representative scanning electron microscopy images showing various leaf microstructures and surface roughness on (**a,b,e,f,i,j,m,n,q,r**) adaxial and (**c,d,g,h,k,l,o,p,s,t**) abaxial leaf surfaces. (**a–d**) of *M. glyptostroboides* (waxes on adaxial surfaces), (**e–h**) *P. yedoensis* (roughened stripe-like grooves and non-glandular trichomes on both adaxial and abaxial surfaces), (**i–l**) *Q. acutissima* (adaxial surfaces with bullate-like grooves and abaxial surfaces with simple uniseriate and stellate trichomes), (**m–p**) *E. japonicus* (smooth adaxial surfaces with thick wax layers and adaxial leaf surfaces with concave stomata), and (**q–t**) *P. densiflora* (elliptical-rounded stomata on both adaxial and abaxial needle surfaces). Note: Images show different surface roughness configurations of grooves, stomata, trichomes, and waxes on adaxial and abaxial leaf surfaces. Scale bars: (**e,f,g,i,k,o,q,s**) 100 μm; (**c,m,p,t**) 50 μm; (**a,d,h,j,l,n,r**) 20 μm; (**b**) 3 μm.

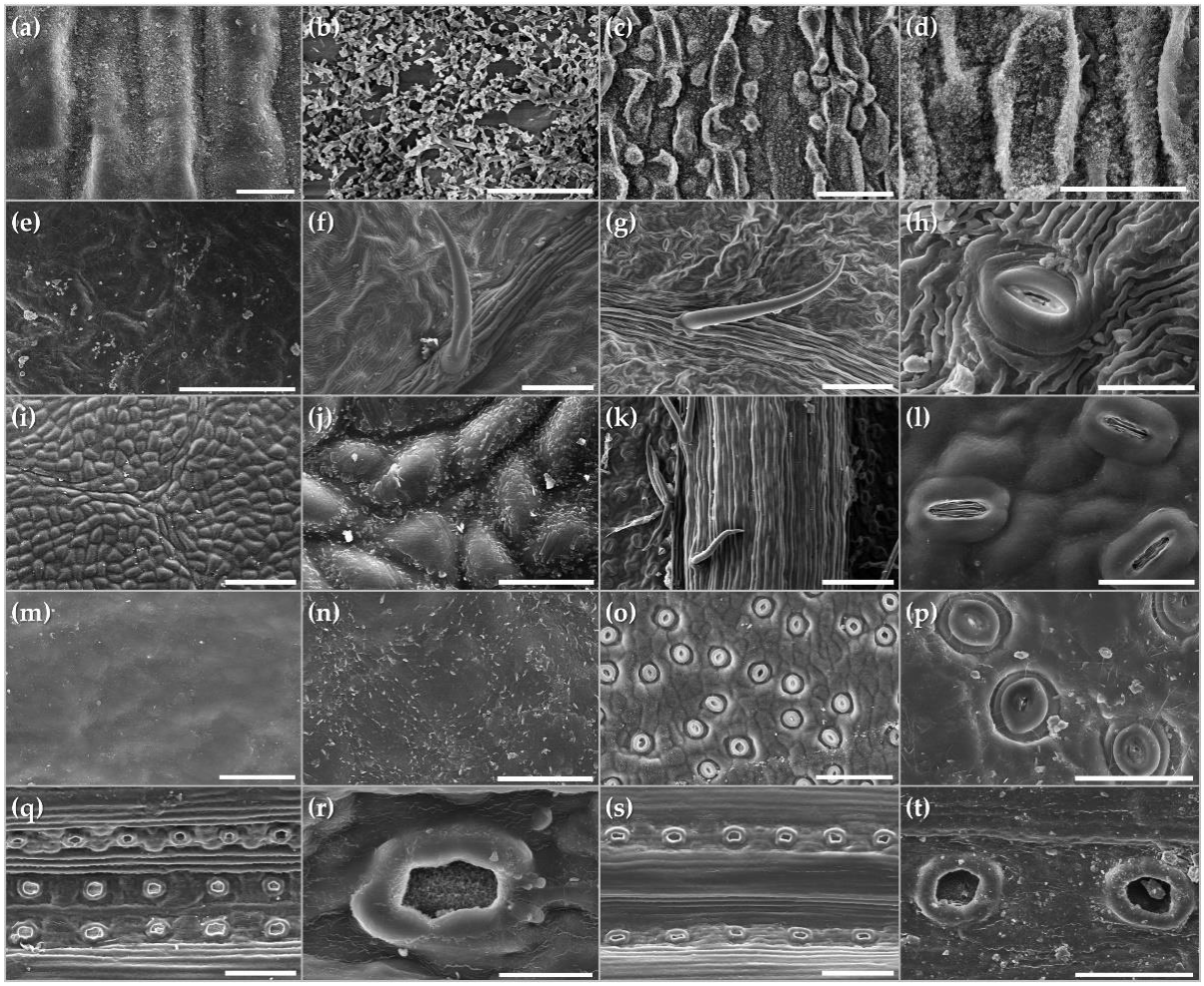


Figure S3. Correlation matrix for all pairs of the observed variables between leaf macro-scale and net PM wash-off ability from the outer crown-positioned leaves. Note: The distribution of each variable is shown on the diagonal. The bivariate scatter plots with a fitted line are displayed on the bottom of the diagonal. The correlation coefficient plus the significance level indicated with asterisks are shown on the top of the diagonal. Each significance level is symbolically encoded at the levels of 0.05 (*), 0.01 (**), and 0.001 (***). Abbreviations: surface PM (OUTER_SPM2.5, OUTER _SPM10), in-wax PM (OUTER_WPM2.5, OUTER_WPM10), and total PM (OUTER_TPM) from the outer crown-positioned leaves, leaf area (LA), perimeter (P), circularity (C), leaf width-to-length ratio (W/L), vein-to-blade ratio (V/B), and leaf roundness index (RI).

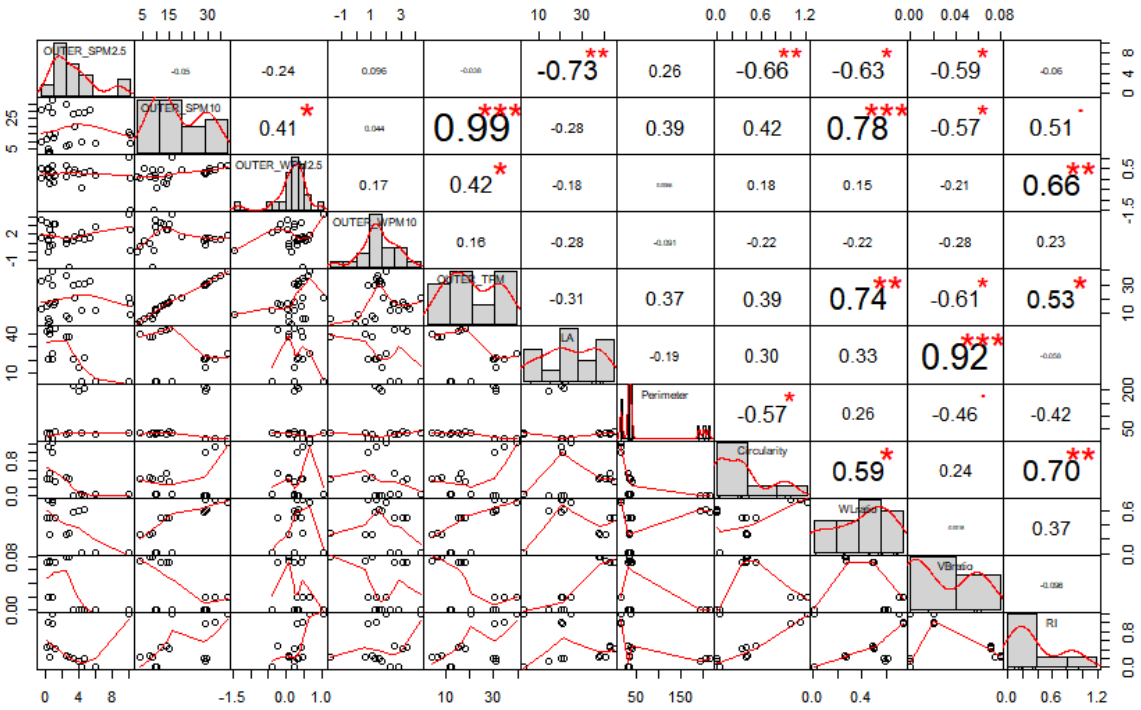


Figure S4. Correlation matrix for all pairs of the observed variables between leaf micro-scale and net PM wash-off ability from the outer crown-positioned leaves. Note: The distribution of each variable is shown on the diagonal. The bivariate scatter plots with a fitted line are displayed on the bottom of the diagonal. The correlation coefficient plus the significance level indicated with asterisks are shown on the top of the diagonal. Each significance level is symbolically encoded at the levels of 0.05 (*), 0.01 (**), and 0.001 (***). Abbreviations: surface PM (OUTER_SPM2.5, OUTER _SPM10), in-wax PM (OUTER_WPM2.5, OUTER_WPM10), and total PM (OUTER_TPM) from the outer crown-positioned leaves, stomatal length (SLab) and width (SWab) on abaxial surfaces, stomatal density on adaxial (SDad) and abaxial (SDab) leaf surfaces, trichome density on vein (TVDad) and blade (TBDad) of adaxial surfaces, trichome density on vein (TVDab) and blade (TBDab) of abaxial surfaces, trichome length on vein (TVLad) and blade (TBLad) of adaxial surfaces, and trichome length on vein (TVLab) and blade (TBLab) of abaxial surfaces.

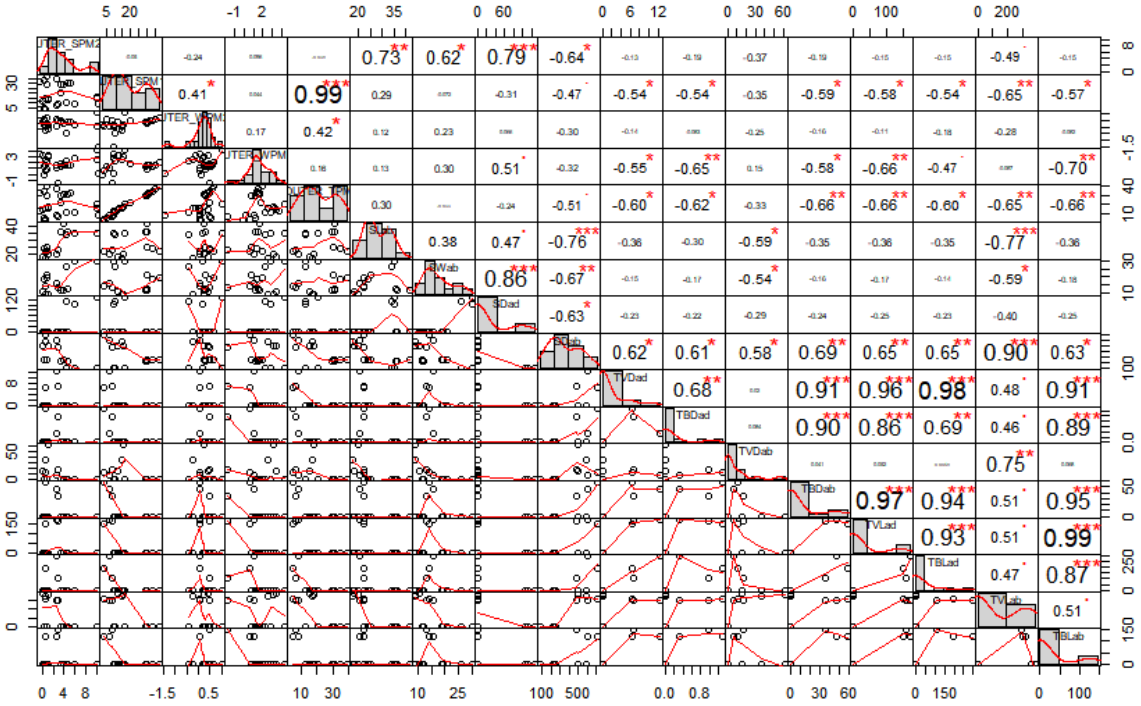


Figure S5. Correlation matrix for all pairs of the observed variables between leaf geometric properties and net PM wash-off ability from the outer crown-positioned leaves. Note: The distribution of each variable is shown on the diagonal. The bivariate scatter plots with a fitted line are displayed on the bottom of the diagonal. The correlation coefficient plus the significance level indicated with asterisks are shown on the top of the diagonal. Each significance level is symbolically encoded at the levels of 0.05 (*), 0.01 (**), and 0.001 (***). Abbreviations: surface PM (OUTER_SPM2.5, OUTER _SPM10), in-wax PM (OUTER_WPM2.5, OUTER_WPM10), and total PM (OUTER_TPM) from the outer crown-positioned leaves, leaf epicuticular wax (Wax), contact angle on adaxial (CAad) and abaxial (CAab) leaf surfaces, and micro-roughness on adaxial (RAad) and abaxial (RAab) leaf surfaces.

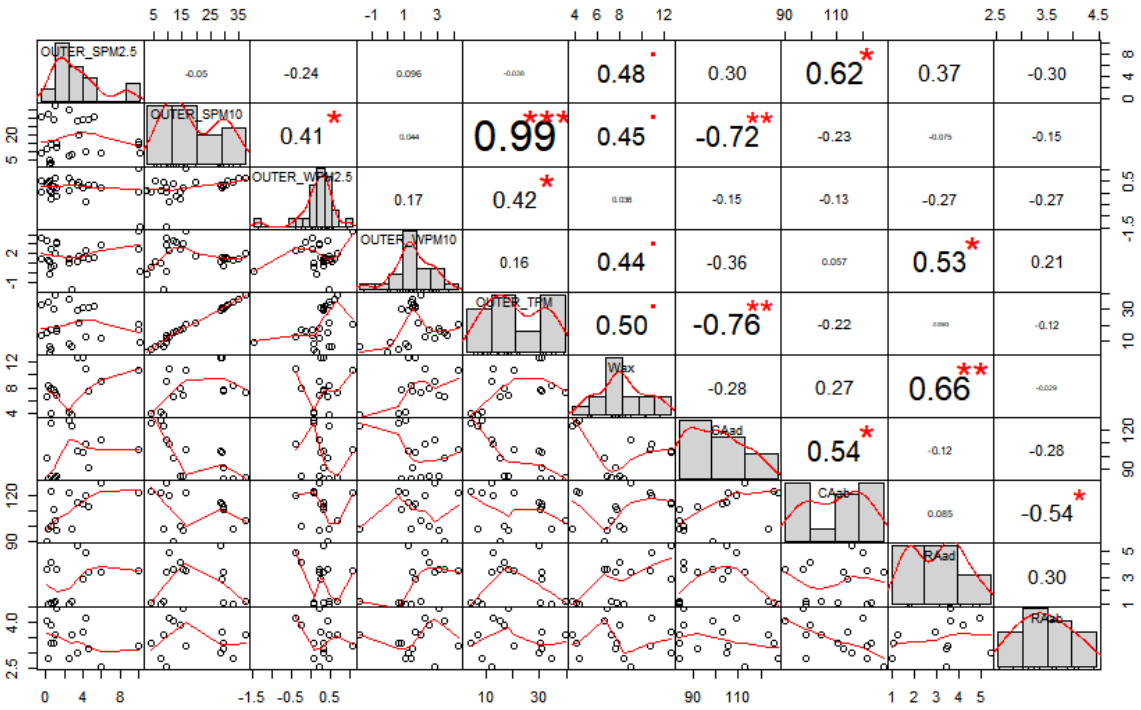


Figure S6. Correlation matrix for all pairs of the observed variables between leaf macro-scale and net PM wash-off ability from the inner crown-positioned leaves. Note: The distribution of each variable is shown on the diagonal. The bivariate scatter plots with a fitted line are displayed on the bottom of the diagonal. The correlation coefficient plus the significance level indicated with asterisks are shown on the top of the diagonal. Each significance level is symbolically encoded at the levels of 0.05 (*), 0.01 (**), and 0.001 (***). Abbreviations: surface PM (INNER_SPM2.5, INNER_SPM10), in-wax PM (INNER_WPM2.5, INNER_WPM10), and total PM (INNER_ITPM) from the inner crown-positioned leaves, leaf area (LA), perimeter (P), circularity (C), leaf width-to-length ratio (W/L), vein-to-blade ratio (V/B), and leaf roundness index (RI).

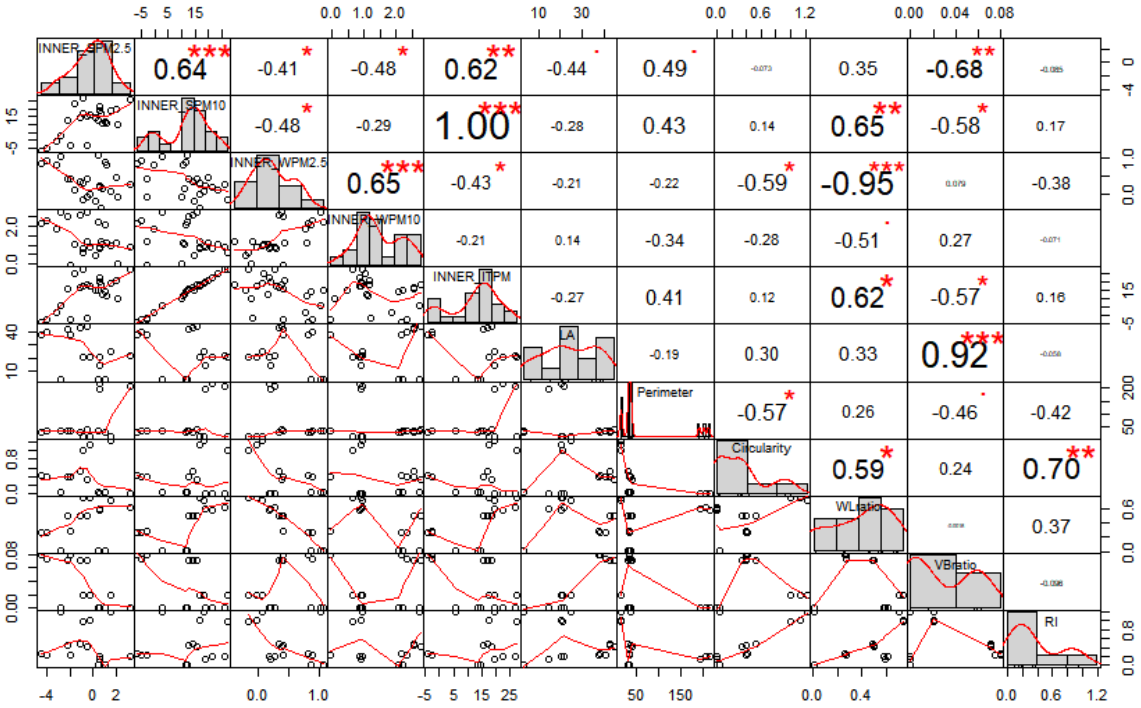


Figure S7. Correlation matrix for all pairs of the observed variables between leaf micro-scale and net PM wash-off ability from the inner crown-positioned leaves. Note: The distribution of each variable is shown on the diagonal. The bivariate scatter plots with a fitted line are displayed on the bottom of the diagonal. The correlation coefficient plus the significance level indicated with asterisks are shown on the top of the diagonal. Each significance level is symbolically encoded at the levels of 0.05 (*), 0.01 (**), and 0.001 (***). Abbreviations: surface PM (INNER_SPM2.5, INNER_SPM10), in-wax PM (INNER_WPM2.5, INNER_WPM10), and total PM (INNER_ITPM) from the inner crown-positioned leaves, stomatal length (SLab) and width (SWab) on abaxial surfaces, stomatal density on adaxial (SDad) and abaxial (SDab) leaf surfaces, trichome density on vein (TVDad) and blade (TBDad) of adaxial surfaces, trichome density on vein (TVDab) and blade (TBDab) of abaxial surfaces, trichome length on vein (TVLad) and blade (TBLad) of adaxial surfaces, and trichome length on vein (TVLab) and blade (TBLab) of abaxial surfaces.

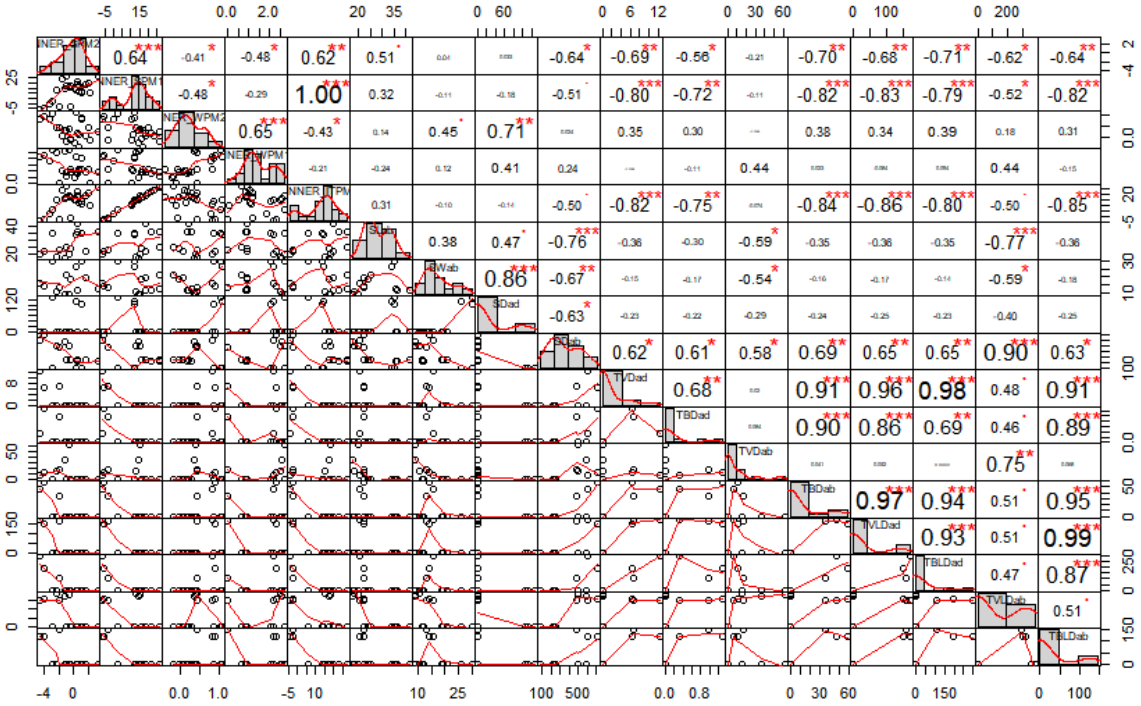
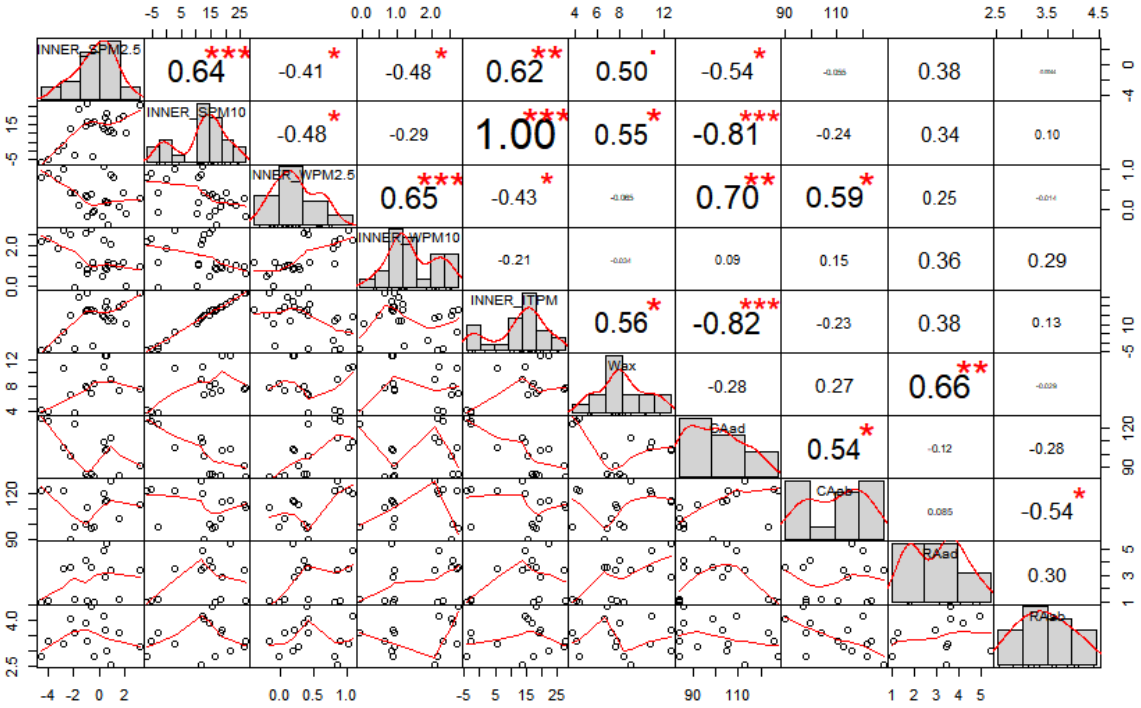


Figure S8. Correlation matrix for all pairs of the observed variables between leaf geometric properties and net PM wash-off ability from the inner crown-positioned leaves. Note: The distribution of each variable is shown on the diagonal. The bivariate scatter plots with a fitted line are displayed on the bottom of the diagonal. The correlation coefficient plus the significance level indicated with asterisks are shown on the top of the diagonal. Each significance level is symbolically encoded at the levels of 0.05 (*), 0.01 (**), and 0.001 (***). Abbreviations: surface PM (INNER_SPM2.5, INNER_SPM10), in-wax PM (INNER_WPM2.5, INNER_WPM10), and total PM (INNER_ITPM) from the inner crown-positioned leaves, leaf epicuticular wax (Wax), contact angle on adaxial (CAad) and abaxial (CAab) leaf surfaces, and micro-roughness on adaxial (RAad) and abaxial (RAab) leaf surfaces.



Supplementary Tables

Table S1. Quantitative assessment of retained PM and wash-off on leaf surfaces and wax layers during rainfall events.

Evaluating the adsorption propensity to PM particles	
PM particles on leaf surfaces	**PM particles in leaf wax layers**
1. PM mass on leaf surfaces before and after rainfall events: 5 species × 5 repetitions × 2 (inner and outer tree crown) × 2 times (before and after rainfall event) × 3 times (rainfall events)	1. PM mass in leaf wax layers before and after rainfall events: 6 species × 5 repetitions × 2 (inner and outer tree crown) × 2 times (before and after rainfall event) × 3 times (rainfall events)
2. Random sampling to achieve a constant leaf surface area (150–250 cm^2) in the sampled twig	2. Chloroform cleaning (CC): After UC, wash the leaves in 250 mL of chloroform for about 40 seconds.
3. Ultrapure water cleaning (UC): soak the sampled leaves in 700 mL of ultrapure water and stir for about 40 minutes (140 rpm)	3. After constant CC process, sequential filtration of the solution containing PM particles with each filter paper through a vacuum filtration device
4. After constant stirring, sequential filtration of the PM-containing solution through each filter paper through a vacuum filtration device	4. The difference in weight of filter paper is measured in the same way as described for measuring PM adsorption on leaf surfaces
5. After filtration, the weight difference between each filtered filter paper (W2) and the original filter paper (W1) is measured as: PM mass on leaf surfaces = W2 − W1	5. After filtration, the weight difference between each filtered filter paper (W4) and the original filter paper (W3) is measured as: PM mass in leaf wax layers = W4 − W3

Table S2. Leaf macro-scale properties of individual leaf shape descriptors in urban forest tree species

Species	Leaf area	Perimeter	Circularity	W/L ratio	V/B ratio	Leaf roundness index
P. yedoensis	43.0 ± 1.0	36.3 ± 4.3	0.421 ± 0.094	0.495 ± 0.006	0.071 ± 0.000	0.440 ± 0.021
Q. acutissima	38.7 ± 1.5	34.7 ± 0.9	0.403 ± 0.015	0.270 ± 0.006	0.078 ± 0.003	0.242 ± 0.007
M. glyptostroboides	19.0 ± 3.9	205.2 ± 10.7	0.006 ± 0.001	0.594 ± 0.019		0.171 ± 0.041
E. japonicus	22.0 ± 2.5	15.8 ± 0.6	1.115 ± 0.114	0.730 ± 0.020	0.020 ± 0.000	1.050 ± 0.119
P. densiflora	3.6 ± 0.1	35.5 ± 3.1	0.037 ± 0.006	0.010 ± 0.001		0.002 ± 0.000

Table S3. Leaf micro-scale structural properties of individual leaf surfaces in urban forest tree species.

Species	Stomatal size		Stomatal density		Trichome length				Trichome density			
	Abaxial		Adaxial	Abaxial	Adaxial		Abaxial		Adaxial		Abaxial	
	Length	Width	Sdad	SDab	Vain	Blade	Vain	Blade	Vain	Blade	Vain	Blade
P. yedoensis	18.6	9.6		524.0			365.7				38.0	
Q. acutissima	21.9	14.4		600.4	161.0	200.8	303.4	129.2	8.58	0.86	11.9	46.8
M. glyptostroboides	37.9	11.8		225.0								
E. japonicus	24.0	19.7		247.7								
P. densiflora	34.7	28.7	115.3	83.1								

Table S4. Leaf geometric properties in urban forest tree species

Species	Wax	CAad	CAab	RAad	RAab
P. yedoensis	7.2 ± 0.7	89.8 ± 7.6	95.5 ± 5.0	3.81 ± 0.33	4.14 ± 0.28
Q. acutissima	4.1 ± 0.3	125.5 ± 2.0	114.3 ± 14.3	1.09 ± 0.13	3.25 ± 0.40
M. glyptostroboides	11.0 ± 2.9	99.7 ± 7.0	114.3 ± 1.4	3.91 ± 1.32	3.27 ± 0.36
E. japonicus	7.8 ± 0.5	83.5 ± 0.1	104.1 ± 6.3	1.09 ± 0.13	3.25 ± 0.40
P. densiflora	10.3 ± 1.0	109.0 ± 3.7	123.4 ± 4.3	3.95 ± 0.85	3.31 ± 0.80