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Immobilized atmospheric particulate matter on leaves of 96 urban plant species

Samira Muhammad*, Karen Wuyts, Roeland Samson

Laboratory of Environmental and Urban Ecology, Department of Bioscience Engineering,
University of Antwerp, Groenenborgerlaan 171, 2020 Antwerp Belgium
samira.muhammad@uantwerpen.be, karen.wuyts@uantwerpen.be,
roeland.samson@uantwerpen.be

*Corresponding author: samira.muhammad@uantwerpen.be; samira.muhammad@gmail.com
Tel: + 32 265 3569

ABSTRACT

Plants provide many ecosystem services in urban environments, including improving ambient air quality. Leaves of plants permit the deposition of particulate matter (PM) and, depending on their leaf traits, PM may be immobilized within the epicuticular wax (EW) layer, on trichomes, on hyphae of fungi, or inside stomatal cavities. In this study, leaves of 96 perennial urban plant species consisting of 45 deciduous broadleaf/needle-like trees, 32 deciduous broadleaf shrubs, 12 evergreen needle/scale-like trees, 5 evergreen broadleaf trees, and 2 climber species were investigated in June and September 2016 to determine the effectiveness of distinct leaf surfaces in PM immobilization after leaf washing treatment. The leaf surfaces were washed vigorously using a vortex shaker. The magnetizable component of accumulated and immobilized PM on the leaf surfaces was estimated using Saturation Isothermal Remanent Magnetization (SIRM) of the unwashed and washed leaves, respectively. In June, the washed-leaf SIRM of deciduous (broadleaf/needle-like) tree and shrub species ($n = 77$) ranged between 0.1 to 13.9 μA . In September, the washed-leaf SIRM of all investigated plant species ($n = 96$) ranged between 1.2 to 35.0 μA . Outcomes of this study indicate that leaves of *Buddleja davidii*, *Viburnum lantana*, and *Sorbus intermedia* showed the highest washed-leaf SIRM and thus were the most effective in immobilizing PM on their leaf surfaces while leaves of *Populus alba*, *Robinia pseudoacacia*, and *Abies fraseri* with lowest washed-leaf SIRM were the least effective. On average, more than half (i.e., 60 %) of the magnetic signal still remained after vigorous washing but a large variation exists between species (9 – 96 %). The leaf SIRM of washed leaves of deciduous broadleaf tree and shrub species was significantly higher compared to leaves of evergreen needle/scale-like species. Evidently, the magnetic signal of unwashed leaves was higher than washed ones and higher in September than in June. Leaf traits significantly influenced the magnetic signal of both washed and unwashed leaves: leaves with a high trichome density or high leaf wettability showed a higher unwashed and washed leaf SIRM compared to leaves with no trichomes or low leaf wettability. The effect of epicuticular wax structure types on leaf SIRM was indicated to be only marginally significant. Moreover, also the immobilized fraction of PM was significantly affected by trichome density and leaf wettability, thus substantiating that plant species with high trichome density and/or leaf wettability not only accumulate more PM but are also less prone to PM re-suspension than other species. In general, the results also indicate that leaf SIRM of unwashed leaves can be a good indicator to determine the effectiveness of a plant species in PM immobilization. Plant species effective in immobilizing PM on their leaf surfaces may likely improve ambient air quality when planted in urban environments. However, it is vital that leaves of these plant species, (i.e., with high PM immobilization abilities), are carefully recycled as they may be polluted.

KEYWORDS

Particle immobilization, leaf wettability, epicuticular wax structures, trichome density, nature-based solutions

1. Introduction

Urban forests provide ecosystem services such as cultural services, provisioning habitat, and environmental services (Samson et al. 2017). Concerning the latter, urban forests have a positive role in removal of gaseous air pollutants and particulate matter (PM: Beckett et al. 2000; McPherson et al. 2005; Nowak et al. 2006; Dzierżanowski et al. 2011; Sæbø et al. 2012; Popek et al. 2013; Samson et al. 2017) and as such improve human health (Weber 2013). The health harming effects of PM increase with a decrease in the aerodynamic diameter (Da) of the particulate which are characterized as follows, coarse-particles ($2.5 \mu\text{m} \leq \text{Da} < 10 \mu\text{m}$), fine-particles ($0.1 \mu\text{m} \leq \text{Da} < 2.5 \mu\text{m}$) and ultra-fine-particles ($\text{Da} < 0.1 \mu\text{m}$). Removal of PM from the atmosphere is either by dry deposition, by wet deposition, (i.e., rain, snow), or by occult deposition (i.e., fog, wind-driven cloud water) (Fowler et al. 1989). Dry deposition of PM to plants and soil is relatively a slower process than wet deposition, but it is continuous and affects all exposed (plant) surfaces (Hicks 1986). Dry deposition of particles occurs either through: sedimentation under gravity, diffusion (Brownian motion) and turbulent transfer resulting in impaction and interception (Davidson and Wu 1989). Smaller phoretic processes such as thermophoresis (i.e., transport due to temperature), diffusiophoresis (i.e., induced by concentration gradient), and electrophoresis (i.e., deposition due to electrostatic forces) are also involved in the deposition of PM (Hicks et al. 2016).

PM deposited on exposed plant surfaces, may be dislodged by wind or washed-off during a rainfall event. Particles which come back into the atmosphere are termed as re-suspended particles (Burkhardt and Grantz 2016). Re-suspension of particles increases with particle size and the amount of PM on the leaf before rainfall and decreases with time since deposition (Nicholson 1993; Chen et al. 2017). For particles which are either held within the epicuticular wax (EW) layer (Dzierżanowski et al. 2011; Hofman et al. 2014), affixed on leaf trichomes (Sæbø et al. 2012; Popek et al. 2013) on hyphae of fungi or trapped inside the stomatal cavities (Lehndorff et al. 2006; Song et al. 2015) are considered as immobilized particles. Kaupp et al. (2000) suggest that 20 % of total leaf contamination can be washed off from leaf surfaces by water. Jouraeva et al. (2002) revealed that heavy metals and polycyclic aromatic hydrocarbons (PAH's) associated with PM are removed from leaf surfaces by rain and wind ensuing a storm event. However, re-suspension of immobilized fine-particles either by wind or rain is shown to be negligible (Terzaghi et al. 2013; Przybysz et al. 2014; Hofman et al. 2014). Hofman et al. (2014) examined the PM immobilization on leaves of *Platanus x acerifolia* by hand washing the leaf samples followed by measuring the saturation isothermal remanent magnetization (SIRM) of the washed leaves. The authors revealed that immobilized PM contributed to 38 % of the leaf SIRM signal. Blanusa et al. (2015) demonstrated that leaf surfaces of *Platanus x hispanica* due to the presence of fine firm trichomes were able to immobilize more than 90 % of the total deposited particles compared to 12 % of PM immobilized on *Olea europea* with glossy leaf surfaces. The authors illustrated that PM immobilization varies between plant species due to the differences in their leaf micro-morphology (Blanusa et al. 2015). Popek et al. (2013) examined leaves of 13 plant species for surface and in-wax immobilized PM and found that on average between plant species, a 40 % of in-wax PM contributed to the net accumulated PM. Sæbø et al. (2012) revealed that of the net accumulated PM, leaves of *Betula pendula* showed 82.6 % of in-wax PM while leaves of *Fagus sylvatica* showed only 25% of in-wax PM. The authors attributed the disparity between plant species for in-wax PM to the amount of epicuticular waxes (Sæbø et al. 2012). However, studies of, e.g., Kaupp et al. (2000), Jouraeva et al. (2002), Dzierżanowski et al. (2011) found no relationship between the amount of waxes and PM accumulation. The authors suggest that concerning the immobilization of PM in the wax layer, the chemical composition and structure of the epicuticular wax may be of more relevance compared to the amount of waxes.

To date, many studies have used the gravimetric approach for the quantification of PM immobilized in the EW layer where the leaf material is washed using chloroform followed by filtering and weighing (e.g., Dzierżanowski et al. 2011; Sæbø et al. 2012; Popek et al. 2013). Another technique put forward to estimate immobilized PM includes washing of leaves with water after exposure to simulated rain (e.g., Chen et al. 2017) followed by filtering, weighing and comparison of the washing solutions. The drawback of the latter

technique is that water-soluble particles may not be quantified (Beckett et al. 2000) and the particles trapped in the leaf are not taken into account. Alternatively, not the washing solution but the washed leaves can be analyzed, as done by Hofman et al. (2014) through magnetic analysis of leaves washed with distilled water. However, the manual washing of leaf material is difficult to standardize and is prone to uneven cleaning, causing an overestimation of the immobilized fraction. Therefore, a standardized approach is required, which enables a uniform and reproducible washing procedure of all leaf samples.

The overall aim of our study was to determine PM immobilization of selected perennial urban plant species and relate PM immobilization with leaf surface characteristics such as trichome density, leaf wettability, and epicuticular wax structure (EWS) types. The specific aims of this study were to (I) determine the PM immobilization differences between plant species ($n = 96$) and functional plant types ($n = 5$) commonly found in urban environments, (II) identify the leaf surface characteristics which influence PM immobilization, (III) investigate the seasonal variation in PM immobilization. We hypothesize that an increase in PM immobilization would be due to an increase in trichome density and leaf wettability. To reduce the variation in the washing process, we introduce a new standardized methodology to vigorously wash leaf samples in water using a vortex shaker.

2. Materials & methods

2.1 *Experimental set-up and plant material*

The study was conducted as a common garden experiment on the premises of the University of Antwerp (Antwerp, Belgium), i.e., on a secluded parcel of the Groenenborger campus (51° 10'46.0"N, 4° 25' 0.02"E). The set-up of the common-garden is fully described by Muhammad et al. (2019). In summary, 96 perennial plant species commonly used in the Western-European urban environments were selected (Table 1), of which, 45 species were deciduous (broadleaf/needle-like) trees, 32 deciduous broadleaf shrubs, 12 evergreen needle/scale-like, five evergreen broadleaves, and two climber species. For each plant species, five replicates were bought from one pesticide-free nursery (Houtmeyers in Eindhout-Laakdal, Belgium) potted in 15 L pots with organic soil and slow-release fertilizer (Peltracom NV, Belgium) and randomly placed in a common-garden in a 1.5 m x 1.5 m setting. All plants were regularly watered and monitored for any pests or disease, yet not all replicates of each species remained by the time of sampling. Two sampling campaigns were organized during the growing season of 2016; first in June and second in September. For the June sampling campaign, leaves of the evergreen species were about one year old while leaves of the deciduous species ($n = 77$) were from the current growing season. During the considered leaf season (1st April – 30th September 2016) the mean total PM₁₀ and PM_{2.5} concentrations measured at the nearest air quality monitoring station (42R817, Antwerpen Groenenborgerlaan, at 250 m from the common-garden site, operated by Flanders Environment Agency, VMM) were 21.8 and 11.2 µg/m³ respectively. Meteorological data were obtained from the station Antwerpen Luchtbal (station 42M802, Havanastraat, Antwerp operated by VMM). From April – September 2016, the mean for precipitation, air temperature, wind speed, and relative air humidity was 74.3 mm, 15.4 °C, 3.1 m/s, and 72 % respectively.

2.2 *Leaf washing protocol*

Mature and undamaged leaves were harvested from plant replicates ($\sim n = 3 - 5$) of the investigated perennial urban plant species ($n = 96$) on a span of two days in June (9th, 10th) and in September (1st, 2nd) to minimize variations for differences in exposure time. Leaf samples from each investigated plant species and their respective replicates were collected from the south-east direction of the plant, to eliminate within canopy orientation bias. The harvested leaf samples were stored in labeled paper bags and taken to the Laboratory of Environmental and Urban Ecology. To determine the leaf surface-accumulated particles and the leaf-immobilized particles, each leaf sample from each replicate of each plant species was subsequently divided into two sub-samples while maintaining a minimum leaf area of 100 – 150 cm². The leaf area of fresh leaves from each sub-sample was measured using a leaf area meter (Li-3100, LiCor Biosciences).

To evaluate the immobilized PM in and on leaves, we applied a novel methodology for the removal of surface particles using a vigorous vortex shaker. For each sample, all leaves of one of the two paired sub-samples were placed in 50 mL falcon tubes (Greiner Bio-one) and 50 mL of mineralized ultra-pure distilled water with a conductivity of $0.01 \mu\text{S cm}^{-1}$ was added. The falcon tubes were closed tightly with a lid and fastened to a Vortex-Genie 2 (MO BIO Laboratories Inc. New York, USA). The samples were vigorously shaken at a speed of 3200 rpm for three minutes precisely. After washing, the leaf samples were removed from the falcon tubes with the help of tweezers and placed in Petri dishes for air drying. Once the leaf samples were air-dried, the ferromagnetic and magnetizable component of immobilized PM was evaluated using Saturation Isothermal Remanent Magnetization (see § 2.3). By standardizing the washing procedure, we eliminated the possibility of an uneven surface cleaning between replicates and plant species and avoided mechanical damage to the leaf, which can be a risk if the leaf washing process was performed manually. The washing protocol for all leaf samples was performed in the Laboratory of Applied Microbiology and Biotechnology.

The duration and intensity of washing the leaf samples were selected based on preliminary tests. Leaves of *Hedera helix*, an evergreen climber species, were used for all pre-testing. An iterative process was performed to test the dislodging of particles at several motor speeds (3, 5, 7, and 10; with 3 being gentle shaking and 10 being forceful vortexing) and durations (1, 3, 5, and 10 minutes). The preliminary tests showed that after one minute of leaf washing on average, 20 % of the magnetic signal was removed. However, after three minutes of leaf washing on average, 60 % of the magnetic signal was removed. Washing the leaf samples for any prolonged duration (5 and 10 minutes) showed only minor further differences in the magnetic signal. The results showed that the magnetic signal decreased by 30 % at a motor speed of 3 while at a motor speed of 7, this was 55 %. Hence, we used the maximum motor speed of 10 (3200 rpm) and a shaking duration of three minutes to remove the surface accumulated particles.

2.3 Saturation isothermal remnant magnetization (SIRM)

The ferromagnetic and magnetizable component of PM accumulated and PM immobilized on leaf surfaces was estimated using saturation isothermal remnant magnetization (SIRM). The pre-processing protocol of Hofman et al. (2013) was followed. Each leaf sample was tightly packed in a cling film and pressed in a 6.7 cm^3 plastic container. The sample containers were magnetized at a magnetic field of 1 T using a pulse magnetizer model 660 (2G Enterprises, Mountain View, California, USA). Soon after the remanent magnetic intensity was determined using a 2G magnetometer (2G Enterprises). For each measurement, the leaf sample container was placed at 'load position at 0° '. Next, the samples were placed and measured at 'background-position' and finally at 'measurement position' to minimize measurement errors. The magnetic moment measured in emu/cm^3 was multiplied by 10^{-3} to convert it to (Am^2). The resultant was divided by leaf surface area to obtain a leaf surface area (m^2) normalized SIRM values. The final leaf area-normalized SIRM value was denoted as A ($A = \text{Am}^2/\text{m}^2$) and expressed in μA . All magnetic measurements were carried out at the Royal Meteorological Institute of Belgium in Dourbes, Belgium. The leaf SIRM after leaf washing was considered to reflect the immobilized PM and denoted as SIRM_w . The leaf SIRM determined without leaf washing represents the surface-accumulated particles and is denoted as SIRM_u . The SIRM_u values were previously reported on by Muhammad et al. (2019). The fraction of immobilized PM is calculated as the ratio of $\text{SIRM}_w/\text{SIRM}_u \times 100\%$. For the deciduous tree and shrub, species ($n = 77$), an absolute difference in leaf SIRM ($\Delta\text{SIRM} = \text{SIRM}_u - \text{SIRM}_w$) was calculated for both June and September. For the evergreen species with leaves of about one-year-old in June, the ΔSIRM was calculated only for September.

2.4. Leaf characteristics

Leaf wettability, trichome density, and epicuticular wax structure types were determined from the same replicate trees in June and September from the same growing season. The data and the full description of the methodologies applied for leaf wettability and trichome density have been previously reported by Muhammad et al. (2019).

2.4.1 Leaf wettability: drop contact angle (DCA)

For leaf wettability measurements, leaf samples were harvested separately, and in batches, on a span of ten days (13th – 24th June and 12th – 23rd September) after the main leaf harvesting (see § 2.2) campaign. Soon after harvesting, drop contact angle (DCA) measurements were conducted on the right side of the fresh leaf sample from each available replicate (~ n = 3 - 5) of each plant species (n = 96) according to the method described by Kardel et al. (2012). The leaf samples were affixed on wooden laths, using double-sided tape. A 7.5 µL droplet of distilled water (for broadleaves) and 4 µL droplet (for needles) was carefully placed on the leaf sample using a micropipette. The DCA was obtained from both the abaxial (AB) and the adaxial (AD) leaf surfaces. Images of droplets were acquired using a Canon EOS 550D camera attached to a macro lens (MP-E 65mm 1:2.8) with 3x magnification. The left and the right inside contact angles between the droplet outline intersecting the solid surface (i.e., leaf surface) within a droplet were measured using ImageJ and the drop snake analysis plugin developed by Stalder et al. (2006). The DCA for a single replicate was calculated as an average of the left and the right angle whereas, the DCA for a plant species was calculated by averaging over all replicates.

2.4.2. Trichome density (TD)

Leaves of all plant species were inspected for trichomes on their surfaces using a binocular microscope. When trichomes were present, one small disc (approx. 12 mm in diameter) was punched out using a leaf perforator from the left side of the leaf, from each available replicate (~ n = 3 - 5) in both June and September 2016. Subsequently, following the chlorophyll clearing protocol (Gudesblat et al. 2012) the leaf discs, were placed in 95 % ethanol (3 days) followed by 1.25 M NaOH: EtOH (1:1 v/v) solution for two hours, finally followed by 85 % lactic acid (3 – 5 days). All leaf discs were washed with 35 % ethanol before mounting the discs on microscope slides. A droplet of glycerin was placed on the slide and using tweezers the cleared leaf discs were gently placed on the slide and covered with a glass coverslip. The procedure was followed for both the adaxial and the abaxial leaf side. All prepared slides were imaged using a light microscope (Olympus CX41) at a magnification of 4×10. Using the cell counter plugin in ImageJ, the acquired images were analyzed for TD estimation. For each replicate and leaf side, ten images were analyzed. An average trichome count of all replicates divided by the surface area of the images analyzed (mm²) was considered as a final reading for TD per plant species.

2.4.3. Epicuticular wax structure (EWS) type

The characterization of epicuticular wax structure (EWS) type on leaves of the 96 species was accomplished by Scanning Electron Microscope (SEM) images collected from leaf discs of both the AB and the AD leaf sides in both June and September. Leaf discs of ~12 mm in diameter were punched out from the lower left side of the leaf using a leaf perforator avoiding the midrib. The leaf discs were placed on an aluminum stub (Ted Pella Inc.) and affixed in place using conductive double-sided tape (PELCO Tabs 12mm, 16084-1). Next, the leaf discs were air-dried at room temperature. Three days before imaging for EWS, the leaf discs were coated with a layer of carbon approximately 20 nm (Leica EM ACE600) in a vacuum environment to avert charge build-up effect. Lastly, the leaf discs were inspected and imaged using a Quanta 250 Field Emission Gun Environmental Scanning Electron Microscope (FEG-ESEM) which requires a high vacuum setting of 10⁻³ Pa. A spot size of 2.5 was selected, and the distance between the electron emitter and the sample stage was set to 10 mm. A magnification of 10000 – 50000x was used depending on the sharpness of the feature. Lastly, an accelerating voltage of 20 kV was set before imaging the leaf samples. The observed EWS types were classified according to the types described by Barthlott et al. (1998). ‘Gaps’ within the wax crystals (Marcell and Beattie 2002) were evaluated qualitatively using digital micrographs for the leaf samples collected in June and compared against those collected in September. Plant species with a different EWS type in June compared to September were classified as “EWS change”, plant species with the same EWS type in both June and September but showed an increase in gaps or reduced clustering between the wax crystals were classified as “cluster decrease”, plant species with no decrease in clustering of the wax crystals and no change in EWS type were classified as “no change”.

2.5 Data Analysis

In June, one-way analysis of variance (ANOVA) followed by Tukey-HSD post hoc test was applied to analyze the differences in SIRM_w between deciduous (broadleaf/needle-like) tree and deciduous broadleaf shrub species while in September all investigated functional plant types (n = 5) were included. The median leaf SIRM_w values were calculated for functional plant types in June (n = 2) and September (n = 5). To identify the effects of leaf characteristics on leaf SIRM, we applied a linear mixed-effect regression (LMER) model due to the repeated measurements to the same replicates and the hierarchical nature of the dataset. The LMER model was built using the plant id as a random effect, whereas the fixed effects were time with two levels (June and September), treatment with two levels (unwashed and washed leaf samples), and leaf characteristics, i.e., the EWS types at four levels (crusts, platelets, thin film, tubules following the classification of Barthlott et al. 1998) and drop contact angle and trichome density as a continuous variables. The response variable leaf SIRM was natural log (ln) transformed. The LMER model was initiated by including all fixed effects. Subsequently, model parameters with non-significant (p > 0.05) estimates were successively removed. The LMER was also applied to the immobilized fraction of PM with time (i.e., June and September) and leaf characteristics (i.e., drop contact angle, trichome density, EWS types) as fixed effects and plant-id as random effect to determine the effective leaf characteristics in PM immobilization. The Akaike Information Criterion (AIC) was used to compare the performances of different model structures. Normality of residuals was checked statistically using the Shapiro-Wilk test and also by Normal Q-Q plot. An ANOVA was performed on the seasonal variation in SIRM of washed leaves, (i.e., SIRM_D = SIRM_w in September - SIRM_w in June) with a change in EWS from June to September. Pearson correlation was computed on ln(SIRM_U) and ln(SIRM_w) to identify the relationship between SIRM of unwashed leaves to SIRM of washed leaves for June (n = 77) and September (n = 96). In addition, separate correlations were computed for leaves with and without trichomes in September and for the identified EWS types in both June and September.

Leaf samples of evergreen needle/scale-like (n = 12), evergreen broadleaf (n = 5) and evergreen climber species (n = 2) were about one year old in June. Therefore, in June, we did not calculate the median values, absolute difference (Δ SIRM § 2.3) and the immobilized fraction of PM for plant species within the above-mentioned functional plant types. The SIRM values obtained in June for the evergreen plant species (n = 19) were not included in either correlation tests or LMER model. In September, the SIRM values of all investigated plant species (n = 96) were included in all statistical analysis as then the magnetic analysis was performed on leaves that emerged in that growing season, including those of the evergreen species. As such, leaves of all investigated plant species in September were fairly of equal age to obtain impartial comparisons in PM immobilization between species. All analyses were performed using the software R, version 3.4.2 (R Development Core Team 2017), the *Stats* package (R Core Team and contributors worldwide) and the add-on package lmerTest (Kuznetsova et al. 2017). The box plots and XY-plots were generated using the lattice package (Deepayan, 2008).

3. Results

3.1 Differences in leaf SIRM_w between plant species and plant types

The SIRM of washed leaf samples (SIRM_w) varied between plant species (Table 1, Fig. 1). In June, the leaf SIRM_w for the deciduous (broadleaf/needle-like) tree and shrub species (n = 77) ranged from 0.1 to 13.9 μ A. The lowest leaf SIRM_w was identified on leaves of *S. purpurea* (0.1 μ A), *L. anagyroides* (0.4 μ A) and *P. alba* (0.5 μ A). The highest leaf SIRM_w was identified on leaves of *V. lantana* (13.9 μ A), *S. intermedia* (9.9 μ A) and *R. rubiginosa* (6.1 μ A).

In September, the leaf SIRM_w consisting of all investigated plant species (n = 96) ranged from 1.2 to 35.0 μ A. The lowest leaf SIRM_w was observed on leaves of *P. alba* (1.2 μ A), *R. pseudoacacia* (1.9 μ A) and *A. fraseri* (2.0 μ A). The highest leaf SIRM_w was observed on leaves of *B. davidii* (35.0 μ A), *V. lantana* (34.5 μ A) and *S. intermedia* (26.0 μ A).

Table 1 Analyzed plant species (n = 96) with indication of family (n = 29) denoted as (1 – 29; see text box below) and functional plant type (n = 5, EN = evergreen needle/scale-like, EB = evergreen broadleaf, DT = deciduous needle-like and broadleaf tree, DS = deciduous broadleaf shrub, CL = climber) in June and September 2016 calculated from ~ 3 – 5 replicates per species for leaf SIRM (Saturation Isothermal Remanent Magnetization in μA) of unwashed and washed leaf samples, immobilized fraction of particles expressed as %, drop contact angles (DCA in $^\circ$) on the abaxial (AB) and the adaxial (AD) leaf sides and trichome density (mm^{-2}), “+++” indicates leaves with dense fibrous network of trichomes for which trichome density could not be estimated. Epicuticular wax structure (EWS) types and the change in EWS from June to September, is “↓” a decrease in clustering of wax crystals within the same type of wax structure, “Δ” a change in the type of EWS or “↔” no change in type or clustering of EWS. Leaves of plant species names in the bold text were one year old in June 2016. Missing data are indicated by “-”. [Except for leaf SIRM of washed leaves and percent PM immobilized all other data have been previously reported by Muhammad et al. (2019) and Muhammad et al. (2020)]

PLANT SPECIES	JUNE					SEPTEMBER					Epicuticular wax structures			
	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	Trichome density	EWS June	EWS September	EWS Change
<i>Abies fraseri</i> (EN) ²¹	-	-	-	73	72	11.68	2.02	17	90	56	-	Crusts	Crusts	↓
<i>Abies koreana</i> (EN) ²¹	-	-	-	115	89	10.49	3.07	29	111	66	-	Crusts	Tubules	Δ
<i>Abies nordmanniana</i> (EN) ²¹	-	-	-	72	68	10.28	2.88	28	64	64	-	Crusts	Crusts	↔
<i>Acer campestre</i> (DT) ²⁶	7.91	5.61	71	69	83	28.88	22.07	76	67	78	4.04	Thin film	Thin film	↔
<i>Acer ginnala</i> (DT) ²⁶	3.99	1.62	41	88	81	14.47	11.32	78	61	73	-	Thin film	Thin film	↔
<i>Acer platanoides</i> (DT) ²⁶	5.58	3.95	71	86	96	20.96	10.80	52	76	67	-	Platelets	Platelets	↔
<i>Acer pseudoplatanus</i> (DT) ²⁶	9.07	4.70	52	133	76	6.46	11.12	-	106	63	-	Platelets	Platelets	↔
<i>Aesculus hippocastanum</i> (DT) ²⁶	6.80	4.61	68	97	84	29.59	23.00	78	88	62	9.96	Thin film	Crusts	Δ
<i>Alnus glutinosa</i> (DT) ⁶	8.15	5.36	66	65	65	9.00	6.91	77	59	58	0.46	Thin film	Platelets	Δ
<i>Alnus incana</i> (DT) ⁶	5.27	3.49	66	115	75	20.43	14.00	68	98	69	9.00	Thin film	Crusts	Δ
<i>Amelanchier lamarckii</i> (DS) ²⁴	3.71	2.47	67	113	85	17.55	13.09	75	77	85	-	Tubules	Tubules	↓
<i>Betula pendula</i> (DT) ⁶	3.53	1.59	45	73	75	10.35	7.15	69	76	74	-	Platelets	Platelets	↔
<i>Buddleja davidii</i> (DS) ²⁷	6.41	4.97	78	133	76	37.97	35.04	92	124	63	+++	Thin film	Thin film	↔
<i>Carpinus betulus</i> (DT) ⁶	6.32	3.01	48	89	76	30.95	23.51	76	67	74	1.17	Platelets	Crusts	Δ
<i>Castanea sativa</i> (DT) ¹⁵	6.02	2.32	39	68	73	15.72	9.08	58	64	70	13.58	Crusts	Crusts	↔
<i>Catalpa bignonioides</i> (DT) ⁷	3.70	1.25	34	94	79	9.73	9.44	97	80	62	5.29	Thin film	Thin film	↔
<i>Cedrus deodara</i> (EN) ²¹	-	-	-	96	101	12.77	6.29	49	71	79	-	Tubules	Tubules	↓
<i>Chamaecyparis lawsoniana</i> (EN) ¹¹	-	-	-	111	117	12.59	5.84	46	108	104	-	Tubules	Tubules	↓
<i>Cornus alba</i> (DS) ¹⁰	3.80	2.77	73	120	88	11.13	7.46	67	111	73	21.54	Thin film	Platelets	Δ
<i>Cornus mas</i> (DT) ¹⁰	4.86	3.22	66	78	83	11.00	6.05	55	62	74	5.96	Platelets	Platelets	↓
<i>Cornus sanguinea</i> (DS) ¹⁰	3.16	1.30	41	81	74	12.84	9.09	71	63	74	15.63	Thin film	Platelets	Δ
<i>Corylus avellana</i> (DS) ⁶	6.37	3.76	59	77	76	20.27	17.19	85	63	69	4.38	Platelets	Platelets	↓
<i>Corylus colurna</i> (DT) ⁶	8.18	4.78	58	62	56	20.43	15.32	75	57	63	9.50	Thin film	Thin film	↔

PLANT SPECIES	JUNE					SEPTEMBER					Epicuticular wax structures			
	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	Trichome density	EWS June	EWS September	EWS Change
<i>Crataegus monogyna</i> (DT) ²⁴	3.81	1.75	46	98	78	16.15	10.90	67	72	65	1.08	Crusts	Thin film	Δ
<i>Elaeagnus angustifolia</i> (DT) ¹²	8.11	5.79	71	147	85	14.12	8.37	59	124	79	45.13	Crusts	Crusts	↔
<i>Euonymus europaeus</i> (DS) ⁹	4.99	3.08	61	88	88	15.24	7.99	52	63	74	-	Platelets	Platelets	↓
<i>Fagus sylvatica</i> (DT) ¹⁵	8.55	4.86	57	92	90	22.07	16.05	73	69	75	9.67	Crusts	Crusts	↓
<i>Fraxinus excelsior</i> (DT) ²⁰	4.34	2.83	65	71	80	14.44	10.32	71	55	64	-	Thin film	Thin film	↔
<i>Fraxinus ornus</i> (DT) ²⁰	4.05	2.19	54	80	67	12.46	9.26	74	67	67	-	Thin film	Thin film	↔
<i>Ginkgo biloba</i> (DT) ¹⁶	3.14	1.07	34	131	127	12.89	4.62	36	117	70	-	Tubules	Tubules	↔
<i>Hedera helix</i> (CL)³	-	-	-	74	82	9.09	4.04	44	72	74	0.58	Platelets	Platelets	↓
<i>Hibiscus syriacus</i> (DS) ¹⁹	3.90	1.42	36	77	73	14.60	11.18	77	60	62	1.29	Thin film	Platelets	Δ
<i>Hippophae rhamnoides</i> (DS) ¹²	7.11	3.79	53	117	86	16.11	8.22	51	101	84	-	Thin film	Thin film	↔
<i>Ilex aquifolium</i> (EB)⁴	-	-	-	93	89	9.54	4.68	49	80	83	-	Thin film	Platelets	Δ
<i>Juglans regia</i> (DT) ¹⁷	3.26	0.91	28	76	71	17.33	7.44	43	60	69	-	Crusts	Crusts	↓
<i>Juniperus communis</i> (EN)¹¹	-	-	-	99	89	18.59	13.73	74	81	72	-	Tubules	Tubules	↓
<i>Laburnum anagyroides</i> (DT) ¹⁴	2.41	0.42	18	133	113	13.02	5.76	44	115	76	11.13	Platelets	Platelets	↓
<i>Larix decidua</i> (DT) ²¹	6.58	3.43	52	114	105	8.39	3.06	36	84	76	-	Tubules	Tubules	↓
<i>Larix kaempferi</i> (DT) ²¹	5.95	5.69	96	111	112	7.29	2.80	38	101	87	-	Tubules	Tubules	↓
<i>Ligustrum ovalifolium</i> (DS) ²⁰	4.52	2.01	44	85	79	13.37	8.03	60	60	71	-	Thin film	Thin film	↔
<i>Ligustrum vulgare</i> (DS) ²⁰	5.51	2.83	51	95	98	12.94	6.22	48	85	74	-	Thin film	Thin film	↔
<i>Liquidambar styraciflua</i> (DT) ²	3.61	1.70	47	98	98	7.88	3.49	44	83	67	-	Platelets	Thin film	Δ
<i>Liriodendron tulipifera</i> (DT) ¹⁸	2.44	0.56	23	135	133	7.80	3.71	48	125	93	-	Crusts	Crusts	↓
<i>Lonicera periclymenum</i> (CL)⁸	-	-	-	134	123	9.83	5.29	54	105	93	3.92	Tubules	Tubules	↔
<i>Lonicera tatarica</i> (DS) ⁸	3.74	1.50	40	137	136	17.30	11.31	65	112	58	-	Tubules	Tubules	↔
<i>Lonicera xylosteum</i> (DS) ⁸	3.62	1.75	48	140	134	19.70	11.28	57	112	69	8.83	Tubules	Tubules	↓
<i>Magnolia kobus</i> (DT) ²⁰	4.89	2.69	55	101	104	12.83	5.38	42	77	64	5.75	Platelets	Platelets	↓
<i>Mahonia aquifolium</i> (EB)⁵	-	-	-	132	86	12.42	6.99	56	89	69	-	Thin film	Platelets	Δ
<i>Malus sylvestris</i> (DT) ²⁴	7.33	3.06	42	93	81	12.62	8.95	71	87	76	+++	Thin film	Platelets	Δ
<i>Mespilus germanica</i> (DT) ²⁴	7.91	5.67	72	92	85	22.70	18.47	81	71	74	21.33	Thin film	Thin film	↔
<i>Picea abies</i> (EN)²¹	-	-	-	100	104	11.78	2.65	23	66	82	-	Tubules	Tubules	↓
<i>Picea pungens glauca</i> (EN)²¹	-	-	-	80	82	17.97	6.29	35	88	93	-	Tubules	Tubules	↓
<i>Pinus nigra</i> (EN)²¹	-	-	-	76	86	12.30	4.14	34	75	77	-	Crusts	Tubules	Δ
<i>Platanus × acerifolia</i> (DT) ²²	4.92	1.79	36	99	83	7.01	3.76	54	55	80	2.08	Platelets	Thin film	Δ
<i>Populus alba</i> (DT) ²⁵	2.40	0.45	19	93	85	3.08	1.19	39	75	76	-	Thin film	Thin film	↔

PLANT SPECIES	JUNE					SEPTEMBER					Epicuticular wax structures			
	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	Trichome density	EWS June	EWS September	EWS Change
<i>Prunus avium</i> (DT) ²⁴	5.39	2.65	49	87	86	17.88	12.90	72	74	64	3.17	Platelets	Platelets	↓
<i>Prunus laurocerasus</i> (EB) ²⁴	-	-	-	85	85	9.60	2.82	29	81	78	-	Thin film	Platelets	Δ
<i>Prunus padus</i> (DS) ²⁴	5.89	3.65	62	126	92	18.15	12.53	69	96	69	0.13	Platelets	Platelets	↓
<i>Prunus spinosa</i> (DS) ²⁴	7.07	1.17	17	100	86	20.33	12.10	59	82	66	8.17	Thin film	Platelets	Δ
<i>Pseudotsuga menziesii</i> (EN) ²¹	-	-	-	90	84	21.05	8.08	38	91	76	-	Tubules	Tubules	↔
<i>Quercus ilex</i> (EB) ¹⁵	13.71	10.5	77	130	71	24.08	17.26	72	100	66	+++	Platelets	Platelets	↓
<i>Quercus palustris</i> (DT) ¹⁵	5.56	3.05	55	99	87	8.48	3.42	40	57	65	-	Crusts	Crusts	↔
<i>Quercus petraea</i> (DT) ¹⁵	5.93	3.70	62	133	93	22.10	16.74	76	110	75	13.38	Platelets	Platelets	↓
<i>Quercus robur</i> (DT) ¹⁵	4.26	2.46	58	131	119	21.89	12.44	57	94	80	-	Platelets	Platelets	↓
<i>Quercus rubra</i> (DT) ¹⁵	5.36	3.70	69	122	104	14.67	6.08	41	76	75	-	Platelets	Platelets	↓
<i>Rhamnus cathartica</i> (DS) ²³	8.41	4.18	50	84	68	16.60	9.77	59	76	68	1.17	Platelets	Platelets	↓
<i>Rhamnus frangula</i> (DS) ²³	4.98	3.10	62	91	83	20.16	14.21	70	62	71	-	Thin film	Crusts	Δ
<i>Rhododendron</i> (EB) ¹³	-	-	-	58	76	15.06	7.24	48	55	59	-	Platelets	Platelets	↓
<i>Robinia pseudoacacia</i> (DT) ¹⁴	1.71	1.34	78	141	132	4.41	1.85	42	125	123	31.79	Platelets	Platelets	↔
<i>Rosa canina</i> (DS) ²⁴	5.66	0.96	17	97	123	9.22	6.10	66	89	103	-	Crusts	Crusts	↔
<i>Rosa glauca</i> (DS) ²⁴	3.14	1.18	37	131	129	7.77	2.04	26	126	124	-	Crusts	Crusts	↔
<i>Rosa pimpinellifolia</i> (DS) ²⁴	5.00	3.14	63	128	128	16.91	13.50	80	90	80	-	Platelets	Platelets	↔
<i>Rosa rubiginosa</i> (DS) ²⁴	7.31	6.10	83	69	89	24.61	16.44	67	59	66	9.88	Thin film	Thin film	↔
<i>Rosa rugosa</i> (DS) ²⁴	5.76	4.30	75	124	81	28.90	24.16	84	100	58	28.88	Crusts	Platelets	Δ
<i>Salix alba</i> (DT) ²⁵	3.78	2.00	53	125	74	14.68	8.27	56	110	67	19.83	Crusts	Crusts	↓
<i>Salix aurita</i> (DS) ²⁵	4.60	3.19	69	134	120	22.80	14.85	65	126	68	16.21	Platelets	Platelets	↔
<i>Salix caprea</i> (DT) ²⁵	4.74	2.53	54	133	71	12.27	7.74	63	125	64	11.13	Crusts	Crusts	↓
<i>Salix cinerea</i> (DS) ²⁵	4.95	3.49	70	130	85	18.72	11.32	60	124	83	20.46	Crusts	Crusts	↔
<i>Salix purpurea</i> (DS) ²⁵	1.34	0.13	9	130	132	8.07	3.05	38	121	112	-	Platelets	Platelets	↔
<i>Salix repens</i> (DS) ²⁵	6.00	3.84	64	129	69	21.89	12.11	55	123	81	38.42	Crusts	Crusts	↔
<i>Salix rosmarinifolia</i> (DS) ²⁵	5.18	3.05	59	137	69	17.71	9.45	53	128	78	+++	Platelets	Platelets	↔
<i>Salix viminalis</i> (DS) ²⁵	5.53	3.48	63	130	85	15.78	11.62	74	128	84	16.96	Thin film	Platelets	Δ
<i>Sambucus nigra</i> (DS) ¹	4.92	2.46	50	56	64	15.58	11.54	74	54	64	1.38	Thin film	Thin film	↔
<i>Sorbus aria</i> (DT) ²⁴	7.36	4.57	62	139	82	30.21	23.75	79	130	61	+++	Thin film	Thin film	↔
<i>Sorbus aucuparia</i> (DT) ²⁴	10.13	4.67	46	131	78	17.47	9.93	57	86	75	3.29	Platelets	Tubules	Δ
<i>Sorbus intermedia</i> (DT) ²⁴	13.87	9.94	72	135	79	23.53	26.02	-	110	63	+++	Thin film	Thin film	↔

PLANT SPECIES	JUNE					SEPTEMBER					Epicuticular wax structures			
	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	SIRM Un-washed	SIRM Washed	Immobilized fraction of particles	DCA (AB)	DCA (AD)	Trichome density	EWS June	EWS September	EWS Change
<i>Sorbus torminalis</i> (DT) ²⁴	5.12	3.93	77	84	77	13.49	9.44	70	61	59	10.46	Platelets	Platelets	↔
<i>Symphoricarpos</i> × <i>chenaultii</i> (DS) ⁸	4.34	0.72	17	140	135	12.70	9.05	71	126	92	19.46	Tubules	Tubules	↔
<i>Syringa vulgaris</i> (DS) ²⁰	4.59	1.79	39	56	79	15.59	8.87	57	56	63	-	Thin film	Thin film	↔
<i>Taxus baccata</i> (EN) ²⁸	-	-	-	94	75	11.69	5.21	45	86	66	-	Tubules	Tubules	↔
<i>Thuja plicata</i> (EN) ¹¹	-	-	-	104	83	19.54	15.84	81	93	64	-	Tubules	Tubules	↓
<i>Tilia cordata</i> (DT) ¹⁹	3.61	2.03	56	74	66	12.76	8.57	67	70	76	-	Platelets	Platelets	↓
<i>Tilia platyphyllos</i> (DT) ²⁴	5.61	1.47	26	84	59	21.39	18.40	86	61	59	6.75	Thin film	Platelets	Δ
<i>Ulmus glabra</i> (DT) ²⁹	6.12	5.06	83	85	85	27.06	22.80	84	67	55	10.29	Platelets	Platelets	↔
<i>Viburnum lantana</i> (DS) ¹	15.74	13.9	88	79	76	39.77	34.51	87	58	71	8.38	Platelets	Platelets	↔
<i>Viburnum opulus</i> (DS) ¹	5.40	5.10	94	95	74	31.01	22.92	74	77	71	22.29	Thin film	Platelets	Δ

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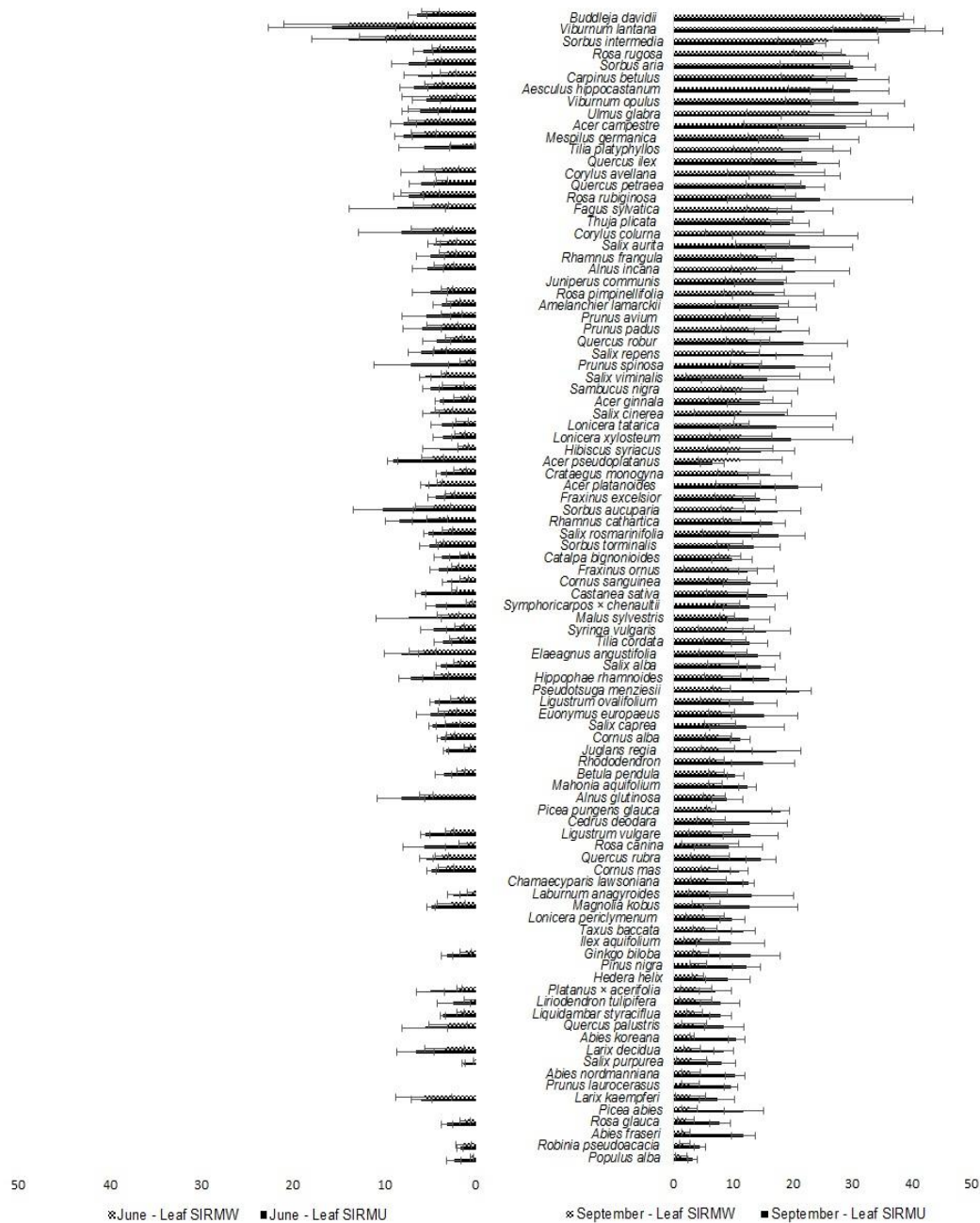
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# Plant families:	1 = Adoxaceae	2 = Altingiaceae	3 = Apiaceae	4 = Aquifoliaceae
5 = Berberidaceae	6 = Betulaceae	7 = Bignoniaceae	8 = Caprifoliaceae	9 = Celastraceae
10 = Cornaceae	11 = Cupressaceae	12 = Elaeagnaceae	13 = Ericaceae	14 = Fabaceae
15 = Fagaceae	16 = Ginkgoaceae	17 = Juglandaceae	18 = Magnoliaceae	19 = Malvaceae
20 = Oleaceae	21 = Pinaceae	22 = Platanaceae	23 = Rhamnaceae	24 = Rosaceae
25 = Salicaceae	26 = Sapindaceae	27 = Scrophulariaceae	28 = Taxaceae	29 = Ulmaceae

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351 **Fig. 1** Mean leaf-area normalized leaf SIRMU - unwashed leaves (black bars, μA) and leaf SIRMw - washed
352 leaves (textured bars, μA) from ($\sim n = 3 - 5$) replicates of the selected perennial urban plant species ($n =$
353 96) in June (left) and September (right) 2016. Error bars indicate the standard deviation. Note: leaves of
354 evergreen needle/scale-like, evergreen broadleaf and climber species sampled in June were excluded
355 because their leaves were developed in the previous growing season and were about one-year-old at the
356 time of sampling. Leaf SIRM data of unwashed leaves from June and September 2016 have been
357 previously reported by Muhammad et al. (2019).

In June, the median leaf SIRM_w for deciduous (broadleaf/needle-like) trees and shrubs was 2.8 μ A, and 3.1 μ A respectively. One-way analysis of variance (ANOVA) revealed that leaf SIRM_w of deciduous (broadleaf/needle-like) trees and shrubs were not significantly different [$F_{1, 73} = 0.002$, $p = 0.963$] (Table 2). In September, the median leaf SIRM_w was, 4.7 μ A for climbers, 5.5 μ A for evergreen needle/scale-like, 7.0 μ A for evergreen broadleaf, 9.4 μ A for the deciduous (broadleaf/needle-like) trees, and 11.3 μ A for the deciduous broadleaf shrub species. An ANOVA indicated that the deciduous (broadleaf/needle-like) trees ($p = 0.027$) and deciduous broadleaf shrub species ($p = 0.012$) differed significantly from evergreen needle/scale-like species. No significant differences in leaf SIRM_w were observed between other functional plant types (Fig. 2a). A significant increase ($p < 0.001$) in leaf SIRM_w from June to September was observed for deciduous (broadleaf/needle-like) tree and deciduous broadleaf shrub species (Fig. 2b).

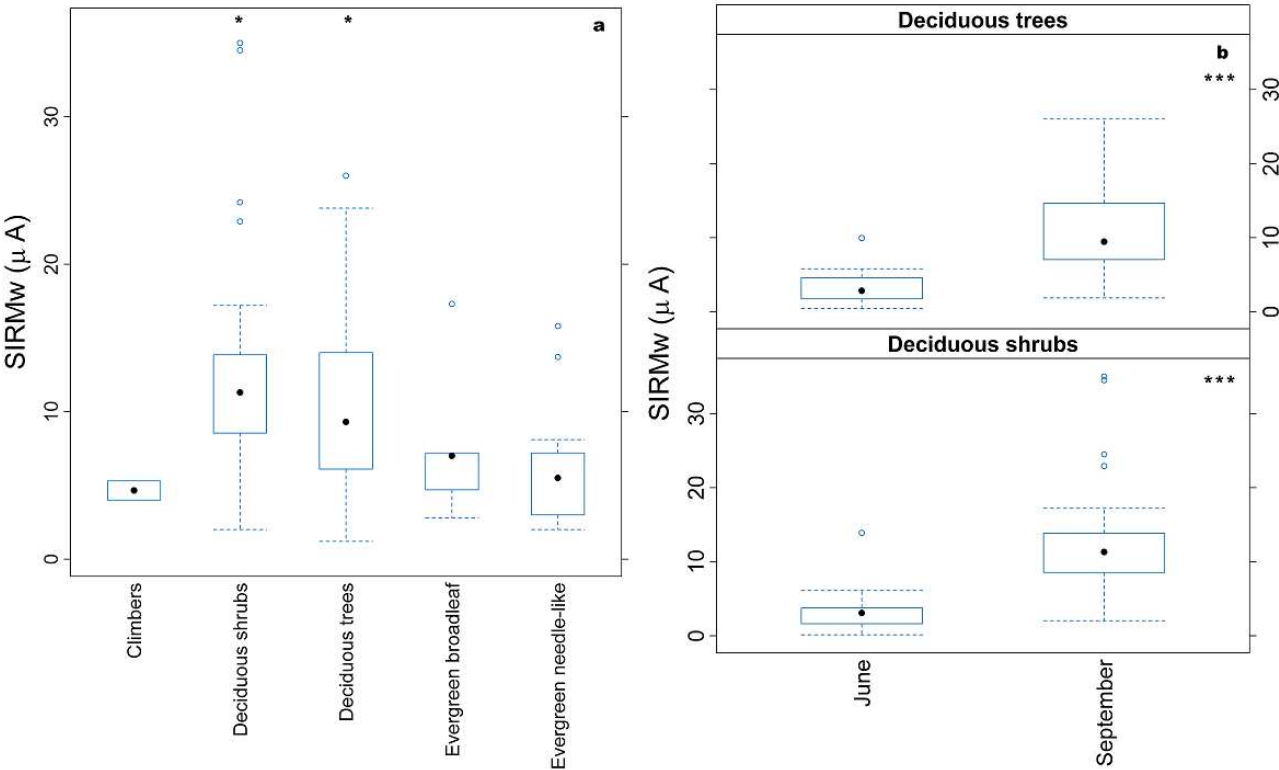


Fig. 2 (a) Leaf SIRM_w by functional plant types ($n = 5$) consisting of all investigated perennial urban plant species ($n = 96$) in September. Significant differences in leaf SIRM_w of deciduous (broadleaf/needle-like) tree and shrub species from evergreen needle/scale-like species using Tukey's HSD post-hoc test with 95 % confidence level ($p < 0.05$) are indicated with an asterisk "*" **(b)** Box plots of leaf SIRM_w (washed leaf samples) for deciduous (broadleaf/needle-like) trees and deciduous broadleaf shrubs ($n = 77$) in June and September 2016. Results of the paired sample t-test indicate a significant ("***": $p < 0.001$) increase in leaf SIRM_w in September. The bars extending vertically from both sides of the box are the lower and upper whiskers indicating the minimum and maximum leaf SIRM_w. The lower end of the box indicates the first quartile; the top indicates the third quartile. The black filled dots are median leaf SIRM_w, and the hollow circles are the outliers.

3.2 Absolute difference in SIRM (Δ SIRM) and the immobilized fraction of PM

In June, the absolute difference (Δ SIRM) between leaf SIRM of unwashed leaves (SIRM_u) to leaf SIRM of washed leaves (SIRM_w) for deciduous (broadleaf/needle-like) trees and shrubs species ($n = 77$) ranged

between 0.2 to 5.9 μA with the lowest and the highest ΔSIRM observed on leaves of *L. kaempferi* and *P. spinosa* respectively. Leaves of other plant species such as *V. opulus*, *R. pseudoacacia*, *C. alba*, *U. glabra*, *S. torminalis* also showed a low ΔSIRM whereas *S. aucuparia*, *R. canina*, *A. pseudoplatanus*, *M. sylvestris*, and *R. cathartica* showed a high ΔSIRM . In September, considering all investigated plant species ($n = 96$), the ΔSIRM ranged between 0.2 to 12.9 μA with the lowest and the highest ΔSIRM observed on leaves of *C. bignonioides* and *P. menziesii* respectively. Leaves of other plant species which also showed a low ΔSIRM in September were, for example, *P. alba*, *A. glutinosa*, *R. pseudoacacia*, *B. davidii*, and *T. platyphyllos* whereas *P. glauca*, *A. platanooides*, *J. regia*, *S. repens*, and *A. fraseri* showed a high ΔSIRM . The median difference between SIRM_{U} and SIRM_{W} was 2.1 μA in June and 5.3 μA in September.

In June, the immobilized fraction of PM ranged between 9 to 96 % with the lowest fraction observed on leaves of *S. purpurea* and highest on leaves of *L. kaempferi*. Several other plant species such as *P. spinosa*, *S. chenaultii*, *R. canina*, *L. anagyroides*, and *P. alba* also showed a low immobilized fraction of particles whereas *V. opulus*, *V. lantana*, *R. rubiginosa*, *U. glabra*, and *R. pseudoacacia* showed a high immobilized fraction of particles on their leaf surfaces. In June, the immobilized fraction of particles was indicated as not significantly different between deciduous (broadleaf/needle-like) tree and deciduous shrub species [$F_{1, 73} = 0.025$, $p = 0.874$]. In September, the immobilized fraction of particles ranged from 17 to 97 % with the lowest fraction observed for *A. fraseri* and the highest for *C. bignonioides*. Leaves of plant species such as *P. abies*, *R. glauca*, *A. nordmanniana*, *A. koreana*, and *P. laurocerasus* showed a low immobilized fraction of particles whereas leaves of *B. davidii*, *V. lantana*, *T. platyphyllos*, *C. avellana*, and *U. glabra* showed a high immobilized fraction of particles. In September, the ANOVA indicated significant differences [$F_{4, 89} = 7.55$, $p < 0.001$] in the immobilized fraction of particles between functional plant types. The immobilized fraction of particles was significantly higher on leaves of deciduous (broadleaf/needle-like) tree ($p < 0.001$) and deciduous broadleaf shrub species ($p < 0.001$) compared to the immobilized fraction observed on leaves of evergreen needle/scale-like species. No significant differences in the fraction of immobilized particles were observed between other functional plant types.

3.3 Leaf SIRM: the effect of washing treatment, time and leaf micro-morphology

The median values of leaf SIRM_{W} were 3.1 μA in June ($n = 77$) and 9.1 μA in September ($n = 96$). The median values of leaf SIRM_{U} were 5.2 μA in June and 14.9 μA in September. The results of the linear mixed effect regression (LMER) model (Table 2) indicated a significant effect of time on leaf SIRM. A significantly higher leaf SIRM was obtained in September compared to June. A significant effect of treatment was indicated, the leaf SIRM of washed leaves (SIRM_{W}) was significantly lower than the leaf SIRM of unwashed leaves (SIRM_{U}). Concerning the leaf-micromorphology, the effect of trichome density on leaf SIRM was significant and positive, while the effect of DCA on leaf SIRM was significant and negative (Table 2). The effect of epicuticular wax structures (EWS) types on leaf SIRM was marginally significant, plant species with EWS type of thin-film showed a significantly higher leaf SIRM compared to leaf SIRM of plant species with EWS type of crusts, platelets, and tubules. The interaction effects between time, treatment, and leaf characteristics were not significant and thus were excluded from the model.

The LMER applied on the immobilized fraction of PM indicated a significant effect of leaf wettability and trichome density (Table 2). The effect of time (i.e., June, September) and the EWS types on the fraction of immobilized PM were not significant and hence were removed from the LMER model. So although the median fraction of immobilized particles was 55 % in June ($n = 77$), and increased to 60 % in September ($n = 96$), this difference was not significant. The immobilized fraction of PM increased with increasing leaf trichome density. In June, the median immobilized fraction of PM was 52 % for plant species with no leaf trichomes and 62 % for plant species with leaf trichomes. In September, the median immobilized fraction of PM was 48 % for plant species with no leaf trichomes and 70 % for plant species with leaf trichomes. The fraction of immobilized PM decreased with an increasing DCA. Leaves with low wettability showed a low immobilized fraction of PM compared to leaves with high wettability.

Table 2

ANOVA of fixed factors in the linear mixed-effect regression (LMER) model on response variable $\ln(\text{SIRM})$ and immobilized fraction of PM. The random effect is plant-id as and time (June, September), treatment (unwashed and washed leaves), epicuticular wax structure (EWS: crusts, platelets, thin film and tubules), leaf wettability and trichome density as fixed effects. The LMER on the immobilized fraction of PM does not include treatment as a fixed effect. Significant effects ($p \leq 0.05$) are shown in bold

Response	Source of variation	F value	p-value
Leaf SIRM (n = 384)	Time	82.63	< 0.001
	Treatment	111.10	< 0.001
	EWS type	2.98	0.032
	Leaf wettability (DCA)	26.90	< 0.001
	Trichome density	16.75	< 0.001
Immobilized fraction of PM (n= 192)	Leaf wettability (DCA)	6.78	0.010
	Trichome density	4.47	0.037

3.4 The effect of change in EWS types with time on SIRM_D

In June, the leaf SIRM_W by EWS type ranged between 3.1 to 3.5 μA with the lowest and the highest leaf SIRM_W observed for EWS type of platelets and tubules respectively. In September, the leaf SIRM_W by EWS type ranged between 6.3 to 15.2 μA with the lowest and the highest leaf SIRM_W observed for EWS type of tubules and crusts respectively (Fig 3a). A change in EWS type or clustering of the wax crystals was observed from June to September (Table 1) for more than half of the investigated species ($n = 54$). Few plant species ($n = 23$) such as *M. sylvestris*, *C. alba*, *H. syriacus*, *V. opulus*, and *P. spinosa* were observed to have a change in EWS type from June to September. One-third of the investigated plant species ($n = 31$) mainly with EWS type of platelets and tubules such as *L. anagyroides*, *M. kobus*, *P. glauca*, and *Q. robur* showed no change in EWS type but a decrease in the clustering of wax crystals or an increase in gaps of the wax crystals. Almost half of the investigated plant species ($n = 42$) did not show a change in either EWS type or clustering. One-way analysis of variance (ANOVA) on the leaf SIRM_D (i.e., SIRM_W in September minus SIRM_W in June) for deciduous (broadleaf/needle-like) tree and shrub species ($n = 77$), indicated no significant effect of a change in EWS with the difference in leaf SIRM_W from June to September [$F_{2,74} = 2.62$, $p = 0.08$] (Fig. 3b).

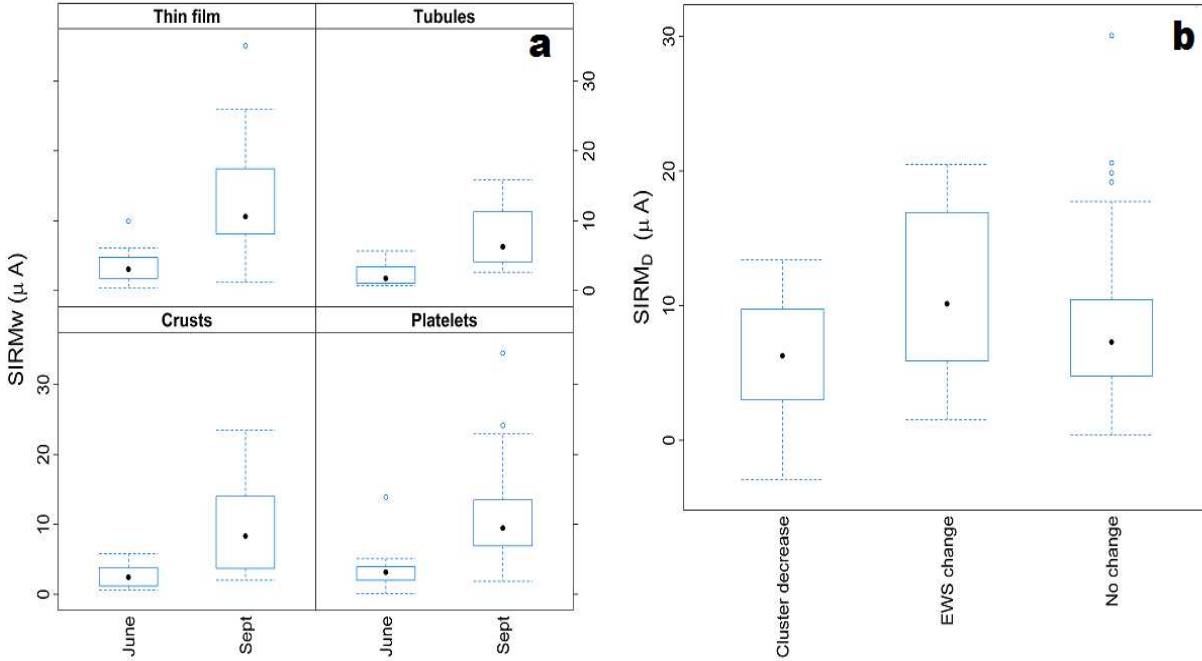


Fig. 3 (a) Box plots of SIRM_w (washed leaves SIRM) by epicuticular wax structure types (thin film, tubules, crusts, platelets) in June and September **(b)** The difference in leaf SIRM_w from June to September denoted as SIRM_b for leaves of deciduous (needle-like/broadleaf) tree, and shrubs species ($n = 77$) with a change in EWS type from June to September ($n = 23$) or decrease in clustering of wax crystals within the same EWS type ($n = 31$) or no change either in EWS type or clustering ($n = 42$). The bars extending vertically from both sides of the box are the upper and lower whisker indicating the minimum, and maximum, the box indicates the first quartile while the top indicates the third quartile. The black filled dots indicate median values. The hollow circles above the upper whisker indicate outliers.

3.5 Correlation between leaf SIRM_u and leaf SIRM_w

Pearson correlations between natural log transformed $\ln(\text{SIRM}_u)$, i.e., leaf SIRM of unwashed leaves and natural log transformed $\ln(\text{SIRM}_w)$, i.e., leaf SIRM of washed leaves were computed. A significant correlation was indicated for both June [$r = 0.84$, $n = 77$, $p < 0.001$] and September [$r = 0.89$, $n = 96$, $p < 0.001$] (Fig. 4a and b). The Pearson correlation were also computed on $\ln(\text{SIRM}_u)$ and $\ln(\text{SIRM}_w)$ for plant species with and without leaf trichomes in September and for the different EWS types in both June and September. A significant but weak correlation ($r = 0.76$, $n = 45$, $p < 0.001$) was indicated between $\ln(\text{SIRM}_u)$ and $\ln(\text{SIRM}_w)$ for plant species with no leaf trichomes (Fig. 4b) compared to the significant and steeper correlation obtained for plant species with leaf trichomes ($r = 0.95$, $n = 51$, $p < 0.001$) (Fig. 4b). The Pearson correlation between $\ln(\text{SIRM}_u)$ and $\ln(\text{SIRM}_w)$ for each EWS type was significant and fairly similar between EWS types i.e., crusts ($r = 0.92$, 0.90), platelets ($r = 0.89$, 0.87), thin film ($r = 0.84$, 0.90), and tubules ($r = 0.93$, 0.83) in both June and September respectively (Fig. 4c, d).

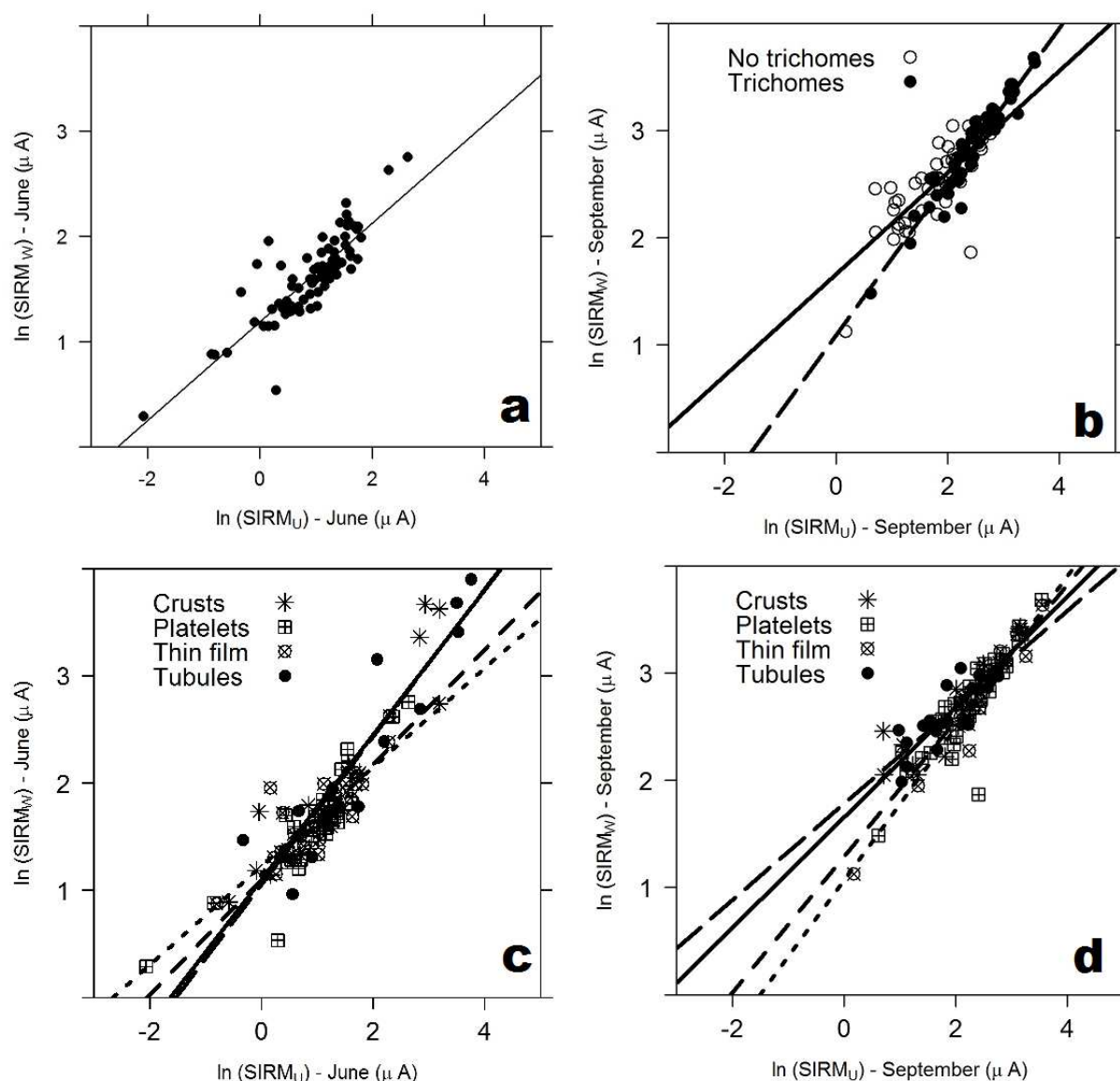


Fig. 4 Scatter plot with fitted regression lines showing natural log transformed leaf SIRM of unwashed leaf samples $\ln(\text{SIRM}_U)$ plotted against natural log transformed leaf SIRM of washed leaf samples $\ln(\text{SIRM}_W)$ in (a) June for deciduous broadleaf species ($y = 1.37x - 0.56$, $r = 0.84$, $n = 77$, $p < 0.001$) (b) in September for plant species with no leaf trichomes ($y = 1.33x - 0.63$, $r = 0.76$, $n = 45$, $p < 0.001$) and with leaf trichomes ($y = 1.39x - 0.69$, $r = 0.95$, $n = 51$, $p < 0.001$) (c) for epicuticular wax structure (EWS) types of crusts ($y = 1.19x - 0.47$, $r = 0.92$), platelets ($y = 1.54x - 0.64$, $r = 0.89$), thin film ($y = 1.39x - 0.60$, $r = 0.84$), and tubules ($y = 1.27x - 0.49$, $r = 0.93$) in June (d) for EWS types of crusts ($y = 1.59x - 0.98$, $r = 0.90$), platelets ($y = 1.21x - 0.45$, $r = 0.87$), thin film ($y = 1.32x - 0.54$, $r = 0.90$) and tubules ($y = 1.56x - 0.97$, $r = 0.83$) in September. Lines shown are regression lines – “solid” for EWS type of crusts, “dashed” for platelets, “dotted” for thin film and “long dash” for tubules.

4. Discussion

4.1 Differences in leaf $SIRM_w$ and $SIRM_u$ between plant species and functional plant types

Our study enables an impartial comparison of leaves of perennial plant species in immobilizing PM because of the common-garden set-up and the standardized washing procedure using a vigorous vortex shaker at a consistent speed and for a precise duration. The ferromagnetic and magnetizable component of PM after leaf washing treatment was estimated using saturation isothermal remanent magnetization and expressed as ($SIRM_w$). The SIRM signal should be considered as an indicator of anthropogenic PM rather than as total PM loadings on leaf surfaces. The link between the SIRM signal and traffic-related PM has been established in previous studies of (e.g., Maher et al. (2008), Kardel et al. (2012), and Sant'Ovaia et al. (2012). Furthermore, industrial activities (Hansard et al. 2011; Zhang et al. 2012) and railway lines (Moreno et al. 2003; Lorenzo et al. 2006) have also been recognized as contributing sources of magnetic PM. When comparing leaf $SIRM_w$ of investigated functional plant types ($n = 5$) with leaves approximately of similar age, the results of ANOVA (Fig. 2a) indicated that leaf $SIRM_w$ of deciduous (broadleaf/needle-like) tree and shrub species were significantly higher than leaf $SIRM_w$ of evergreen needle/scale-like species. There were no significant differences between leaf $SIRM_w$ of evergreen needle/scale-like, evergreen broadleaf, and climber species in September (Fig. 2a). The disparity in PM immobilization between functional plant types can be attributed to their micro-morphological leaf characteristics such as trichome density, leaf wettability, and epicuticular wax structure types which are discussed in detail in § 4.2.

Concerning leaf $SIRM_w$ in absolute numbers, the results indicate that leaves of *B. davidii*, *V. lantana*, and *S. intermedia* were the most effective in PM immobilization while leaves of *P. alba*, *R. pseudoacacia* and *A. fraseri* were the least effective. In terms of leaf $SIRM_u$, the most effective plant species in PM accumulation were *V. lantana*, *B. davidii*, and *V. opulus* and the least effective plant species were *P. alba*, *R. pseudoacacia* and *A. pseudoplatanus* (Table 1). It is worth noting here that leaves of *B. davidii*, and *V. lantana* showed a high leaf $SIRM_u$ as well as a high leaf $SIRM_w$ similarly, leaves of *P. alba* and *R. pseudoacacia* showed a low leaf $SIRM_u$ as well as a low leaf $SIRM_w$ suggesting that PM immobilized on leaf surfaces would be in proportion to the net accumulated PM. Indeed the natural log transformed leaf $SIRM_u$ and leaf $SIRM_w$ correlated significantly (Fig 4a and b) few plant species deviated from the regression line. For example *A. fraseri*, *A. koreana*, *A. nordmanniana* and *P. abies* showed a high leaf $SIRM_u$ but a low $SIRM_w$ resulting in an immobilized fraction of PM between 17 to 29 %. Based on these results it can be concluded that $SIRM_u$ in general can be a good indicator to determine the effectiveness of a plant species in immobilization of PM.

The above-mentioned plant species are some examples exhibiting leaf $SIRM_w$ in proportion to leaf $SIRM_u$, for complete results refer to Table 1. The immobilized fraction of PM on leaf surfaces of plant species ranged between 9 to 96% in June and 17 to 97% in September. It is noteworthy that the leaf-washing protocol used in our study probably does not reflect the particle load which can be blown-off by wind or washed-off after a rainfall event. But it is rather a technique to estimate the absolutely immobilized particles which were either held within the epicuticular wax layer, affixed on leaf trichomes or hyphae of fungi or trapped inside the stomatal cavities. Findings from our study concerning the immobilized fraction of PM determined after leaf washing treatment were overall in agreement with other studies (e.g., Dzierżanowski et al. 2011; Sæbø et al. 2012; Popek et al. 2013; Hofman et al. 2014). However, a direct comparison of our results with that of previous studies can be difficult because of the following reasons (a) the duration of treatment, i.e., leaf washing in this study was precisely 3 minutes while in previous studies the duration of leaf washing ranged between 60 seconds to ≤ 2 minutes (Dzierżanowski et al. 2011; Sæbø et al. 2012; Popek et al. 2013; Hofman et al. 2014; Przybysz et al. 2014); (b) the mechanical and repeatable leaf washing methodology applied in the present study was in contrast to the leaf washing methodology of former studies, for example, Hofman et al. (2014) removed the surface accumulated particles by hand washing, as such the scrubbing intensity may vary between leaf samples and induce bias when comparing between plant species effectiveness in PM immobilization. Previous studies, for example, Popek et al. (2013), Terzaghi et al. (2013), Przybysz et al. (2014) performed the leaf washing treatment using either a

basic shaker or a sonic bath. However, the shaking intensities were not reported, and hence, a direct comparison of results may be difficult.

The results of this study indicated that, on average, more than half (i.e., 60 %) of the magnetic signal still remained after vigorous washing, but a large variation exists between species. The immobilized fraction of PM on leaves of *P. x acerifolia* was approximately 54 % in our study while Hofman et al. (2014) reported the leaf SIRM of washed leaves of *P. x acerifolia* accounted for 38 % of the total SIRM signal. The reason for the discrepancy between the two studies could be due to differences in sampling locations. The study of Hofman et al. (2014) was conducted in a traffic intense urban street canyon in the city of Antwerp, Belgium while our study was also performed in the same city but at a background location away from a direct pollution source. Popek et al. (2013) estimated that 40 % of the particles were immobilized within the wax layer of leaves. One of the reason for the reported disparity in an overall immobilized fraction of PM in this study (~ 60%) compared to that of Popek et al. (2013) (~ 40%) may have resulted due to differences in sample size, i.e., the number of investigated plant species (n = 96, n = 13) respectively. Moreover, the differences in leaf characteristics (i.e., trichome density, leaf wettability) of the investigated plant species need to be taken into account which may have affected the overall immobilized fraction of PM. Another possible reason for discrepancy could be because leaf samples in the study of Popek et al. (2013) were collected at the end of the growing season i.e., October whereas in the present study leaves of investigated plant species were sampled and analyzed for PM immobilization in June and early September hence a low immobilized fraction of PM was reported by Popek et al. (2013). Hofman et al. (2014) analyzed the seasonal dynamics of PM immobilization on leaves of *P. x acerifolia* via SIRM analyses and reported a decline in October-November due to leaf senescence. The authors revealed that when leaf senesce, i.e., discoloration of leaf due to loss of chlorophyll, the leaves cease epicuticular wax production, which may cause a reduction in PM immobilization as was observed in the study of Popek et al. (2013). Hence, it would be desirable to analyze the PM immobilization abilities of plant species before leaf senescence. However, when taking into account the same investigated plant species (e.g., *G. biloba*, *S. nigra*, *Q. rubra*) in this study and that of Popek et al. (2013), the immobilized fraction of PM was observed to be fairly identical. Hence, it can be concluded that PM accumulation and immobilization on leaf surfaces are in conformity of their leaf micro-morphology.

4.2 The effect of leaf micro-morphology on washed leaf SIRM ($SIRM_w$) and fraction of immobilized PM

The trichome density significantly affected the total leaf SIRM ($SIRM_U$ and $SIRM_w$): with higher trichome density, more PM was accumulated and immobilized by the leaves. The median leaf $SIRM_w$ of plant species with dense leaf trichomes in September was higher (i.e., 13.6 μA) compared to median leaf $SIRM_w$ of plant species with no leaf trichomes (i.e., 6.0 μA). In addition, plant species such as *B. davidii*, *S. intermedia*, *S. aria*, and *Q. ilex*, for which trichome density was so high that it could not be determined were found to have a high (~ 17 - 35 μA) leaf $SIRM_w$ (Table 1). The lack of significant interaction effect between trichome density and treatment, (i.e., unwashed and washed leaves) suggests that re-suspension of particles through leaf washing treatment was not affected by trichome density on top of an effect of trichome density on deposition. However, analysis of the immobilized fraction shows a significant effect of trichomes (Table 2) illustrating that for the same PM accumulation, plant species with leaf trichomes were more effective in immobilizing PM compared to plant species with no leaf trichomes. Plant species with trichomes displayed a high (~ 70 %) immobilized fraction of PM compared to plant species with no trichomes (~ 48%). This different immobilization behaviour due to trichome presence is confirmed by the different relationships between the natural log transformed leaf $SIRM_U$ and leaf $SIRM_w$ for plant species with (n = 51) and without (n = 45) trichomes: a steeper relationship was obtained for plant species with leaf trichomes compared to leaves with no trichomes (Fig 4b). This illustrates that for the same PM accumulation the PM immobilization is enhanced in the presence of leaf trichomes. The effect of trichomes in PM immobilization has been corroborated by several other authors (Beckett et al. 2000, Sæbø et al. 2012; Popek et al. 2013). The presence of trichomes increases the surface area on the leaves where particles can be deposited (De

Nicola et al. 2008). Moreover, the boundary layer resistance for leaves with trichomes is decreased compared to leaves with no trichomes, which makes PM prone to being re-suspended but also enhancing the capture of PM (Bakker et al. 1999). Our study, however, indicates that immobilization of PM is enhanced in leaves with trichomes not only by a higher supply of particles to the leaf surface but also by a higher immobilization efficiency by the leaf, since from the same amount of accumulated PM a larger amount is retained by the leaf due to the presence of trichomes. Possibly the trichomes hold the particles strongly or prevent the wash water to make contact with the epidermis of the leaf and making them more water repellent, and hence prevent the deposited particles from being washed away.

Plant species with low leaf wettability (e.g., *G. biloba*, *L. tulipifera*, *R. pseudoacacia*, *R. glauca*, and *S. purpurea*), were found to have an immobilized fraction of PM between (~ 26 – 48 %) compared to leaves, (e.g., *T. platyphyllos*, *U. glabra*, *C. colurna*, *S. nigra*, *V. lantana*, *V. opulus*) with high leaf wettability and displaying a high immobilized fraction of PM between (~ 74 – 87 %) on their leaf surfaces of the net deposited particles, (i.e., SIRM_U, Table 1). Leaves with a low wettability can be anti-adhesive concerning PM immobilization (Neinhuis and Barthlott 1998). The contaminating particles on leaves with low wettability do not adhere to the leaf surface as much but adhere more to the water droplets and may be removed when water droplets roll off the leaf surface (Neinhuis and Barthlott 1997). Fogg (1947) indicated that low leaf wettability can be due to the surface roughness of the leaf, mainly caused by convex epidermal cells, trichomes, and epicuticular wax structures (Neinhuis and Barthlott 1997). We did not characterize the type of epidermal cells on leaves of investigated plant species but observed that leaves with trichomes showed a remarkable surface roughness resulting in low wettability. A droplet of water when deposited on leaves with trichomes, bends the trichomes but the stiffness of the trichomes prevents contact with the leaf surface as was observed in the present study (Fig. 5) and by Otten and Herminghaus (2004). Outcomes from our study also indicated that plant species such as *B. davidii* with a high trichome density and low leaf wettability showed a high immobilized fraction of PM, i.e., 92 % on their leaf surfaces after leaf washing treatment whereas leaves, for example, of *Q. rubra* and *Rhododendron* with no trichomes and a high leaf wettability showed a low immobilized fraction of PM, (i.e., 41 – 48 %) on their leaf surfaces after leaf washing. Hence, it can be concluded that a combination of leaf traits may be responsible for an increase in particle immobilization and not limited in the entirety of any single leaf trait.

The SIRM_w was significantly affected by epicuticular wax structure type. Plant species, for example, *M. germanica*, *S. aria*, *S. intermedia*, *V. opulus* with thin-film as EWS type generally showed a higher leaf SIRM_w (Table 1) compared to leaves of plant species with tubules as EWS type. A possible explanation may be that crystalline protuberances similar to that of tubules with elongated and cylindrical structures result in low adhesive force which can make the adherence of particles difficult due to the reduced contact area (Wang et al. 2015). Dzierżanowski et al. (2011) suggest that chemical composition and structure of the epicuticular wax layer may likely be of relevance in PM immobilization while Sæbø et al. (2012) identified the quantity of epicuticular waxes as an important leaf trait in immobilization of PM of all size fractions. In this study, we did not quantify the amount of leaf-waxes by plant species, but, to the best of our knowledge, this study has been the first to link EWS types to PM immobilization. The results of LMER indicated a significant effect of EWS type on total SIRM signal (i.e., SIRM_U and SIRM_w), independent of washing treatment. However, the effect of EWS types on the immobilized fraction of PM was indicated as not significant. Leaves of plant species which showed a change in EWS type or clustering of wax crystals within the same EWS type from June to September did not show significant differences in leaf SIRM from June to September (i.e., SIRM_D, Fig. 3b). Also, the Pearson correlation indicated a significant and fairly identical correlation for the four identified EWS types. Hence, concluding that immobilization of PM on leaf surfaces would be irrespective of the differences in EWS types. Based on these results, we conclude that the effect of EWS types on net particle accumulation (SIRM_U) and net particle immobilization (SIRM_w) remain analogous (Fig. 4c, d).

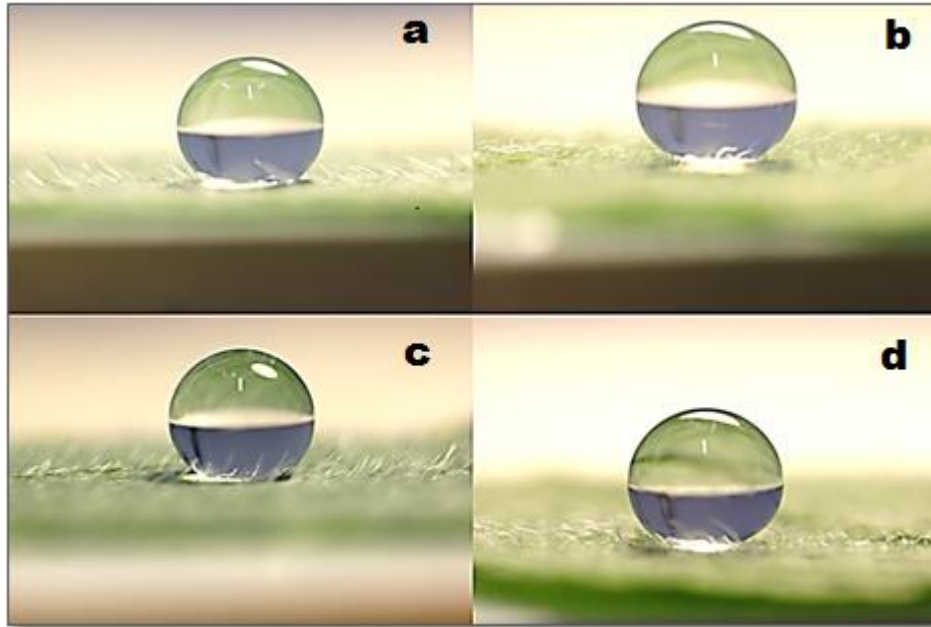


Fig. 5 Water droplets on the abaxial leaf surfaces with trichomes (a) *Cornus alba* (b) *Salix repens* (c) *Laburnum anagyroides* and (d) *Sorbus aucuparia* illustrating the bending of trichomes resulting in minimal contact with the leaf surface.

4.3 Implications for the use of perennial plant species in urban environments

Urban forests consisting of single trees, shrubs, parks, and forests provide ecosystem services which enhance the quality of the environment in cities and its city inhabitants (Roy et al. 2012). Of the many environmental ecosystem services (EES) provided by urban forests, mitigation of air quality and micro-climate regulation remains of utmost importance to the urban environments (Samson et al. 2019). In order to achieve the maximum benefits of urban forests for PM mitigation, the selection of appropriate plant species is of prime importance. For example, deciduous broadleaf plant species, i.e., *Populus* and *Quercus* may take up large amounts of gaseous pollutants, e.g., ozone, which increases during the growing season (Paoletti 2006) but these same species may be strong emitters of biogenic volatile organic compounds (BVOC's) (Calfapietra et al. 2013). Moreover, the performance of a plant species in PM mitigation may vary under stress conditions (Sicard et al. 2018). Hence, the adaptability of a plant species to local environments needs to be incorporated when defining plant species selection criteria. Following the deposition of particles on leaf surfaces, either by wet, occult, or dry deposition, it is possible that particles may be re-suspended back into the atmosphere either being blown away by wind or washed-off during a rain event (Burkhardt and Grantz 2016). The re-suspension rate depends on the aerodynamic diameter of the particle size, (i.e., higher for coarse PM and negligible for fine PM) (Nicholson 1993; Hofman et al. 2014). Plant species showing notable effectiveness in PM immobilization on their leaf surfaces have exceptional health relevance in urban environments. Especially the removal of fine particles is of importance, as they are considered to be more toxic to human health compared to coarse particles (Dockery et al. 1993). The present study comprehensively focused on the effectiveness of plant species in PM immobilization. The results indicated that for the same PM accumulation, plant species with leaf trichomes were more effective in immobilizing PM whereas plant species with low leaf wettability had a small immobilized fraction of PM. The significant correlation between leaf $SIRM_U$ and leaf $SIRM_W$ suggests that the selection of urban plants species for PM mitigation and for the protection human health through PM immobilization can be based on leaf $SIRM_U$. Though some plant species, for example, *A. fraseri*, *A. koreana*, *A. nordmanniana* deviated from the regression line, most plant species showed a significant correlation between leaf $SIRM_U$ and $SIRM_W$, contributing to more than 70 % of the immobilized fraction of PM. The importance of leaf micro-morphology in PM mitigation has been elaborated in many previous studies (Beckett et al. 2000; Freer-Smith et al. 2004, 2005; Mitchell et al. 2010; Dzierżanowski et al. 2011; Sæbø et al. 2012; Popek et al.

2013; Przybysz et al. 2014; Muhammad et al. 2019). At an individual tree level, the crown volume and the leaf area density (LAD - leaf area/unit volume, m^2m^3) of plant species need to be considered. Because crown traits such as crown geometry, foliage distribution, affect the wind flow, which determines the residence time of air pollutants in and around the crowns (Samson et al. 2017). Fowler et al. (1989) suggest that large tree crowns cause turbulent air movements, resulting in the interception and scavenging of particles from the atmosphere. Therefore, it is possible that a plant species with a low leaf SIRM_w but with a high LAI and LAD may effectively remove atmospheric PM from the ambient air while a plant species with a high SIRM_w at leaf level but with a low LAI and LAD, such as *B. davidii* could be overall less effective in PM immobilization.

5. Conclusion

A new standardized methodology was employed in this study to vigorously wash leaf samples in water using a vortex shaker for an impartial comparison in PM immobilization on leaves of perennial urban plant species ($n = 96$) using magnetic measurements of washed and unwashed leaves. Findings from our study indicated that the highest leaf SIRM_w, i.e., PM immobilization was observed on leaves of *V. lantana*, *S. intermedia*, *R. rubiginosa* in June and on leaves of *B. davidii*, *V. lantana* and *S. intermedia* in September. The lowest leaf SIRM_w was observed on leaves of *S. purpurea*, *L. anagyroides*, *P. alba* in June and on leaves of *P. alba*, *R. pseudoacacia*, and *A. fraseri* in September. Immobilization in deciduous (broadleaf/needle-like) tree and shrub species was significantly higher compared to evergreen needle/scale-like species. Trichome density, leaf wettability and EWS type significantly affected both the PM immobilization and PM accumulation on the leaves. Although the effect of these leaf traits seemed similar for immobilization and accumulation, we found that the immobilized fraction of PM increased with increasing trichome density and leaf wettability. These results indicate that leaves with (more) trichomes and higher wettability not only, as a start, accumulate more PM but are also more effective in entrapping a given amount of accumulated PM, making them more effective in immobilizing PM and less prone to PM re-suspension. It can be concluded that PM immobilization on leaf surfaces is governed by a combination of effects of leaf traits, rather than in the entirety of any single leaf trait on both deposition as well as immobilization. The correlation between leaf SIRM_u to leaf SIRM_w was significant in both June and September although some plant species such as *A. fraseri*, *A. koreana*, *A. nordmanniana*, *P. abies* deviated from the regression line with high leaf SIRM_u and low leaf SIRM_w. In general we conclude that leaf SIRM_u can be a good indicator for leaf SIRM_w although it should be taken into account that species with more hairy and more wettable leaves were more effective in PM immobilization than would be expected from the unwashed leaf magnetic signal.

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