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Species-specific dynamics in magnetic PM accumulation and immobilization for six deciduous and evergreen broadleaves

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ABSTRACT

Numerous studies have demonstrated the effectiveness of plants in accumulating airborne particulate matter (PM) on their leaf surfaces. However, it is not fully clear whether leaf traits or atmospheric conditions influence the accumulation and immobilization dynamics of magnetic PM on leaf surfaces. In this study, leaves of two deciduous broadleaf trees (*Quercus petraea*, *Quercus robur*), two deciduous broadleaf shrubs (*Prunus padus, Sambucus nigra*) and two evergreen broadleaf shrubs (*Prunus laurocerasus, Rhododendron* sp.) were sampled from a common garden every 48 to 72-hour to determine the dynamics of magnetic PM accumulation and immobilization. The mass of water-insoluble removable PM was estimated using gravimetric analysis. The ferro-magnetic and magnetizable component of leaf surface accumulated PM, leaf immobilized PM and water-insoluble removable PM in three size fractions (PM > 10, 3 -10, and 0.2 – 3 µm) was determined using Saturated Isothermal Remanent Magnetization (SIRM). Leaf SIRM for both surface accumulated PM and leaf immobilized PM differed in the following order *Q. robur* < *Q. petraea* < *S. nigra* < *P. laurocerasus* < *P. padus* < *Rhododendron* sp. indicating that PM immobilization on leaves of plant species is a function of net accumulated PM. In proportion to the SIRM signal of leaf surface accumulated PM, on average 4 % was recovered in the SIRM of the water-insoluble removable PM and 63 % was found in the SIRM signal of the immobilized PM while 33 % of the SIRM signal of the leaf surface accumulated PM could not be recovered in the immobilzed or water-insoluble removable PM. The leaf surface accumulated SIRM related with leaf wettability. The mass and SIRM of water-insoluble removable PM were significantly affected by leaf traits and meteorological conditions i.e., (i) negatively by leaf wettability, precipitation and wind speed and (ii) positively by relative humidity and ambient $PM_{2.5}$ concentrations. These results indicate that magnetic PM accumulation is influenced by both the atmospheric conditions as well as by the micro-morphological leaf traits of plant species. Leaves of *Rhododendron* sp. followed by *Q. robur* showed the highest median net deposition velocities for both coarse and fine-particles. Based on the results of this study we recommend *S. nigra* a deciduous broadleaf shrub species as a preferred choice to mitigate PM pollution in urban environments as it accumulated the highest proportion of ferro-magnetic particles in the mass of water-insoluble removable PM considering the harmful

effects of these particles on human health. In addition, *S. nigra* is known to have low biogenic volatile organic compound (BVOC) emissions and provides a good provision for birds and insects.

KEYWORDS: Net deposition velocity, coarse-particles, fine-particles, water-insoluble removable PM, ferromagnetic PM

1. Introduction

Urban green infrastructures which comprise of strategically planned network of natural, semi-natural and cultivated areas deliver a broad range of ecosystem services (Salbitano et al. 2016) including carbon dioxide (CO_2) sequestration (Fares et al. 2017), storm water run-off, protecting biodiversity (Pinho et al. 2017) and most importantly mitigation of particulate matter (PM) pollution (Sæbø et al. 2012; Popek et al. 2013). Particulate matter is emitted from both natural and anthropogenic sources. Natural sources of PM include biogenic and geogenic particles (Zeb et al. 2018). Biogenic particles consist of pollen, parts of plants, animals, and microorganisms (WHO, 2006; Alghamdi et al. 2014), while geogenic particles consist of soil, dust, and sea salt (WHO, 2006). Anthropogenic sources of PM include vehicular, industrial and agricultural activities (AQEG 2005). Anthropogenic PM typically consists of metals such as Fe, Pb, Zn, Ba, Mn, Cd and Cr (Matzka and Maher 1999; Tomašević et al. 2005; Mitchell and Maher 2009; Hansard et al. 2011, 2012; Sant'Ovaia et al. 2012; Castanheiro et al. 2016; Weerakkody et al. 2017). Airborne Fe-bearing PM usually occurs as a complex mixture of magnetite (Fe_3O_4), haematite (α-Fe_2O_3) and maghemite (γ - Fe_2O_3), with some metallic Fe (α-Fe) has also been reported (Maher et al. 2008; Hansard et al. 2011). Traffic related PM is typically the dominant source of magnetic particles not only due to the exhaust and/or combustion emissions, but also due to the mettalic wear and abrasion, brake wear (Gonet et al. 2021) and resuspension of street and road dust (McIntosh et al. 2007; Rai 2013).

Plants in urban environments increase the roughness of the ground. As a result, wind speeds and the mobility of particles is reduced (He et al. 2019). This happens because plants consist of stem, leaves, barks and branches, the flow momentum is absorbed and skin-friction drag in the canopy reduces the air flow velocity (Tong et al. 2015). Due to reduced wind speed and more turbulence due to vegetation, particles in the air have more time to deposit on exposed plant surfaces (i.e., stem, leaves, barks and branches) thereby decreasing the PM concentrations from air (Steffens et al. 2012). The transfer of airborne particulates from the atmosphere to plant surfaces 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 particles, whether solid or liquid has been generalized to be governed by four processes: gravitational settling (i.e., sedimentation), turbulent transfer through impaction and interception, and transfer by Brownian motion (Fowler et al. 1989). Particles through dry deposition are deposited in the function of their diameter (Slinn1982). Deposition through gravitational settling is effective for particles with a diameter (Dp) > 8 μm, while Brownian motion is the primary way of moving gases and fine particulates with Dp < 0.1 µm. Impaction and turbulent transfer are essential for medium and large particles 0.1 < Dp ≤ 10 µm (Beckett et al. 2000). Larger particles tend to quickly fall out of the airflow, permitting them to collide with obstacles and deposit on them (Fowler et al. 2004). Ultra-fine particles stay suspended in the air for an extended duration and can drift far distances from an emission source (Fowler et al. 2004). Slinn (1982) indicated that particle acceleration due to gravity is reduced with decreasing aerodynamic particle diameter. Apart from the four main deposition processes, smaller phoretic processes such as thermophoresis, diffusiophoresis and electrophoresis may contribute to the deposition of PM (Hicks et al. 2016). The PM deposited on leaves and exposed plant surfaces can be re-suspended (i.e., when aerodynamic lift forces are greater than adhesive forces) into the atmosphere by wind or washed off by rain (Freer-Smith et al. 2004; Dzierżanowski et al. 2011; Sæbø et al. 2012; Nowak et al. 2006; Blanusa et al. 2015). Gillette et al. (2004) stated that re-suspension of particles by wind occurs in either of the following conditions (a) when the force of airflow is large enough to detach the particle or (b) energized turbulent air disrupts the boundary layer and lifts the particles away into the airflow or (c) particles are detached from vibration or shaking of the whole leaf. If the re-suspended PM reaches the soil surface, the organic components of PM are either decomposed by natural processes, taken up by vegetation or leached from soil to the ground water, by run-off or by volatilization whereas the inorganic components of PM are accumulated in the soil and the soil solution (Dzierżanowski et al. 2011). If the re-suspended PM falls on a paved surface, it is more likely to be re-suspended back into the atmosphere (Przybysz et al. 2014) at the onset of suitable conditions such as surface drying or high wind speeds (Nicholson 1993). For particles which are trapped within the epicuticular wax layer (Dzierżanowski et al. 2011), stomatal cavities (Lehndorff et al. 2006; Song et al. 2015) or trichomes (Sæbø et al. 2012) are considered as immobilized particles and re-suspension of those particles by wind or by rain would be negligible (Terzaghi et al. 2013; Hofman et al. 2014; Przybysz et al. 2014).

The deposition of PM on a leaf surface can be evaluated using deposition velocity (Vd), i.e., the ratio of the deposition flux of the specified pollutant to the pollutant concentration (Seinfeld and Pandis 2006). Several factors, such as particle size, wind speed, and tree species, can affect the deposition velocity of particles (Beckett et al. 2000; Freer-Smith et al. 2005; Litschke and Kuttler 2008). Freer-Smith et al. (2005) demonstrated the effect of particle size on the deposition velocity revealing that deposition velocities were highest for ultra-fine particles (Dp < 0.1 µm). Previous studies have established the effect of plant species, illustrating a higher PM deposition on leaves of needle-like plant species compared to those of broadleaved species (Beckett et al. 1998; 2000; Freer-Smith et al. 2005; Dzierżanowski et al. 2011; Sæbø et al. 2012; Mo et al. 2015; Chen et al. 2017). For deciduous broadleaf plants, the roughness of the leaf surface is an essential factor in particle deposition compared to smooth leaf surfaces (Beckett et al. 2000; Mitchell et al. 2010). Evergreen needle/scale-like tree species due to their aerodynamic structure, high leaf area index (LAI: leaf surface area per unit ground area, m^2 m^{-2}), and presence of foliage throughout the year have been observed as more effective PM_{10} collectors (Beckett et al. 1998, 2000; Freer-Smith et al. 2005: Sæbø et al. 2012) than deciduous broadleaf plant species.

Our research aims to determine the dynamics in PM accumulation and immobilization on leaves of six deciduous and evergreen broadleaf plant species using a combination of magnetic and gravimetric analyses. The specific aims of this research were as follows (i) investigate the temporal variation in surface accumulated, immobilized and water-insoluble removable PM (ii) determine the ferromagnetic and magnetizable mass fraction of PM in three size fractions, (iii) calculate the species-specific net deposition velocity (i.e., the net result of PM deposition and re-suspension) of coarse and fine-particles and (iv) identify the effect of leaf traits (i.e., specific leaf area, drop contact angles and trichome density) and meteorological conditions (wind speed, relative humidity, precipitation) on the species-specific net deposition velocity. The SIRM of surface accumulated and immobilized PM was determined prior to and after vigorous leaf washing respectively. The mass of water-insoluble removable PM was analyzed using gravimetric filter analyses of washing water in three size fractions (PM > 10, 3 – 10, 0.2 – 3 µm). We hypothesize that PM accumulation on leaf surfaces is reduced with increasing wind speed and after precipitation events whereas it increases with an increase in trichome density and wettability of leaf surfaces.

2. Material and Methods

2.1 Experimental set-up

The study was conducted as a common-garden experiment on the premises of the University of Antwerp, i.e., on a secluded parcel of the Groenenborger campus (51º 10'46.0"N, 4º 25' 0.02"E) away from a direct pollution source. The set-up of the common-garden is fully described in a study by Muhammad et al. (2019). In brief, the investigated plant species and their respective replicates were bought from one pesticide-free nursery (Houtmeyers in Eindhout-Laakdal, Belgium) in March 2016 and potted in 15 L pots with organic soil infused with 150 g of Multicote 8, controlled-release fertilizer (Haifa Group N: P: K of 15:7:15 with MgO and trace elements). The pots were randomly placed in the common-garden in a 1.5 m x 1.5 m setting. All plants were regularly watered and monitored for pests or diseases.

2.1.1 Plant material

The investigated plant material comprised of six plant species commonly used in Western-European urban environments, consisting of (Table 1) two deciduous broadleaf trees (*Quercus petraea (*Matt) Liebl and *Quercus robur* L), two deciduous broadleaf shrubs (*Prunus padus* L and *Sambucus nigra* L), and two evergreen broadleaf shrubs (*Prunus laurocerasus* L and a *Rhododendron* sp.). Two replicates were investigated per plant species. From each plant replicate, eight mature, undamaged, and uninfected leaves were harvested at a 48 to 72-hour interval from 6th to 26th June 2017. Leaf samples from the investigated plant species and their respective replicates were collected from the south-east direction of the plant, to eliminate within-canopy orientation bias. The samples were collected at the petiole avoiding leaf surface contact and carefully placed in labeled paper envelopes. After leaf harvesting, the leaf samples were transported to the Laboratory of Environmental and Urban Ecology, University of Antwerp, Belgium, for bio-magnetic and gravimetric analyses.

2.1.2 Atmospheric and meteorological conditions during the study period

The atmospheric and meteorological conditions during the sampling period are described in Table 2. The ambient atmospheric concentrations of PM_{10} and $PM_{2.5}$ during the sampling period were acquired from the nearest air quality monitoring station (42R817, Antwerp Groenenborgerlaan, at 250 m from the experiment site) and the meteorological data were obtained from the nearest station (42M802, Antwerpen Luchtbal, Havanastraat, Antwerp) located approximately 10 km from the experiment site and operated by Flanders Environment Agency, VMM. The morphological leaf characteristics of the investigated plant species described in Table 1 have been previously reported in a study by Muhammad et al. (2019).

Table 1

Analyzed plant species (n = 6) according to functional plant types (n = 3) and their leaf traits [specific leaf area (SLA m^2 kg^{-1}), drop contact angle (DCA °) trichome density (TD mm^{-2})]. The data for leaf traits have been reported in a previous study by Muhammad et al. (2019).

Plant type	Plant species	SLA	DCA	TD
Deciduous broadleaf trees	*Quercus petraea* (Matt) Liebl	14.24	113	13.38
	Quercus robur L	16.94	125	0
Deciduous broadleaf shrubs	*Prunus padus* L	15.59	109	0.13
	Sambucus nigra L	18.22	60	1.38
Evergreen broadleaf shrubs	*Prunus laurocerasus* L	9.90	85	0
	Rhododendron sp.	10.16	67	0

2.2 Gravimetric analyses

Soon after leaf harvesting, each leaf sample from each replicate of each plant species was subsequently divided into two sub-samples to determine the leaf surface-accumulated particles and the leaf-immobilized particles. A minimum leaf area of 100 – 150 cm^2 was maintained for each sub-sample from each replicate of each plant species. The leaf area of fresh leaves was measured using a leaf area meter (Li-3100, LiCor Biosciences).

2.2.1 Removal of leaf surface accumulated PM

Following the leaf area measurements, the leaf samples from the first sub-sample were individually placed in 50 mL falcon tubes (Greiner Bio-one) and 50 mL of mineralized ultra-pure distilled water with a conductivity of 0.01 µS 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 motor speed of 10 (i.e., 3200 rpm) for three minutes precisely. The leaf washing duration and motor speed of the vortex shaker were selected based on preliminary testing which is explained in detail in a study by Muhammad et al. (2020). After leaf washing, the samples were removed from the falcon tubes with the help of tweezers and placed in Petri dishes for air drying and subsequent magnetic analysis. Subsequently, the washing water was stored in labeled falcon tubes and placed in the freezer awaiting filtration.

2.2.2 Filtration of the washing water

The washing water was filtered using Nuclepore track-etched polycarbonate, 47mm membranes (Whatman, UK). The mass of both (clean and loaded) filter membranes was determined using a 1 µg precision Mettler MT5 balance (Mettler-Toledo International Inc., Switzerland). Before weighing both (clean and loaded), the filter membranes were passed through an anti-static ionizer system (Mettler-Toledo International., Switzerland) to neutralize the electrostatic charge effects. Following the European standard guidelines (FprEN 12341:2013), the clean filter membranes were acclimatized in a climate-controlled room for 24-hours with an average relative humidity of 50 % and temperature 21 °C followed by the first pre-weighing (m_{C1}). The second pre-weighing (m_{C2}) was followed by additional acclimatization of filter membranes for ≥ 12-hours. The difference in mass of the clean filter membranes between the two observations ($m_{C1} – m_{C2}$) was expected to be ≤ 40 µg. If the condition was not fulfilled, the clean filter membranes were required additional acclimatization of 12-hours ensuing the third pre-weighing (m_{C3}). The difference between the last two observations ($m_{C2} – m_{C3}$) was supposed to fulfill the condition, or else the filter membranes needed to be discarded. All filter membranes met the weighting criteria. However, in many cases, a third measurement after an additional acclimatization of 24-hours was needed which was sufficient to meet the weighting criteria.

Following the pre-weighing, the clean filter membranes were placed in labeled Petri dishes (Greiner bio-one, diameter 90 mm) covered and left in a dark climate-controlled room awaiting filtration. On the day of filtration, the washing water was shaken for re-suspension of particles using Vortex-Genie 2 (MO BIO Laboratories Inc. New York, USA) at a motor speed of 10 (i.e., 3200 rpm) for 10 minutes. Next, a 47 mm glass filter funnel (GE Healthcare, UK) was connected to a vacuum pump (Vacuubrand, Germany) and placed over the pre-weighed filter membrane. The filtrate was subsequently filtered in three sessions with pore sizes in succession of 10, 3 and 0.2 µm. This enabled us to procure surface accumulated particles on filter membranes in three size fractions: PM >10 µm (very coarse: VC), PM 3 - 10 µm (coarse: C), PM 0.2 – 3 µm (fine: F). The loaded filter membranes were once again placed in labeled Petri dishes, covered and air-dried following the European standard guidelines (FprEN 12341:2013, clause 5.2.2) ensuing post-weighing. The first post-weighing (m_{L1}) was performed after an acclimatizing of the loaded filter membranes for 48-hours. The second post-weighing (m_{L2}) was performed after additional acclimatization of the loaded filter membranes for 24-hours following the first post weighing. The difference between ($m_{L1} – m_{L2}$) was expected to be ≤ 60 µg. If the condition was not fulfilled, the filter membranes were required additional acclimatization of 24-hours ensuing a third post-weighing (m_{L3}). The difference between the last two observations ($m_{L2} - m_{L3}$) was expected to fulfill the condition, or else the measurement was considered invalid, and the filter membrane was discarded. All filter membranes met the weighting criteria. However, in many cases, a third measurement after an additional acclimatization of 24-hours was needed which was sufficient to meet the weighting criteria. The mass of the clean filter membrane was subtracted from the mass of the loaded filter membrane. The difference between the mass of clean filter membrane and the mass of the blank filter membranes, following the entire washing, storage and shaking process, using only Ultrapure water was also subtracted from the calculated mass of the loaded filter membrane. As a result the mass of water insoluble removable PM in three size fractions for each sub-sample of each plant species was obtained. The mass of PM in each size fraction was divided by the leaf area of the washed leaves to obtain the leaf area normalized mass of removable PM in the three size fractions (m_{VC}, m_{C}, m_{F}), expressed as µg cm^{-2}.

2.2.3 Preliminary tests to determine the particle agitation intensity

The duration to shake the washing solution for re-suspension of the particles was determined by performing preliminary tests. Leaf samples of *Hedera helix* with two plant replicates and each replicate with a leaf area of 100 – 150 cm^2 were washed, and the washing water was filtered and stored similarly as that of the investigated plant species. The washing water was shaken using a vortex shaker as described above at 3200 rpm for a duration of 10, 20 and 30 minutes. It was observed that shaking of the washing water for more than 10 minutes showed a reduction of particle mass in three size fractions (i.e., PM > 10 µm, 3-10 µm, 0.2- 3 µm). It is possible that the particles accumulated on the leaf surface were water-soluble which caused a reduction in mass of PM at each iteration. Hence, we selected ten minutes for shaking the washing water at 3200 rpm resulting in a total of ~ 32,000 rotations) to re-suspend the particles within the washing water.

2.3 Saturation isothermal remanent magnetization (SIRM)

2.3.1 Assessment of water-insoluble removable PM in three size fractions using SIRM

Following the post-weighing of filter membranes, the ferromagnetic and magnetizable component of water-insoluble removable PM in three size fractions (i.e., PM > 10 µm, 3-10 µm, 0.2-3 µm) using saturation isothermal remanent magnetization (SIRM) was estimated. Before estimating the SIRM in three size fractions, the pre-processing protocol of Hofman et al. (2013) was followed. Each filter membrane was tightly packed in a cling film (Fresh Cling, 113831) and pressed in a 6.7 cm^3 sample pot (ASC Scientific). The sample pot along with filter membrane was magnetized at a magnetic field of 1 T using a Molspin magnetizer (Molspin Ltd. UK). Subsequently, the remanent magnetic intensity was determined using JR-6 Dual Speed Spinner Magnetometer (AGICO, 09173, Czech Republic). The intensity measurement was an average of two repetitions with an accuracy of ± 2.4 µA/m and corrected for the sample holder, sample pot and cling film. The magnetic intensity obtained in (A/m) was multiplied by the volume of the sample pot (6.7 cm^3) and divided by the leaf area (m^2) of the washed leaves to obtain leaf area normalized SIRM and expressed in µA. The SIRM signal of blank filter membrane of each pore size was determined and comprised of 0.38 ± 0.04 µA for 10 µm, 0.69 ± 0.01 µA for 3 µm and 0.87 ± 0.05 µA for 0.2 µm filter membranes. The SIRM of blank filters was subsequently subtracted from the SIRM of the loaded filter membranes, as such the SIRM of removable PM in three size fractions ($SIRM_{VC}$, $SIRM_{C}$, $SIRM_{F}$) was estimated.

2.3.2 Assessment of leaf surface accumulated and leaf immobilized PM using SIRM

The leaf SIRM of the washed leaves from the first sub-sample (see § 2.2) was determined by following the same pre-processing steps as described above and represents the leaf immobilized PM, i.e., particles immobilized within the epicuticular waxes, or in stomatal cavities or affixed on the trichome or hyphae of fungi and denoted as $SIRM_W$. The leaf SIRM of un-washed leaves was determined from the second sub-sample representing the total leaf surface accumulated particles and denoted as $SIRM_U$.

2.4 Calculation of species-specific net deposition velocities

In this study, the species-specific net deposition velocities (Vd in cm s^{-1}) for the removable fraction of PM_{10} and $PM_{2.5}$ (Eq. 1) were estimated, by analogy with the calculations of deposition velocity by Terzaghi et al. (2013). For each fraction and species, the difference in the leaf area normalized mass of particles (see § 2.2.) between subsequent sampling events (ΔPM mass, µg cm^{-2}) was divided by the product of the mean atmospheric PM concentrations [PM_{10} and $PM_{2.5}$ (µg m^{-3})] for the period in-between the given subsequent sampling events and the time in-between the given subsequent sampling events (seconds, refer Table 2). The calculations for PM_{10} were performed based on the sum of m_C and m_F whereas those of $PM_{2.5}$ were done using m_F (see § 2.2). The PM_{10} and $PM_{2.5}$ concentrations (Table 2) were measured at the nearest air quality monitoring station (42R817, Antwerpen Groenenborgerlaan) approximately 250 m from the experimental site operated by the Flanders Environment Agency (VMM).

$$\text{Vd (species)} = \frac{\Delta\text{PM mass}}{(\text{PM conc} * \Delta t)} \quad \textbf{Eq.1}$$

Where:

Vd (species) = species-specific net deposition velocity (cm s^{-1})

ΔPM mass = difference in leaf area normalized mass of PM_{10} [$\sum(m_C, m_F)$] or $PM_{2.5}$ [(m_F)] accumulated on the leaf (µg cm^{-2}) between the previous and the current sampling event

PM conc = mean PM_{10} or $PM_{2.5}$ air concentrations (µg cm^{-3}) between the previous and the current sampling event

Δt = exposure time (seconds) since previous sampling event

Table 2

The Investigated plant species analyzed during subsequent sampling events (6th – 26th) in June of 2017. The difference in exposure time (Δt) between the previous and current sampling event expressed in hours and seconds, the mean atmospheric [PM_{10} and $PM_{2.5}$ (μgm^{-3})] concentrations and meteorological conditions [accumulated precipitation (mm), wind speed (m s^{-1}), relative air humidity (RH %)] between the previous and current sampling event.

June 2017						
Time period	Δt (Hours, seconds)	PM_{10} (μgm^{-3})	$PM_{2.5}$ (μgm^{-3})	Precipitation (mm)	Wind speed (m/s)	RH (%)
6^{th} - 9^{th}	72, 259200	17.6	6.3	14.8	6.7	65
9^{th} - 12^{th}	72, 259200	16.7	6.8	0.0	4.5	59
12^{th} - 14^{th}	48, 172800	25.1	9.8	0.0	3.6	64
14^{th} - 16^{th}	48, 172800	28.1	10.1	0.0	4.3	54
16^{th} - 19^{th}	72, 259200	29.4	13.2	0.0	3.1	64
19^{th} - 21^{st}	48, 172800	34.7	13.2	0.0	3.3	57
21^{st} - 23^{rd}	48, 172800	32.9	15.3	0.0	4.0	55
23^{rd} - 26^{th}	72, 259200	13.6	5.6	0.0	5.4	72

2.5 Data analysis

The minimum, maximum and median values were calculated of the leaf area normalized mass of water-insoluble removable PM (m_{VC}, m_C, m_F), SIRM of water-insoluble removable PM ($SIRM_{VC}$, $SIRM_C$, $SIRM_F$), the SIRM of leaf surface accumulated PM ($SIRM_U$) and leaf immobilized PM ($SIRM_W$) on the 9 sampling events (6^{th} to 26^{th} June) to evaluate the differences between plant species. The coefficient of variance (CV) calculated as the ratio of standard deviation to the mean and expressed as percentage was calculated on the water-insoluble removable mass of very coarse, coarse and fine-particles. The net deposition velocities of coarse and fine-particles were calculated for the 8 periods in-between subsequent sampling events. A linear mixed-effect regression (LMER: Bates et al. 2015) model was applied separately on the mass and SIRM of water-insoluble removable PM, leaf surface accumulated PM ($SIRM_U$) and leaf immobilized PM ($SIRM_W$). First, we evaluated the effect of plant species, time and particle size, next the effects of leaf traits and finally the effect of atmospheric conditions on the mass and SIRM of water-insoluble removable PM, leaf surface accumulated PM ($SIRM_U$) and leaf immobilized PM ($SIRM_W$). The first LMER model was built using the plant-id nested within plant-replicate as a random effect, and plant species as factor variable, particle size fraction (three levels: very coarse, coarse and fine-particles), time period as a continuous variable, their interaction effects as fixed effects. Next, to determine the effects of leaf traits, the LMER was built using the plant-id nested within plant-replicate as a random effect and leaf traits (SLA, DCA, TD) as fixed effects. Lastly, to determine the effect of atmospheric conditions, the LMER was built using the plant-id nested within plant-replicate as a random effect and ambient PM_{10} and $PM_{2.5}$ concentrations, wind speed, precipitation and relative humidity (RH) as fixed effects. The LMER was also applied on species-specific net deposition velocity of coarse and fine particles separately with wind speed, precipitation, RH and leaf traits (SLA, DCA, TD) as fixed effects and plant id as random effect. The response variables in all LMER models [mass, and SIRM of water-insoluble removable PM, leaf $SIRM_U$, leaf $SIRM_W$ and net deposition velocity (coarse and fine-particles)] were natural log (ln) transformed to normalize the distribution (i.e., symmetric around their mean values). For each

LMER, the model was initiated by including all fixed effects. Subsequently, the model parameters with non-significant ($p > 0.05$) estimates were successively removed. The normality of residuals was checked by Shapiro-Wilk test. The Akaike Information Criterion (AIC) was used to compare the performances of different model structures. All statistical analyses were performed using R 3.2.2 software (R core Team 2015), the *Stats* package (R Core Team and contributors worldwide), stacked bar plots were generated using ggplot2 (Wickham, 2009) and scatter plots were generated using lattice library (Deepayan, 2008).

3. Results

3.1 Mass of removable PM in three size fractions and differences between plant species

The mass of water-insoluble removable PM (refer SM1, Table S1, Fig S1) in the three size fractions [very coarse (m_{VC}), coarse (m_C) and fine (m_F)] differed on leaves of the investigated plant species. The median m_{VC}, (i.e., PM > 10 µm) ranged from 21.7 to 77.0 mg m^{-2}. The lowest and the highest median m_{VC} was observed for *Q. robur* and *Rhododendron* sp. respectively. The median m_C, (i.e., PM 3 – 10 µm) ranged from 6.1 to 16.6 mg m^{-2}. The lowest and the highest median m_C was observed for *P. laurocerasus* and *Q. robur* respectively. The median m_F, (i.e., PM 0.2 – 3 µm) ranged from 5.7 to 11.2 mg m^{-2}. The lowest and the highest median m_F was observed for *Q. robur* and *Rhododendron* sp. respectively. For the six investigated plant species, the mass of removable PM (i.e., ∑ PM > 10, 3 -10, 0.2 – 3 µm) comprised of on average 63 % of m_{VC}, 21 % of m_C and 16 % of m_F (refer SM1, Fig. S2a). The coefficient of variance (CV) in mass of very coarse-particles was 35-66 %, in mass of coarse-particles was 31-56 % and in mass of fine-particles was 25-53 % between the investigated plant species during the sampling period.

3.2 SIRM of removable PM in three size fractions and differences between plant species

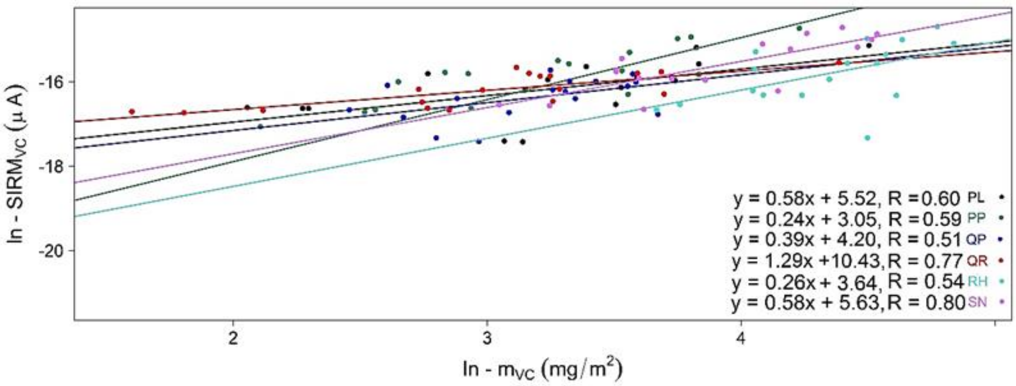
The SIRM of water-insoluble removable PM (refer SM2, Table S2, Fig. S3) in three size fractions was relatively similar on leaves of the investigated plant species. The median $SIRM_{VC}$, (i.e., PM > 10 µm), median $SIRM_C$, (i.e., PM 3-10 µm) and median $SIRM_F$, (i.e., PM 0.2-3 µm) ranged from 0.09 to 0.2 µA, 0.06 to 0.08 µA and 0.06 to 0.07 µA respectively. The lowest and the highest median $SIRM_{VC}$, $SIRM_C$, and $SIRM_F$ were observed for *Q. petraea* and *S. nigra* respectively. For the six investigated plant species, the SIRM of water-insoluble removable PM (i.e., $\sum SIRM_{VC}$, $SIRM_C$ and $SIRM_F$) comprised of, on average, 47 % of $SIRM_{VC}$, 29 % of $SIRM_C$ and 24 % $SIRM_F$ (refer SM1, Fig. S2b).

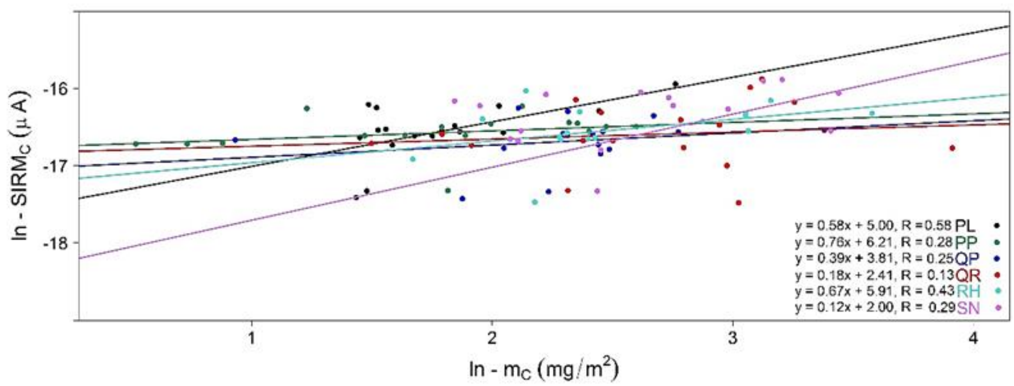
3.3 Leaf SIRM: surface accumulated PM ($SIRM_U$) and leaf immobilized PM ($SIRM_W$)

The median SIRM of leaf surface accumulated particles ($SIRM_U$) and leaf immobilized particles ($SIRM_W$) (refer SM2, Table S2) ranged from 3.7 to 17.6 µA and 1.9 to 12.3 µA respectively. The lowest and the highest median leaf $SIRM_U$ and $SIRM_W$ was observed on leaves of *Q. robur* and *Rhododendron* sp., respectively. The median $SIRM_U$ and median $SIRM_W$ increased in the following order *Q. robur* < *Q. petraea* < *S. nigra* < *P. laurocerasus* < *P. padus* < *Rhododendron* sp. The sum of the SIRM of immobilized particles ($SIRM_W$) and the sum of the SIRM of water-insoluble removable PM fractions, (i.e., ∑ $SIRM_{VC}$, $SIRM_C$, $SIRM_F$ and $SIRM_W$) ranged from 2.1 to 12.5 µA with the lowest and the highest total SIRM observed on leaves of *Q. robur* and *Rhododendron* sp. respectively. Of the SIRM signal of the unwashed leaves, on average 4 % was recovered in the SIRM of the water-insoluble removable PM and 63% was found in the SIRM signal of the immobilized PM (washed leaves) while 33% of the SIRM signal of the unwashed leaves could not be recovered in the immobilzed or removable PM.

3.4 Relationship between mass and SIRM of water insoluble removable PM

Pearson correlations between the SIRM and the mass of water-insoluble removable PM were computed for the three size fractions. When considering all species together, a significant and positive correlation was indicated for PM > 10 µm [$r = 0.50$, $n = 108$, $p < 0.0001$] whereas no significant correlation for PM 3 – 10 µm [$r = 0.16$, $n = 108$, $p = 0.08$] and PM 0.2 – 3 µm [$r = 0.02$, $n = 108$, $p = 0.77$] was indicated. For each species separately (Fig. 1), these relationships between SIRM and mass were significant ($p < 0.05$) for the six investigated plant species in case of PM >10 µm (Fig. 1 top). In case of PM 3 – 10 µm, the relationship between SIRM and mass was significant ($p = 0.02$) only on leaves of *P. laurocerasus* (Fig. 1 center). The relationship between SIRM and mass of PM 0.2 – 3 µm was not significant ($p > 0.05$) for any of the investigated plant species (Fig. 1 bottom). When Pearson correlations were computed, considering all species together between leaf SIRM of surface accumulated PM ($SIRM_U$) on the one hand and the sum of: leaf SIRM of immobilized PM ($SIRM_W$) and SIRM of water-insoluble removable PM in three size fractions, (i.e., ∑ $SIRM_{VC}$, $SIRM_C$, $SIRM_F$ and $SIRM_W$) on the other hand, a significant and positive correlation ($r = 0.47$, $n = 108$, $p < 0.0001$) was indicated. Considering the species separately (Fig. 2), this positive relationship was only significant for *Q. petraea* ($p = 0.003$), *Q. robur* ($p < 0.001$) and *Rhododendron* sp. ($p < 0.001$) and the steepest for the latter.





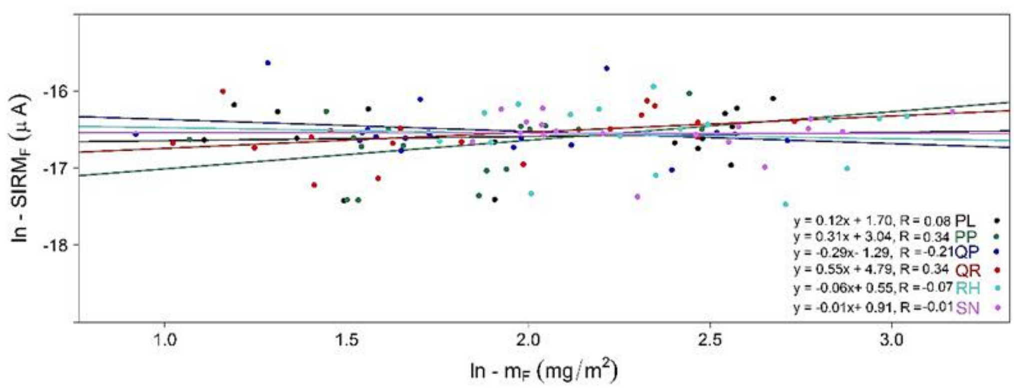


Figure 1. The scatter plot between mass of water-insoluble removable PM and SIRM of removable PM normalized by leaf area during sampling events (6th – 26th June) in three size fractions **(top)** m_{vc} and $SIRM_{vc}$ of very coarse fraction i.e., PM > 10 µm, **(center)** m_c and $SIRM_c$ of coarse fraction, i.e., PM 3 - 10 µm, **(bottom)** m_F and $SIRM_F$ of fine fraction, i.e., PM 0.2 – 3 µm. (Note the difference in the scales of the x and y-axes). The lines represent fitted regression lines for individual plant species in colors; black for *Prunus laurocerasus* (PL, n = 18), light blue for *Rhododendron* sp. (RH, n = 18), green for *Prunus padus* (PP, n = 18), voilet for *Sambucus nigra* (SN, n = 18), blue for *Quercus petraea* (QP, n = 18) and red for *Quercus robur* (QR n = 18). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

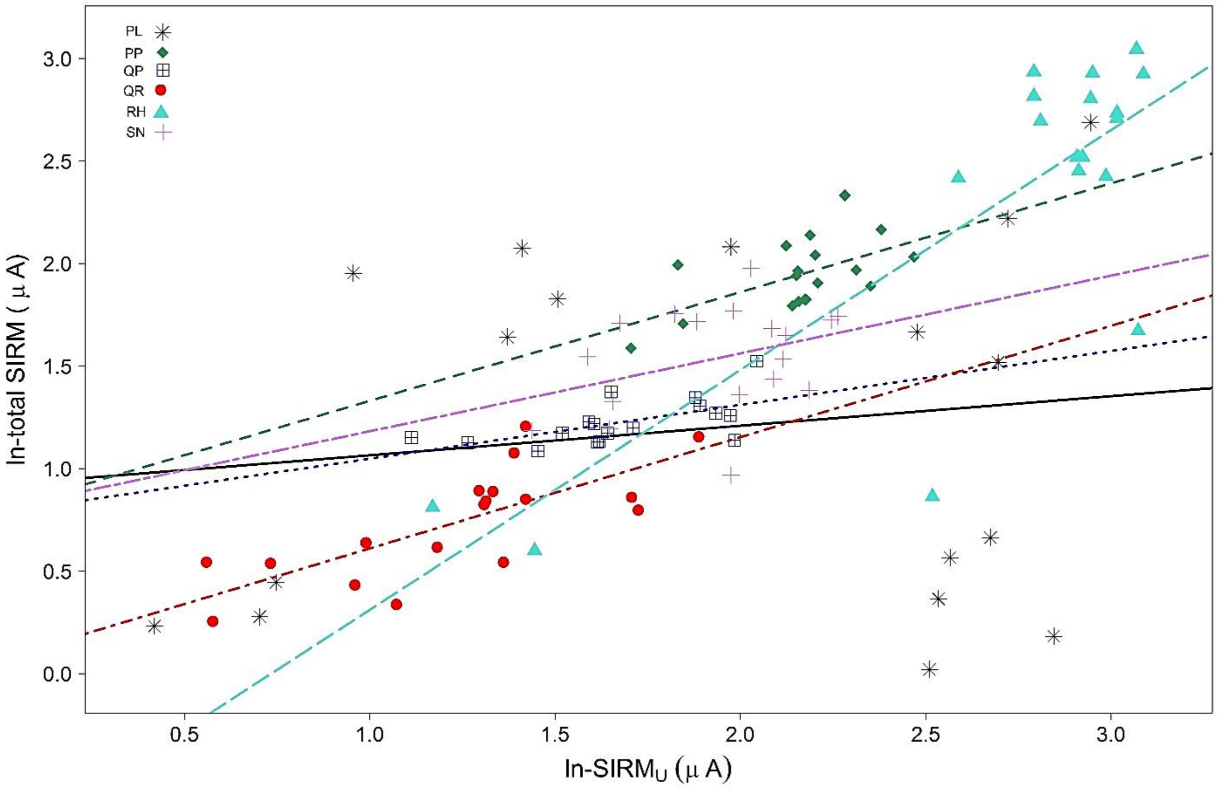


Figure 2. The scatter plot between log-transformed (ln) leaf SIRM of unwashed leaves ($SIRM_U$) and log-transformed (ln) total SIRM signal defined as SIRM of leaf immobilized PM and SIRM of mass of removable PM in three size fractions, (i.e., $\sum SIRM_W$, $SIRM_{VC}$, $SIRM_C$, $SIRM_F$) for the investigated plant species (n = 6) The lines represent fitted regression lines for individual plant species; *Prunus laurocerasus* (PL: solid, y = 0.18x + 0.38, R^2 = 0.04, n = 18), *Rhododendron* sp. (RH: long dash, y = 0.85x + 0.05 R^2 = 0.38, n = 18), *Prunus padus* (PP: dashed, y = 0.31x + 0.57, R^2 = 0.36, n = 18), *Sambucus nigra* (SN: two dash, y = 0.06x + 0.63, R^2 = 0.01, n = 18), *Quercus petraea* (QP: dotted, y = 0.18x +0.41, R^2 = 0.46, n = 18), *Quercus robur* (QR: dot dash, y = 0.72x - 0.06, R^2 = 0.76, n = 18). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3.5 Effect of plant species, time and particle size fraction on leaf SIRM of surface accumulated PM, leaf immobilized PM and on mass and SIRM of water-insoluble removable PM

The linear mixed-effect regression (LMER) model indicated a significant effect of plant species on the leaf SIRM of surface accumulated PM, leaf SIRM of immobilized PM and on mass of water-insoluble removable PM (Table 3). The leaf SIRM of surface accumulated PM was significantly ($p < 0.001$) higher on leaves of *Rhododendron* sp. compared to *P. laurocerasus*. The leaf SIRM of surface accumulated PM on leaves of *P. padus*, *Q. robur*, *Q. petraea* and *S. nigra* were not significantly different compared to *P. laurocerasus*. Considering the leaf SIRM of immobilized PM, leaves of *P. padus* and *Rhododendron* sp. indicated a significantly higher leaf SIRM ($p < 0.001$) compared to *P. laurocerasus* whereas a significantly lower leaf SIRM of immobilized PM was indicated for *Q. robur* ($p = 0.01$) compared to *P. laurocerasus*. A significant effect of plant species was indicated on mass of water-insoluble removable PM. Leaves of *Q. petraea*, *Q. robur*, *Rhododendron* sp. and *S. nigra* had significantly higher mass of water-insoluble removable PM compared to leaves of *P. laurocerasus*. No significant differences in mass of water-insoluble removable PM were indicated between *P. padus* and *P. laurocerasus*. In case of SIRM of water-insoluble removable PM, no significant differences were indicated between the investigated plant species.

A significant positive effect of time (Table 3) was indicated on leaf SIRM of surface accumulated PM ($p < 0.001$), on mass ($p < 0.001$) and SIRM ($p < 0.001$) of water-insoluble removable PM. Overall, the leaf SIRM of surface accumulated PM and mass and SIRM of water-insoluble PM were higher towards the end compared to the start of the sampling period. However, the effect of time on these dependent variables interacted significantly with plant species. The increase in SIRM of unwashed leaves ($SIRM_U$) with time was significant for *Q. petraea* and *P. laurocerasus* but not significant for *Q. robur*, *S. nigra*, *P. padus* and *Rhododendron* sp. The investigated plant species showed a significant increase with time in mass of water-insoluble removable PM but this increase in mass was significantly larger for *Q. robur*, *P. padus*, *P. laurocerasus* and *S. nigra* than for *Q. petraea* and *Rhododendron* sp. The SIRM of water-insoluble removable PM, increased with time on leaves of the investigated plant species. However, leaves of *P. laurocerasus* *P. padus*, and *S. nigra* indicated a significantly high SIRM signal with time whereas leaves of *Rhododendron* sp, *Q. petraea* and *Q. robur* did not indicate a significantly high SIRM signal with time. The effect of time, however, was not significant ($p > 0.05$) on leaf SIRM of immobilized particles.

The effect of particle size fraction on mass of water-insoluble removable PM (Table 3) was significant and positive. Leaves of investigated plant species had significantly ($p < 0.001$) higher proportion of very coarse-particles' mass compared to coarse-particles' mass. The interaction effect between plant species and particle size fraction indicated that leaves of *Q. petraea* ($p < 0.001$) and *Q. robur* ($p < 0.001$) had significantly less mass of very-coarse particles compared to mass of coarse-particles. Concerning the mass of fine-particles, leaves of *Q. petraea* ($p < 0.001$), *Q. robur* ($p < 0.001$), *S. nigra* ($p = 0.01$), and *Rhododendron* sp. ($p = 0.024$) were indicated to have significantly low mass of fine than of coarse-particles. Although no significant differences in SIRM of water-insoluble removable PM were indicated between the three particle size fractions. The interaction effect between particle size fraction and time was indicated as significant and positive. The SIRM of very coarse-particles was higher compared to the SIRM of coarse-particles and increased towards the end of the sampling period.

3.6 The effect of leaf traits, ambient PM concentrations and meteorological conditions on leaf surface accumulated and leaf immobilized SIRM and on mass and SIRM of water insoluble removable PM

The effect of leaf wettability (Table 3) was significant and negative on leaf SIRM of surface accumulated PM and on mass and SIRM of water-insoluble removable PM. Leaves with low wettability had a low mass of particles as well as a low SIRM signal compared to leaves with high leaf wettability. None of the leaf traits (SLA, TD and DCA) indicated a significant effect on leaf SIRM of immobilized PM. Concerning the effect of meteorological conditions, the effect of precipitation was indicated as significant and negative on leaf SIRM of surface accumulated PM and mass of water-insoluble removable PM. Following a period with precipitation, the leaf SIRM of surface accumulated PM was lower compared to the leaf SIRM of surface accumulated PM during dry events (i.e., without precipitation). The effect of wind speed was indicated to be significant and negative on mass of water-insoluble removable PM (Table 3). The effect of $PM_{2.5}$ concentration was significant and positive on the SIRM of water-insoluble removable PM but not it's mass. However, the significant interaction effect between particle size fraction and $PM_{2.5}$ concentration on mass of water-insoluble removable PM indicated a relatively stronger relationship with fine-particles (R^2 = 0.14, p = 0.02) compared to coarse (R^2 = 0.06, p = 0.85) and very coarse-particles (R^2 = 0.01, p = 0.92). The mass of coarse particles was high when ambient $PM_{2.5}$ concentrations were low. The effect of relative humidity was significant and positive on SIRM of water-insoluble removable PM (Table 3). The results of LMER did not indicate a significant effect of either atmospheric concentrations, meteorological conditions or leaf traits on leaf SIRM of immobilized PM (Table 3).

3.7 The effect of leaf traits and meteorological conditions on net deposition velocity

The median net deposition velocity for coarse and fine-particles ranged from 0.044 to 0.407 and -0.046 to 0.116 cm s^{-1}, respectively for the investigated plant species (Table 4). The lowest and the highest median net deposition velocity for both coarse and fine-PM was estimated on leaves of *Q. petraea* and *Rhododendron* sp. respectively. The LMER model indicated a significant effect of SLA on net deposition velocity of coarse PM (Table 5, Fig. 3). Leaves of plant species such as *P. laurocerasus* and *Rhododendron* sp. with a low SLA (Table 1) showed a higher net deposition velocity compared to plant species with a high SLA. A significant and negative effect of precipitation was indicated on the net deposition velocity of coarse PM. The effect of leaf traits (i.e., TD, DCA, and SLA) and meteorological conditions (i.e., RH, precipitation and wind speed) were indicated as non-significant (p > 0.05) on the net deposition velocity of fine-PM.

Table 3

ANOVA of fixed factors in the linear mixed effect regression (LMER) models applied separately with response variables as leaf SIRM of surface accumulated particles [ln($SIRM_U$)], leaf SIRM of immobilized particles [ln($SIRM_W$)], mass of water insoluble removable PM [ln(mass)], leaf SIRM of mass of water-insoluble removable PM [ln($SIRM_{filter}$)]. The random effect was plant-id nested within plant replicate. The fixed effects were particle size fraction (three levels: very coarse, coarse, fine-particles), time (continuous 9 sampling events), leaf traits [(i.e., specific leaf area (SLA), drop contact angle (DCA), and trichome density (TD)] and atmospheric conditions [(ambient PM_{10} and $PM_{2.5}$ concentrations, wind speed, precipitation and relative air humidity (RH)] for LMER models of [ln(mass), ln($SIRM_{filter}$), ln($SIRM_U$) and ln($SIRM_W$)]. The number of observation differ due to fixed effects included in each model [n = 324 (6 plant species x 2 replicates x 9 sampling events x 3 particle size fractions), n = 108 (6 plant species x 2 replicates x 9 sampling events)]. Significant effects ($p < 0.05$) are shown in bold.

	Response variable	Fixed effect	F value	p value
Effect of plant species, particle size, time on (n = 324)	Mass of water-insoluble removable PM	Plant species	13.00	< **0.001**
		Particle size	406.17	< **0.001**
		Time	183.84	< **0.001**
		Plant species x Particle size	9.87	< **0.001**
		Plant species x Time	5.81	< **0.001**
	SIRM of water-insoluble removable PM	Plant species	1.54	0.176
		Time	69.42	< **0.001**
		Particle size	2.92	0.055
		Plant species x Time	2.49	**0.031**
		Particle size x Time	16.251	< **0.001**
	Leaf surface accumulated SIRM ($SIRM_U$)	Plant species	7.46	< **0.001**
		Time	14.42	< **0.001**
		Plant species x Time	2.94	**0.017**
	Leaf immobilized SIRM ($SIRM_W$)	Plant species	16.24	< **0.001**
Effect of leaf traits on (n = 108)	Mass of water-insoluble removable PM	DCA	9.5	**0.001**
	SIRM of water-insoluble removable PM	DCA	5.05	**0.012**
	Leaf surface accumulated SIRM ($SIRM_U$)	DCA	7.95	**0.018**
	Leaf immobilized SIRM ($SIRM_W$)	DCA	2.12	0.183
		SLA	0.36	0.561
		TD	0.03	0.874
Effect of particle size and atmospheric conditions on (n = 324)	Mass of water-insoluble removable PM	Particle size	37.52	< **0.001**
		Wind speed	16.17	< **0.001**
		Precipitation	38.49	< **0.001**
		$PM_{2.5}$ conc	1.71	0.193
		Particle size x $PM_{2.5}$ conc	4.95	**0.027**
	SIRM of water-insoluble removable PM	Particle size	251.17	< **0.001**
		RH	5.01	**0.026**
		$PM_{2.5}$ conc	14.39	< **0.001**
	Leaf surface accumulated SIRM ($SIRM_U$)	Precipitation	16.15	< **0.001**
	Leaf immobilized SIRM ($SIRM_W$)	Wind speed	0.17	0.679
		Precipitation	0.05	0.826
		RH	0.01	0.939
		PM_{10} conc	0.01	0.997
		$PM_{2.5}$ conc	0.16	0.686

Table 4

The median, minimum, and maximum net deposition velocity (cm s^{-1}) estimated for coarse and fine PM on leaves of *P. laurocerasus*, *Rhododendron* sp., *P. padus*, *S. nigra*, *Q. petraea*, *Q. robur* during 6th to 26th June 2017 for 48 to 72-hour interval. The lowest and the highest median values of coarse and fine PM net deposition velocities are indicated in bold.

		Net deposition velocity (cm s^{-1})	
Plant species		Coarse PM	Fine PM
P. laurocerasus L	Median	0.116	0.030
	Min	-0.335	-0.075
	Max	1.087	0.297
Rhododendron sp.	Median	**0.407**	**0.116**
	Min	-1.057	-0.798
	Max	1.028	1.006
P. padus L	Median	0.073	0.071
	Min	-0.249	-0.110
	Max	0.472	0.169
S. nigra L	Median	0.087	-0.001
	Min	-0.298	-0.516
	Max	0.514	0.808
Q. petraea (Matt)Liebl	Median	**0.044**	**-0.046**
	Min	-0.255	-0.460
	Max	0.349	0.450
Q. robur L	Median	0.121	0.092
	Min	-0.369	-1.305
	Max	0.513	0.701

Table 5

ANOVA of fixed factors in the linear mixed effect regression (LMER) models applied separately with response variables net deposition velocity of coarse-particles [ln(vdcoarse)] and net deposition velocity of fine-particles [ln(vdfine)]. The random effect was plant-id nested within plant replicate. The fixed effects were leaf traits [(i.e., specific leaf area (SLA), drop contact angle (DCA), and trichome density (TD)], and meteorological conditions [(ambient PM_{10}, $PM_{2.5}$ concentrations, wind speed, precipitation and relative humidity (RH)]. Significant effects ($p < 0.05$) are shown in bold.

Response variable	**Fixed effect**	**F value**	**p value**
Net deposition velocity (coarse-particles)	Precipitation	10.97	**0.002**
	SLA	4.78	**0.037**
Net deposition velocity (fine-particles)	Wind	0.51	0.480
	Precipitation	0.71	0.411
	RH	0.75	0.395
	SLA	1.70	0.274
	DCA	0.03	0.863
	TD	0.05	0.828

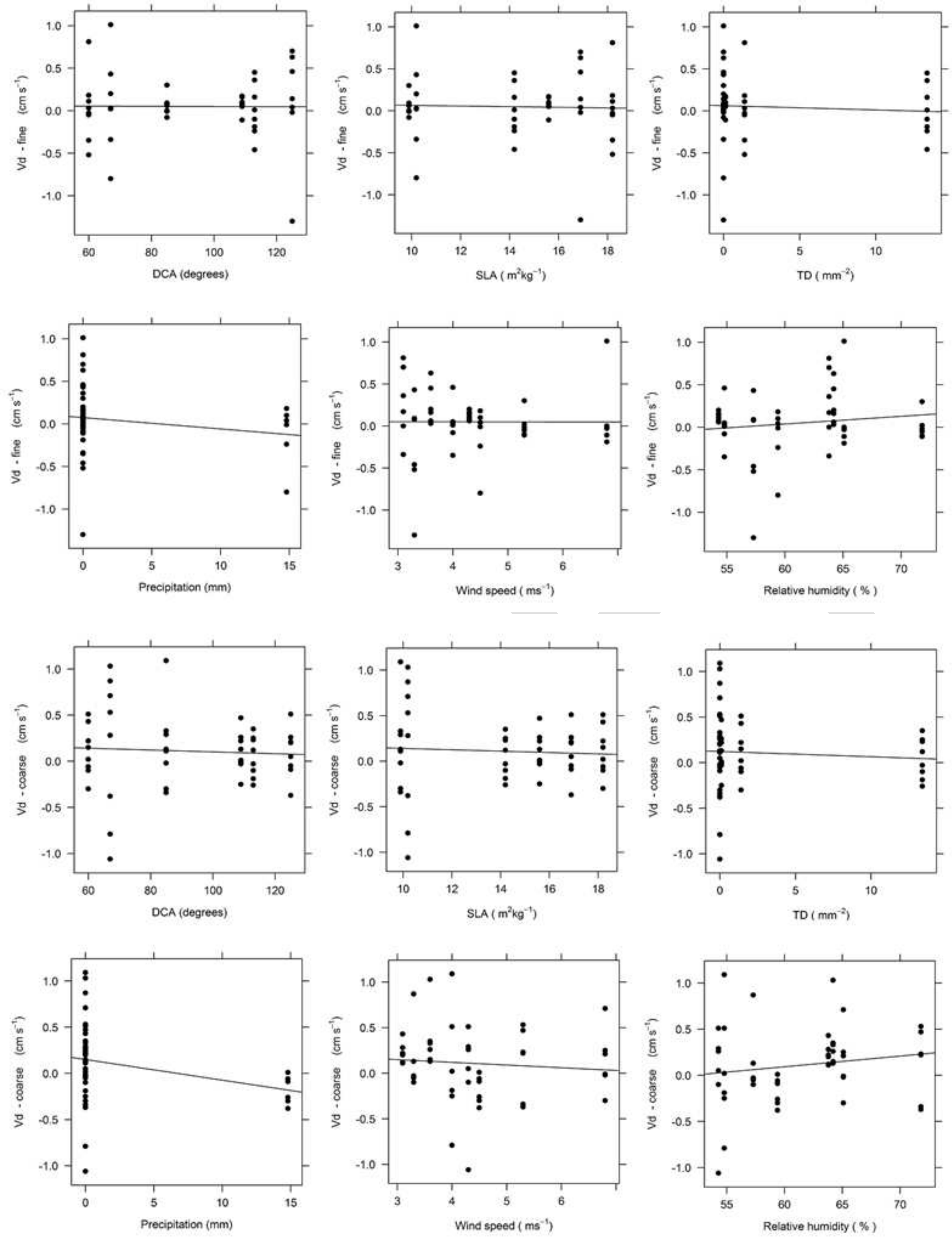


Figure 3. The effect of leaf traits [drop contact angle (DCA), specific leaf area (SLA) and trichome density (TD) shown on 1st and 3rd row] and meteorological conditions [precipitation, wind speed and relative humidity shown on 2nd and 4th row] on net deposition velocity of coarse (1st and 2nd) and fine-particles (3rd and 4th row) expressed in cm s^{-1} for the six investigated plant species. Lines shown are regression lines.

4. Discussion

4.1 Leaf surface accumulated and immobilized particles: differences between plant species and the effect of time, leaf traits and atmospheric conditions

The ferromagnetic and magnetizable component of PM accumulated on the leaf surface and immobilized within the leaf were analyzed using saturation isothermal remanent magnetization (SIRM) and expressed as $SIRM_U$ and $SIRM_W$ respectively. The median leaf $SIRM_U$ and leaf $SIRM_W$ ranged from 3.7 to 17.6 µA and 1.9 to 12.3 µA respectively with an average of 31 % reduction in SIRM signal after leaf washing. The investigated plant species showed an increase in both leaf $SIRM_U$ and leaf $SIRM_W$ in the following order *Q. robur* < *Q. petraea* < *S. nigra* < *P. laurocerasus* < *P. padus* < *Rhododendron* sp. This illustrates that PM immobilization on leaves of a plant species is a function of net accumulated PM. Besides, these differences in $SIRM_U$ and $SIRM_W$ between plant species can be explained with the species-specific leaf traits such as trichome density, leaf wettability, and specific leaf area. The effect of leaf wettability on leaf $SIRM_U$ was significant and negative (Table 3). Leaves with low leaf wettability removed more particles from their leaf surfaces compared to leaves with high wettability. Leaves with low wettability, for example, *Q. robur* and *Q. petraea* (Table 1), tend to easily remove particles from their leaf surfaces due to the surface roughness caused by epicuticular wax crystals, convex epidermal cells and trichomes (Neinhuis and Barthlott 1997). As a result, the contact area and the adhesion between particles and leaf surfaces is reduced while the contact area and adhesion between particles and water droplets is increased. No significant effects of leaf traits (i.e., trichome density, leaf wettability) on $SIRM_W$ were indicated. This may be either due to the small dataset of the investigated plant species or due to the short time-scale of the observations (i.e., 48-72 hours). A similar study by Muhammad et al. (2020) but with a wide array of plant species and longer exposure time between the two sampling events (i.e., June, September), illustrated the effect of leaf wettability and trichome density on the immobilized fraction of PM. It was shown that plant species with leaf trichomes were able to immobilize on average 70 % of leaf-accumulated PM while plant species with no leaf trichomes immobilized on average 48 % of leaf-accumulated PM. Similarly, plant species with low leaf wettability (i.e., DCA > 90 °) on average were able to immobilize 26-48 % of PM as compared to plant species with high leaf wettability (i.e., DCA < 90 °) immobilized on average 74-87 % of PM on their leaf surfaces (Muhammad et al. 2020). Many past studies (e.g., Mitchell et al. 2010; Kardel et al. 2011; Sæbø et al. 2012; Popek et al. 2013) have demonstrated the effect of trichomes on PM accumulation and immobilization. We assume that PM immobilization is enhanced on leaves with trichomes not only by a higher supply of particles due to a decreased boundary layer (Bakker et al. 1999) but for the same amount of accumulated particles a larger amount is immobilized due to the presence of leaf trichomes.

When LMER models were applied separately on leaf $SIRM_U$ and leaf $SIRM_W$, the effect of time on leaf $SIRM_U$ was significant and positive suggesting an increase in leaf $SIRM_U$ and leaf $SIRM_W$ towards the end than at the start of the sampling period. These results are in agreement with previously reported findings of (e.g., McIntosh et al. 2007; Kardel et al. 2012; Hofman et al. 2014). A variation in $SIRM_U$ and $SIRM_W$ occurred from event to event, with lower values in some events than their preceding event. These small $SIRM_U$ reductions in-between subsequent events can be related to the effect of precipitation which was indicated as significant and negative (Table 3) on leaf $SIRM_U$. The SIRM signal of unwashed leaves ($SIRM_U$) was low after a precipitation event but LMER did not indicate a significant effect of precipitation on the SIRM signal of washed leaves ($SIRM_W$). This illustrates that the net accumulated PM on the leaf surfaces is affected by wash-off due to rain, but not the particles immobilized by the leaf. The net surface accumulated PM on the leaf surfaces increased towards the end of the sampling period but are on a short term affected by resuspension by precipitation. The effect of time, atmospheric concentrations and meteorological conditions on leaf $SIRM_W$ were indicated as not significant. So, none of the variables investigated could explain the variation in $SIRM_W$ and thus in immobilized particles.

4.2 Mass of water-insoluble removable PM: differences between plant species and the effect of time, leaf traits, particle size, and meteorological conditions

The mass of water-insoluble removable PM reported in this study was in agreement with the mass ranges reported in literature (e.g., Beckett et al. 2000; Freer-Smith et al. 2005; Dzierżanowski et al. 2011; Sæbø et al. 2012; Popek et al. 2013; Hofman et al. 2014; Song et al. 2015). The results indicated that the mass of water-insoluble removable PM differed between the investigated plant species (n = 6). The median of the sum of all water-insoluble removable PM fractions (i.e., ∑ PM> 10, 3-10, 0.2-3 µm) was highest on leaves of *Rhododendron* sp., followed by *S. nigra* and *P. laurocerasus* (refer SM1, Table S1). The highest median mass of water-insoluble removable PM > 10 and 0.2 – 3 µm was observed on leaves of *Rhododendron* sp., while leaves of *Q. robur* showed the highest median mass of PM 3-10 µm (refer SM1, Table S1). Concerning the effect of leaf traits on mass of water-insoluble removable PM, the effect of leaf wettability was significant and negative. Leaves with low leaf wettability removed more particles after leaf washing compared to leaves with high wettability. As explained in § 4.1, leaves with low wettability (Table 1), tend to easily remove particles from their leaf surfaces due to surface roughness caused by epicuticular wax crystals, convex epidermal cells and trichomes (Neinhuis and Barthlott 1997). However, in the present study leaves of *Rhododendron* sp. and *S. nigra* with high wettability (Table 1) were also able to remove PM off their leaf surfaces. A plausible explanation may be that leaves of *Rhododendron* sp. and *S. nigra* collected 75 % and 70 % respectively of PM > 10 µm of the total mass of water-insoluble removable PM on their leaf surfaces (refer SM1, Table S1). Large particles typically originate from natural and biogenic sources such as pollen, bacteria, fungal spores or soil splash (Tomašević et al. 2005) which can be rapidly deposited through sedimentation under gravity (Freer-Smith et al. 2005), but which can also be easily resuspended back into the atmosphere. Popek et al. (2019) illustrated that large PM fraction (10 -100 µm) is typically the first to be resuspended from leaves whereas the smallest fraction (0.2 - 2.5 µm) remains attached for longer to the leaf as was suggested in a previous study of Nicholson (1993).

Leaves of investigated plant species (n = 6) accumulated the three considered particle size fractions on their leaf surfaces. The PM accumulation on a mass-basis was significantly higher for PM > 10 µm (63 %) compared to PM 3-10 µm (21 %) and PM 0.2-3 µm (16 %) (refer SM1, Fig. S2a). Similar findings were reported by (e.g., Beckett et al. 2000; Dzierżanowski et al. 2011 and Popek et al. 2013) who found less mass of fine particles than coarse particles on leaves of the investigated plant species thus corroborating the findings of the present study. In contrast, Ottelé et al. (2010) observed the greatest number of particles in small size fraction (0.5 – 1 µm) compared to PM ≥10 µm using scanning electron microscopy. Due to the lack of direct relationship between particle mass and particle density (Dzierżanowski et al. 2011), a direct comparison between the results of this study and that of Ottelé et al. (2010) can result in fallacious conclusions. It is possible that a higher mass of coarse-particles compared to the mass of fine-particles results in a high weight per leaf area in the gravimetric analyses (Weerakkody et al. 2017). Besides, Grochowicz and Korytkowski (1996) demonstrated in their study that when fine and ultra-fine particles contribute to 30 % of total PM weight they constitute of 99.9 % of the total number of particles.

The effect of time on mass of water-insoluble removable PM was indicated as significant and positive. This suggests that during the study period, the mass of water-insoluble removable PM increased towards the end of the sampling period. Results of this study are in agreement with the reported findings of McIntosh et al. (2007), Kardel et al. (2011), Hofman et al. (2014) and Rodríguez-Germade et al. (2014) who demonstrated a steady increase in particle accumulation with an increase in exposure time. In terms of particle size fraction, the largest variation in mass of water-insoluble removable PM during the sampling period was observed for very coarse particles (i.e., 35 - 66 %) followed by PM 3 - 10 µm (31 - 56 %) and PM 0.2-3 µm (25 - 53 %) between the investigated plant species. A recent study by Popek et al. (2019) explored the dynamics of on-surface and in-wax accumulated PM on leaves of four Australian plant species. These authors revealed that mass of removable PM not only differed between the investigated plant species but also changed very dynamically with time, i.e., daily between 33 – 35 %, thus corroborating the findings of this study. These changes in PM load due to resuspension (i.e., PM blown-off by wind or washed-off by rain) may remain hidden in weekly measurements (Popek et al. 2019). The authors also emphasize that resuspension of particles was dependent on particle size fraction where the mass of coarse and fine-particles was nearly similar throughout but increased slightly towards the end of the sampling period whereas very coarse-particles showed fluctuations between daily measurements with a rapid increase towards the end of the sampling period (Popek et al. 2019). The results of our study confirm that fluctuations in particle mass occurs mainly because plant surfaces are constantly in contact with their surrounding environments and experience continuous fluctuations in ambient PM concentrations. Moreover, the continuous episodes of wind and precipitation events considering the intensity and duration contribute to the accumulation and removal of PM from the leaf surfaces depending on their leaf micro-morphology. An intense precipitation event for an extended duration may remove PM from leaf surfaces while light precipitation for a short duration might make the leaves sticky which may temporarily enhance the particle accumulation (Wang et al. 2015; Xu et al. 2017). The effect of precipitation and wind speed on mass of water-insoluble removable PM (Table 3) were indicated as significant and negative suggesting that precipitation and/or wind events can resuspend a large fraction of the leaf surface accumulated PM back into the atmosphere resulting in a lower accumulated mass of removable PM. The mass of water-insoluble removable PM as shown in Figure S1 (refer SM1) was evidently low on 12th June after a precipitation event on the 9th June for all investigated plant species hence demonstrating particle resuspension. The resuspension of particles happens when drag forces on particles increase in relation to adhesive forces (Hinds 1986) thus easily pulling-off large particles of a leaf surface and re-suspending them back into the atmosphere (Pullman 2009). No effect of ambient PM concentrations on the accumulated mass of water-insoluble removable PM was observed. In the shorter time scale of 48- 72 h in our study, effects of resuspension by precipitation and wind were perceivable in the mass of removable PM but exposure did not, as suggested by Mitchell et al. (2010), lead to a dynamic equilibrium between leaf surface and atmosphere but to an increase in water-insoluble removable PM mass. This concludes that mass of water-insoluble removable PM is influenced by a combination of factors including leaf traits, particle size, and meteorological conditions.

4.3 SIRM of water-insoluble removable PM: differences between plant species and the effect of time, leaf traits, particle size and atmospheric conditions

The lowest and the highest median SIRM signal for the three considered size fractions was observed for *Q. petraea* and *S. nigra*, respectively (SM2, Table S2). It is worth mentioning that plant species which showed a high mass of water-insoluble removable PM from their leaf surfaces did not show a high SIRM of water-insoluble removable PM. Leaves of *Rhododendron* sp. showed the highest median mass of water-insoluble removable PM (sum of all fractions; refer SM2, Table S2) but leaves of *S. nigra* consistently showed the highest median SIRM of water-insoluble removable PM (sum of all fractions: refer SM1, Table S1). This implies that besides particle mass, the particle composition differs between plant species. This is further substantiated by the correlations between the mass and SIRM of the water-insoluble removable fraction for the three size fractions. The correlation between the SIRM and the mass of water-insoluble removable PM (refer § 3.4) was indicated to be significant for PM > 10 µm (Fig. 1 top) while insignificant for PM 3-10 and 0.2-3 µm (Fig. 1 center, bottom), respectively. The SIRM of water-insoluble removable PM in the considered size fractions (> 10, 3-10, and 0.2-3 µm) were relatively low compared to the reported SIRM results of Hofman et al. (2014). A plausible explanation would be the differences in leaf micro-morphology of the investigated plant species as well as the study site and exposure time. In the study of Hofman et al. (2014) leaf samples of *Platanus x acerifolia* were harvested in September whereas leaf samples in this study were harvested in June. Hence, the reduced exposure time may have contributed to low PM load including the magnetizable component of PM in our study. The results of LMER indicated a significant and positive effect of time on SIRM of water-insoluble removable PM, i.e., higher towards the end than at the start of the sampling period just like on its mass.

The effect of leaf traits on SIRM of water-insoluble removable PM was similar (see § 4.2) to that of its mass (Table 3). Concerning the effect of atmospheric PM concentrations and meteorological conditions on SIRM of the removable PM fraction, $PM_{2.5}$ concentrations and RH were indicated as significant and positive. This suggests that SIRM signal increased when ambient $PM_{2.5}$ concentrations and RH were high. For fine-particles, hygroscopicity is of utmost importance for a direct interaction with the leaf surface (Burkhardt 2010). The SIRM signal appropriately characterizes the fraction of atmospheric PM that is derived from combustion processes or metallic wear/abrasion (Lehndorff et al. 2006; McIntosh et al. 2007) which typically constitute of fine and ultra-fine particles (Tomašević et al. 2005) therefore the effect of relative humidity and ambient $PM_{2.5}$ concentrations were indicated on SIRM and not on mass of water-insoluble removable PM.
Theoretically, the SIRM of the removable PM in three size fractions ($SIRM_{VC}$, $SIRM_{C}$, $SIRM_{F}$) plus the SIRM of immobilized PM ($SIRM_{W}$) should sum up to be equal (SM2, Fig. S3) to the SIRM of surface accumulated PM ($SIRM_{U}$), but instead a difference was obtained. The sum of $SIRM_{VC}$, $SIRM_{C}$, $SIRM_{F}$ and $SIRM_{W}$ was systematically lower than $SIRM_{U}$ for the six investigated plant species. We assume that the difference was due to the water-soluble fraction of PM (Freer-Smith et al. 2005). In relation to the SIRM signal of unwashed leaves, on average 4 % of the SIRM signal was recovered in the SIRM of the water-insoluble removable PM and 63 % was recovered in the SIRM signal of the immobilized PM (washed leaves) while 33 % of the SIRM signal of the unwashed leaves could not be recovered in the SIRM of the immobilized or removable PM. Similar findings were reported by Xu et al. (2019) who determined the ratio of water-soluble ions to total water-insoluble PM which ranged between 7 to 50 % on leaf surfaces. Freer-Smith et al. (2005) revealed that the water-soluble ultra-fine PM can be dissolved promptly in precipitation and drip of the canopy in throughfall.
The particle size fraction that contributes to the SIRM signal of leaf immobilized PM ($SIRM_{W}$), remains unknown in this study. Nonetheless, if we take into account the findings of this study that 63 % of PM is immobilized within the leaf, and the findings of Terzaghi et al. (2013) that immobilization is effective for PM <10 µm and negligible for PM > 10.6 µm. As such we increase the relevancy of urban green infrastructures in PM mitigation by several folds.

4.4 Plant species-specific net deposition velocities

The plant species-specific net deposition velocities for PM_{10} and $PM_{2.5}$ were estimated, by analogy with the calculations of deposition velocity by Terzaghi et al. (2013). The net deposition velocities reported in this study (refer § 3.7, Table 4) were relatively in agreement with the deposition velocities reported by Terzaghi et al. (2013) (i.e., 0.02 to 0.04 cm s^{-1}) but lower than those reported by Freer-Smith et al. (2005) (i.e., 0.44 to 36.24 cm s^{-1}) and White and Turner (1970) (i.e., 3.0 to 7.1 cm s^{-1}). In a separate study using a wind tunnel, Freer-Smith et al. (2004) reported deposition velocities between 0.018 to 6.04 cm s^{-1} for seven plant species commonly found in Europe and semi-arid regions. A direct comparison of deposition velocities may be difficult due to differences in the investigated plant species and methodologies (i.e., field measurements versus wind tunnel). When the deposition velocities of broadleaf plant species reported by Beckett et al. (2000) and Freer-Smith et al. (2004) are exclusively taken into consideration, the deposition velocities ranged from 0.03 to

2.11 cm s^{-1} and 0.0018 to 3.134 cm s^{-1} respectively. These deposition velocities are comparable to the values obtained in the present study which range in between both formerly mentioned ranges.

The principal difference in the term "net deposition velocity" used in this study to the term deposition velocity used in literature, (e.g., Terzaghi et al. 2013) is in terms of its calculation. Terzaghi et al. (2013) used the average mass of particles (µg cm^{-2}) estimated over the 125 days of exposure time. The net deposition velocities (Eq. 1) were calculated every 48-72- hours for each investigated plant species using the difference in mass of particles (µg cm^{-2}) and a mean value of the atmospheric PM concentrations in-between subsequent sampling events. Accordingly, the estimated species-specific net deposition velocities varied between sampling events such that negative deposition velocities were recorded for some sampling events. This may suggest that the rate of PM resuspension was greater than the rate of PM accumulation. The effect of precipitation was indicated on the net deposition velocity of coarse but not of fine-particles. As illustrated in Figure S2a (refer SM1), the investigated plant species substantially accumulated coarse PM on their leaf surfaces compared to fine PM which may have contributed to the rapid resuspension of particles following a precipitation event. Leaves of *Rhododendron* sp. showed the most fluctuation in both coarse and fine-PM net deposition velocity with sharp decline and peaks between sampling events. A significant and negative effect of SLA was indicated on the net deposition velocity of coarse-PM (Table 5). This suggests that net deposition velocity of coarse-PM was higher on leaves of evergreen broadleaf shrub species with low SLA. Leaves of evergreen plant species have a considerably greater mesophyll tissue volume per unit leaf area resulting in low SLA compared to deciduous plant species (Villar et al. 2013). Plant species with a low SLA (i.e., evergreen) have been reported for high magnetic PM accumulation whereas plant species with high SLA (i.e., deciduous) have been reported for a low magnetic PM accumulation on their leaf surfaces (Muhammad et al. 2019). A straight forward explanation would be that due to the extended life span of evergreen plant species they tend to accumulate more PM on their leaf surfaces compared to deciduous plant species. Nonetheless, a high net deposition velocity of evergreen plant species may likely be due to the differences in leaf traits, such as, leaf wettability (i.e., high wettability) which hinders the self-cleaning mechanism of leaf surfaces (Neinhuis and Barthlott 1997) and thereby increasing the particle residence time on leaf surfaces. Furthermore, plant species with no leaf trichomes would imply to have a low PM accumulation on their leaf surfaces (Muhammad et al. 2019) whereas plant species with a high trichome density such as *Q. petraea* would suggest to have a high PM accumulation (Muhammad et al. 2019), but instead leaves of *Q. petraea* showed a low to negative net deposition velocity. This can be explained by the presence of leaf trichomes which would also result in low leaf wettability because the adhesion between particles and leaf surface is reduced resulting in enhanced self-cleaning of leaf surfaces. It can be concluded that particle accumulation on leaves is influenced by a combination of leaf traits and not in the entirety of any single leaf trait. The effect of leaf traits on net deposition velocity of both coarse and fine-particles were not indicated by the LMER. In addition, no effect of meteorological conditions on the net deposition velocity of fine-PM were indicated. We suppose that because of the shorter time scale (i.e., 48-72 h) measurements and very small differences in mass of water-insoluble removable PM between the investigated plant species, the effects of leaf traits were not found. It is possible that the effects of leaf traits on net deposition velocity may become significant during longer compared to shorter study period (i.e., three-weeks). It is therefore recommended that these experiments are performed for an extended duration (i.e., throughout the in leaf season).

4.5 Implications

Findings of this study can be of significance for generic models requiring species-specific net deposition velocities to estimate the benefits of urban green infrastructures. Furthermore, the exact identical order of plant species for SIRM$_U$ and SIRM$_W$ suggests that plant species accumulate and immobilize PM on their leaf surfaces in conformity of their leaf micro-morphology. In addition, SIRM$_U$ is a good indicator of SIRM$_W$ (Muhammad et al. 2020) because for the given leaf surface accumulated PM, plant species with a high leaf trichome density and a high leaf wettability immobilize more particles on their leaf surfaces (Muhammad et al. 2020). Moreover, the tedious and time consuming process of leaf washing can be avoided as SIRM$_W$ has been observed to be directly proportional to SIRM$_U$ for most (90 %) of the investigated plant species with very few (10 %) exceptions (Muhammad et al. 2020). Among the investigated plant species (n = 6) the deposition velocity for *Q. robur* and *Q. petraea* have been previously reported in studies of, (e.g., White and Turner 1970; Freer-Smith et al. 2004). To the best of our knowledge, the net deposition velocity for deciduous and evergreen shrubs included in this study (i.e., *P. padus*, *S. nigra*, *Rhododendron* sp., and *P. laurocerasus*) have not been reported thus far. Past studies (e.g., Beckett et al. 2000, Freer-Smith 2004; 2005; Räsänen et al. 2013; Terzaghi et al. 2013) have suggested that evergreen needle-scale-like plant species are better PM accumulators resulting in a high deposition velocities. It is recommended that besides the usual evergreen needle/scale-like plant species, various other plant species should be investigated to make informed choices in accordance to site requirements. In this study, leaves of *Q. robur* and *Rhododendron* sp. showed a high net deposition velocity of both coarse and fine-PM although not similar to evergreen needle/scale-like plant species yet substantial. To mitigate the effects of PM pollution in urban environments, it would be reasonable to have a mix of plant species that effectively accumulated either coarse or fine-PM, or preferentially both. The significant and negative effects of precipitation and wind speed on surface accumulated PM, mass of water-insoluble removable PM and net deposition velocity of coarse-PM illustrates that PM resuspension is a function of the particles' aerodynamic diameter. However, neither ambient atmospheric PM concentrations nor meteorological conditions (Table 5) were shown to have an effect on immobilized particles suggesting that PM once immobilized through leaf trichomes or epicuticular wax crystals or within stomatal cavities may not be readily resuspended back into the atmosphere. Hence, plant species with high PM immobilization abilities can be a preferred choice in urban environments to mitigate the effects of PM pollution.

To improve the air quality in urban environments using plants, the choice of plant species and the design (i.e., height and density of plants) should be selected based on site-specific and micro-climatic conditions. In addition, the role of shrubs should not be underestimated (Mori et al. 2018) because shrub species, as *S. nigra* did not show a high mass of water-insoluble removable PM but consistently showed a high SIRM signal for the three studied particle size fractions. This emphasizes that besides PM mass, the composition of the leaf surface accumulated PM differs between plant species. It also suggests that SIRM signal adequately characterizes the ferro-magnetic and magnetizable component of PM which is mainly composed of fine-PM (Tomašević et al. 2005). These ferro-magnetic particles may not have a considerable mass but can be detected thoroughly through high-precision magnetometers. Plant species which effectively collect these ferro-magnetic PM on their leaf surfaces would be of great significance because small sized particles are more toxic to human health compared to coarse-particles as they can infiltrate deep into the respiratory system (Dockery et al. 1993). At leaf level, the effective leaf traits in PM accumulation for example leaf wettability should not be disregarded as this can additionally improve the regulation of throughfall (Holder 2007).

The leaf washing methodology used in this study eliminated the possibility of uneven cleaning unlike if the leaf samples were hand washed. We recommend that future studies investigate the washing water of leaf samples for the water-soluble PM using ionic chromatography and inductively coupled plasma mass spectrometry (Ristorini et al. 2020) to determine the composition of leaf surface accumulated particles which may be leached or washed-off by rain. Based on the findings of our study on this set of six species, we recommend that besides trees, deciduous broadleaf shrub species such as *S. nigra* can be used in urban environments for PM mitigation, as they can also ambulate a high proportion of ferro-magnetic particles in the mass of water-insoluble removable PM. As for the ecosystem services provided by *S. nigra*, it is known to have low BVOC emissions (Samson et al. 2017) and provides a good provision for insects and birds (Samson et al. 2017) and therefore can be a preferable choice for PM mitigation in polluted urban environments. In general, also dis-services of plants (i.e., BVOC emissions, allergenicity) which can further exacerbate the urban air quality need to be taken into consideration (Grote et al. 2016) during plant species selection process.

5. Conclusions

The dynamics of magnetic PM accumulation on leaves of evergreen and deciduous broadleaf plant species were investigated by analyzing the SIRM of leaf surface accumulated and leaf immobilized PM and the mass and SIRM of water-insoluble removable PM. Significant differences between investigated plant species were observed for leaf surface accumulated PM, leaf immobilized PM and mass of water-insoluble removable PM. The investigated plant species showed the exact identical order from least to most for both surface accumulated PM and leaf immobilized PM suggesting that PM immobilization is a function of net PM accumulation. Plant species which showed a high mass of water insoluble removable PM from their leaf surfaces did not necessarily show a high SIRM of water-insoluble removable PM illustrating that besides particle mass, the composition of leaf surface accumulated particles differs between plant species.

The leaf surface accumulated PM, mass and SIRM of water-insoluble removable PM increased towards the end of sampling period but not monotonously. The mass of water-insoluble removable PM was influenced by precipitation and wind speed whereas ambient $PM_{2.5}$ concentrations and RH influenced the SIRM of water-insoluble removable PM. Based on the results of this study, it can be concluded that the effect of leaf traits (i.e., leaf wettability), atmospheric PM concentrations and meteorological conditions were of significance in both PM accumulation and removal from leaf surfaces. However, neither atmospheric PM concentrations nor meteorological conditions were shown to have an effect on immobilized particles suggesting that PM once immobilized through leaf trichomes or epicuticular wax crystals or within stomatal cavities may not be readily resuspended back into the atmosphere. Hence, plant species with high PM immobilization abilities should be a preferred choice in urban environments to mitigate the effects of PM pollution.

The standardized leaf washing procedure used in this study eliminated the possibility of uneven cleaning and enabled a reproducible washing procedure for all leaf samples. However, due to the loss of 33 % in SIRM signal we recommend that future studies investigate the washing water of leaf samples to determine the composition of water-soluble removable PM which may be leached or washed-off by rain. Finally, when selecting plant species for PM mitigation it is important to consider the site-specific conditions as well both ecosystem services and ecosystem dis-services provided by a specific plant species (Muhammad et al. work in progress). Based on the results of this study we recommend *S. nigra*, a deciduous broadleaf shrub species, as a good option to be also used in urban environments for PM mitigation because it collected the highest proportion of ferro-magnetic particles in the mass of water-insoluble removable PM. Moreover, it has low BVOC emissions and provides a good provision for insects and birds.

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