



Precipitation plays a key role in the processes of accumulation, retention and re-suspension of particulate matter on *Betula pendula*, *Tilia cordata* and *Quercus robur* foliage

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ABSTRACT

Particulate matter (PM) is one of the most dangerous air pollutants. Urban vegetation, especially trees, can accumulate PM and reduce its concentration in the air. However, knowledge about the dynamics of PM accumulation, retention and re-suspension is limited. This study examined the effects of rainfall on the dynamics of PM (0.2–2.5, 2.5–10 and 10–100 μm , surface PM, wax-embedded PM) accumulation, wash off and re-accumulation on *Betula pendula*, *Tilia cordata* and *Quercus robur* trees throughout the growing season. Irrespective of the species, rain affected PM deposited on plants, reducing the amount of PM on foliage by 17%, 24%, and 37% for *B. pendula*, *Q. robur* and *T. cordata* respectively. Rain had a lesser effect on the amount of wax-embedded PM washed off than on the amount of surface PM. After each rainfall event, PM re-accumulation was recorded. If the amount of PM washed off from the foliage were taken into consideration, the amount of PM that plants actually removed from the air would be 55% (*B. pendula*), 66% (*T. cordata*) and 62% (*Q. robur*) greater than the amount deposited on leaves at the end of growing season. Thus, many studies underestimate plants' potential for air purification.

Keywords: Air pollution; Particulate matter; Rain; Water; Phytoremediation; Trees

1. Introduction

Particulate matter (PM) is a serious global environmental issue influencing air quality, the environment and human health [1,2]. The negative effects of PM depends on its chemical composition, size and concentration in the atmosphere [3,4]. Chemically, PM is rich in toxic components such as trace elements, polycyclic aromatic hydrocarbons, dibenzofurans, polychlorinated biphenyls and black carbon [5,6]. Large PM (10–100 μm) can be a local problem, whereas small PM (0.001–2.5 μm) is a global threat because it is more dangerous to health and can travel thousands of kilometres from the emission source [7]. Many studies have shown that the growing concentration of PM in the air is

associated with an increased number of respiratory and cardiovascular diseases [8–10]. More than 90% of the global population is breathing polluted air and PM is responsible for approximately 7 million deaths each year [11].

Urban vegetation plays a key role in air phytoremediation and PM accumulation by vegetation is a widely documented phenomenon [12–14]. Due to their huge biologically active surface area, trees have the greatest potential to prevent PM dispersion, and adsorb and absorb PM on foliage [15–17]. It has been shown that trees can remove large amounts of PM of different sizes and origins from urban air [18,19]. PM is accumulated mostly by trees that have leaves covered in a wax layer, surface irregularities/roughness or various convex three-dimensional structures (e.g., leaf hairs, trichomes)

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[20–23]. PM accumulation also depends on stomatal density and size, the convexity of veins, the length of the petiole and the shape of the leaf edge [24,25]. The species with a high accumulation of PM are *Betula pendula* L., having high amount of waxes on the leaves in which PM can be stuck; *Quercus robur* L., whose stiff leaves remain on the tree for a long time, accumulating PM even in winter; and *Tilia cordata* Mill, characterized by a dense crown causing air turbulence which increases PM accumulation) [14]. However, the dynamics and efficiency of PM accumulation and retention by plants are greatly influenced by meteorological conditions, especially the frequency and intensity of precipitation [26,27].

The PM fate in the atmosphere and the efficiency of its phytoremediation are affected by air temperature and humidity [28]. However, the principal direct role in PM accumulation/re-suspension dynamics on plant foliage is played by wind and precipitation [26,29–31]. Air movement creates turbulence above the tree crowns and micro-turbulences close to the leaf surface, usually resulting in increased PM accumulation [32]. Depending on the complexity of the branch structure, wind can also blow PM off the leaf surface [33,34]. Strong wind can re-suspend between 27%–36% [27] and 76% [31] of accumulated PM on leaves. PM wash-off from foliage due to rainfall is a complex process that depends on rain duration and intensity, PM size, and plant species and their morphological features [29,31,35–38]. Artificial rainfall, equivalent to 10 mm in outdoor conditions, removes between 23% and 33% of previously accumulated PM (mainly the largest PM with a diameter of 10–100 μm) from plants [39]. A study by Xu et al. [38] showed that simulated rain washed off 51%–70% of surface PM, demonstrating that most PM not accumulated in wax is only deposited temporarily on foliage. About 12%–22% of PM washed off foliage is water-soluble PM [40]. Overall, it can be concluded that more precipitation removes more PM from the leaf surface [27]. Plants retain fine (0.2–2.5 μm) PM more than coarse (2.5–10 μm) and large (10–100 μm) PM [27,39,41]. The presence of a thick wax layer and trichomes make it difficult for PM to be washed off leaves [32,42,43].

The amount of PM accumulated on leaves can vary from week to week and even from day to day, increasing or decreasing depending on the level of PM emission and weather conditions [37]. The re-suspension of PM from the plant surface should not be regarded as a negative process. Foliage PM accumulation capacity will become saturated in the absence of its re-suspension. The PM load on foliage has been shown to reach its maximum after about 24 d [36]. Removal of PM from leaves by rain and wind could be treated as a cleaning of the biological filter, thus preparing the leaves for more deposition [37]. PM re-suspended from plants may be dispersed in the air or accumulated on another biological surface (plants growing under trees), on soil or on a paved surface (pavement, road) [26,27,33].

Although the influence of weather conditions (especially rain) on the dynamics of PM accumulation by plants is indisputable, this factor has not been taken into account in most studies on PM phytoremediation. Even when rainfall is included in the research plan, it is artificial rainfall, which only simulates natural rainfall to a certain extent. In order to fully understand the role of rain in air phytoremediation processes, experiments should be carried out

in natural conditions. Furthermore, in most studies, PM accumulation on plants is assessed only once during the growing season, usually at the end of the season or after a number of days without rain. The aim of this study was therefore to evaluate the effect of natural precipitation on the dynamics of PM accumulation, retention and re-suspension on three tree species over an entire vegetation season. The following hypotheses were tested: (i) under natural conditions, the amount of PM deposited on foliage differs from the amount when there is no rain, (ii) precipitation washes off PM deposited on foliage, and (iii) the ability of trees to accumulate PM is underestimated because washed-off PM is not included in the total amount of PM removed from the air by plants.

2. Materials and methods

2.1. Experimental approach

Planted *Betula pendula* L., *Quercus robur* L. and *Tilia cordata* Mill were exposed to different rain regimes (natural rain – plants treated with natural rain, no rain – plants protected from the rain and artificial rain conditions – plants treated with simulated rain of intensity 15 mm) to assess the effect of precipitation on the dynamics of PM accumulation on urban trees.

2.2. Study location and plant material

The experimental site was a botanical garden in central Poland (at 52°14' N and 21°01' E). The study area is characterised by low urbanisation and a very small number of air pollutant emission sources. The nearest, not particularly busy road was located 500 m from the garden's perimeter. No other anthropogenic sources of air pollutant emissions were identified. The average annual precipitation in the study location is 630 mm, with the highest in July (80 mm) and the lowest in January (23 mm). The average annual temperature is 9.8°C, with the hottest month in July (18.2°C) and the coldest in January (–1°C). The experiment was conducted during the 2020 vegetation season. Meteorological data from Institute of Meteorology and Water Management (IMGW-PIB) (average daily temperature, daily cumulative rainfall and days of rain) for the period between May and September 2020 are given in Table 1. July was much wetter than the long-term average (130.2 mm). The remaining months, particularly September (only 15.7 mm of rain and 4 d with precipitation), were dryer than average.

The objects of this study were three tree species: silver birch (*Betula pendula* L.), English oak (*Quercus robur* L.) and

Table 1
Meteorological data in the study period

Month of measurement	May	June	July	August	September
Precipitation (mm)	32.0	50.1	130.2	30.2	15.7
Days with rain	7.0	9.0	14.0	6.0	4.0
Temperature (°C)	16.0	19.0	19.5	18.3	17.1

small-leaved lime (*Tilia cordata* Mill.). Twelve plants of each species free of mechanical damage, diseases and pests were purchased from a local nursery and planted in the experimental location in March 2019. The plants were acclimatised in these locations for a year before the experiment commenced. At the beginning of experiment they were of the same age (4 y) and height (1.8–2.2 m) and with a similar crown size.

The experiment consisted of three treatments with four plants of each species: (i) control plants treated with natural precipitation, (ii) plants sheltered from natural precipitation, and (iii) plants sheltered from natural precipitation but treated with artificial rain of intensity 15 mm. For simplicity, these three experimental treatments were designated as 'Natural Rain', 'No Rain' and 'Artificial Rain' respectively. Plants in the Natural Rain treatment were grown in natural weather conditions (fully exposed to rain and wind), while plants in the No Rain and Artificial Rain treatments were protected from natural precipitation. The plants were sheltered from natural rain by a roof of reinforced garden foil that was permeable to sunlight and allowed air to flow freely.

2.2.1. Effect of natural rain on PM deposition on foliage

The Natural Rain and No Rain treatments were used to test the impact of natural precipitation on PM accumulation by the examined plants species. Plant material was harvested four times at monthly intervals (June, July, August and September). From each tree individual (biological replication), four samples were harvested [total n per treatment and harvest = 3 species \times 4 trees (biological replicates) \times 2 rain conditions].

2.2.2. Dynamics of PM accumulation and re-accumulation processes after rain events

Trees from the Artificial Rain treatment were used to assess the dynamics of PM accumulation and re-accumulation processes after rain events. Plant material was harvested four times: (i) at the beginning of May when the first leaves reached their maximum size, (ii) in mid-June, (iii) at the beginning of August and (iv) in mid-September at the end of growing season. For simplicity, the time periods between harvest events are defined as accumulation periods: the 'spring accumulation period' (from early May to mid-June), the 'summer accumulation period' (from mid-June to early August) and the 'autumn accumulation period' (from early August to mid-September). After each harvest, trees were subjected to intensive artificial precipitation (equivalent to 15 mm in outdoor conditions) using spray nozzles for 30 min. The water used for spraying came from rainwater no older than 2 d. After drying (for about 24 h), leaves were harvested from the trees again. From each tree individual (biological replication), four samples were harvested [total n per harvest = 3 species \times 4 trees (biological replicates)].

A single plant sample consisted of 5–10 leaves (depending on their size) collected from different parts of the plant. In order to avoid filters clogging during the PM-filtering process, the total area of each sample did not exceed 300 cm². After harvest, the samples were placed in paper

envelopes and stored at a constant temperature and constant humidity conditions until analysis.

2.3. Quantitative assessment of PM and wax content

Accumulation of PM and the amount of epicuticular waxes on foliage were determined gravimetrically using the method of Dzierzanowski et al. [45]. The amount of PM was determined as the amount of surface PM ($_{\text{s}}\text{PM}$) that was washed off foliage with water, and in-wax PM ($_{\text{w}}\text{PM}$) that was washed off with chloroform (PM immobilised in waxes). Both categories were analysed in three size fractions: 0.2–2.5, 2.5–10 and 10–100 μm . Leaf samples were washed with distilled water first and then with chloroform. The solutions obtained were then passed through a 100 μm mesh sieve (to remove particles larger than 100 μm) and were then sequentially filtered using pre-weighed filters of type 91 (paper filter with a pore size of 10 μm), type 42 (paper filter with a pore size of 2.5 μm) and polytetrafluoroethylene (PTFE) membrane filters (a pore size of 0.2 μm) (all Whatman, UK). After filtration, all the filters were weighed again. The quantity of epicuticular waxes was weighed after evaporation of the chloroform collected in pre-weighed beakers. The area of leaves from each sample was determined using an Image Analysis System (Skye Instruments Ltd., UK) and SkyeLeaf software (Skye Instruments Ltd., UK). The amounts of PM from filters and waxes were then recalculated to give the $\mu\text{g}/\text{cm}^2$ of leaves.

2.4. Statistical analysis

Data were subjected to analysis of one-factorial analysis of variance using Statgraphics Plus 4.1 (Statpoint Technologies Inc., Warrenton, VA, USA). The Shapiro–Wilk test was used to examine the normality of distribution, while Bartlett's test verified the homogeneity of variances. Differences between means of combinations were evaluated by post hoc Tukey's honestly significant difference (HSD) test. Means were considered to be significantly different at $P < 0.05$.

3. Results

3.1. Effect of natural rain on PM deposition on foliage

The examined plants accumulated PM throughout the growing season, but foliage deposition varied depending on the rain treatments and the time in the season (Fig. 1). Irrespective of the species and harvest date (except at the beginning of the experiment), PM accumulation in No Rain conditions was always higher, mostly significantly so, than in plants exposed to natural precipitation. Natural Rain conditions reduced PM deposition on tree foliage by 12%, 18% and 21% in *B. pendula*, by 4%, 31% and 35% in *Q. robur*, and by 32%, 24% and 56% in *T. cordata* in July, August and September respectively (Fig. 1).

The amount of PM deposited on tree foliage was dynamic and changed in the subsequent months (Fig. 1). In the period from June to August, the amount of PM accumulated on leaves increased gradually in both the Natural Rain and No Rain treatments. The greatest PM accumulation was found in August for all the studied species. In September, the

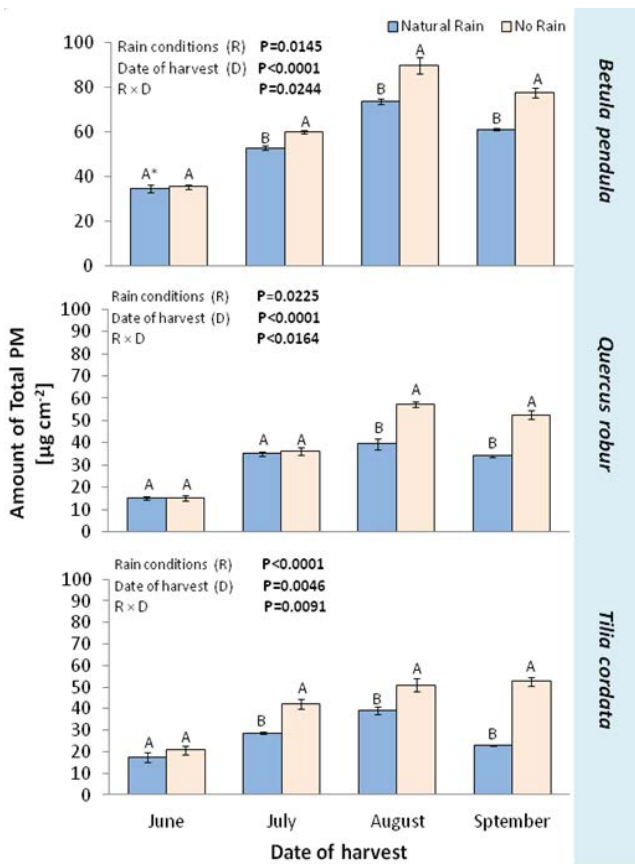


Fig. 1. Amount of total PM on the foliage of *B. pendula*, *Q. robur* and *T. cordata* growing in Natural Rain and No Rain conditions. Data are means \pm SE, $n = 12$. *Different uppercase letters indicate a significant difference between Natural Rain and No Rain conditions in individual months at $P < 0.05$ by Tukey's HSD test.

amount of PM deposited on the leaves usually decreased, with exception of *T. cordata* grown in No Rain conditions. The September decrease in the amount of PM on leaves was more pronounced in plants exposed to natural rain, reducing by 18%, 14% and 41% respectively for *B. pendula*, *Q. robur* and *T. cordata*. The difference in PM deposition between the end and the start of the experiment was 26.0 and 42.3 $\mu\text{g}/\text{cm}^2$ in *B. pendula*, 18.9 and 37.3 $\mu\text{g}/\text{cm}^2$ in *Q. robur*, and 5.6 and 31.7 $\mu\text{g}/\text{cm}^2$ in *T. cordata* for plants grown in Natural Rain and No Rain conditions respectively (Fig. 1).

The studied species differed in the share of PM deposited on the leaf surface (s_{PM}) and permanently embedded in the waxes (w_{PM}) (Fig. 2). On foliage of *B. pendula*, regardless of rain conditions, the majority of PM was accumulated as w_{PM} (on average 62% of total PM). The share of s_{PM} and w_{PM} in *Q. robur* depended on the plants' exposure to precipitation. Plants of *Q. robur* treated with Natural Rain accumulated more PM as w_{PM} (on average 54% of total PM), while those grown in No Rain conditions accumulated slightly more s_{PM} (on average 52% of total PM). *T. cordata* exposed to natural rain accumulated very similar amounts of s_{PM} and w_{PM} , but in No Rain conditions most of the PM deposited on the foliage belonged to the s_{PM} category (on average 57% of total PM) (Fig. 2).

The amount of both s_{PM} and w_{PM} was always greater in plants in the No Rain treatment (Fig. 2). The differences in PM accumulation between plants exposed to Natural Rain and grown in No Rain conditions were greater in s_{PM} than in w_{PM} . Deposition of s_{PM} was greater in the No Rain treatment by on average 10% (with the greatest difference recorded in July) in *B. pendula*, by 17% (with the greatest difference recorded in September) in *Q. robur* and by 22% (with the greatest difference recorded in September) in *T. cordata*. The differences in w_{PM} accumulation were smaller and on average amounted to 6% (with the greatest difference recorded in September) in *B. pendula*, 7% (with the greatest difference recorded in August) in *Q. robur* and 19% (with the greatest difference recorded in September) in *T. cordata*. In all the examined species and rain treatments, the accumulation of s_{PM} and w_{PM} was greater at the end of the experiment than at the beginning, by 12% for s_{PM} and 42% for w_{PM} in *B. pendula*, by 47% for s_{PM} and 20% for w_{PM} in *Q. robur* and by 89% for s_{PM} and 75% for w_{PM} in *T. cordata* (Fig. 2).

Irrespective of the rain conditions, all the species accumulated PM mostly as large PM (10–100 μm), followed by coarse PM (2.5–10 μm) and fine PM (0.2–2.5 μm) (Table 2). The average ratio between different PM size fractions was 47%:27%:26% (Natural Rain) and 46%:26%:28% (No Rain) in *B. pendula*, 56%:32%:11% (Natural Rain) and 51%:36%:13% (No Rain) in *Q. robur*, and 56%:30%:14% (Natural Rain) and 53%:36%:11% (No Rain) in *T. cordata*. The accumulation of PM size fractions varied over time. The greatest accumulation of all PM size fractions was usually recorded in August, while the lowest was most often in June. Between August and September, a decrease in the amount of PM (except for large PM in *B. pendula*) deposited on plants was recorded. The accumulation of all PM size fractions on plant foliage was higher in No Rain conditions in most cases. The September decline in PM deposition was also greater in trees grown in Natural Rain conditions (Table 2).

Foliage of *B. pendula* was covered with significantly the greatest amount of epicuticular waxes, 11 and 17 times greater than *Q. robur* and *T. cordata* respectively (Table 2). The amount of wax did not differ significantly between harvest dates or rain conditions (Table 2).

3.2. Dynamics of PM accumulation

PM accumulation by rain-protected *B. pendula*, *Q. robur* and *T. cordata* plants was lowest at the beginning of the growing season (Fig. 3). At the start of the experiment, the largest amount of total PM was deposited on the foliage of *B. pendula*, followed by *T. cordata* (42% less than *B. pendula*), and lastly *Q. robur* (61% less than *B. pendula*). Plants of all the examined species responded similarly to the regular treatments (every 45 d) with artificial rain (Fig. 3A). The amount of total PM on foliage, regardless of the species, was always significantly lower after 15 mm simulated rain. The processes of PM accumulation (during accumulation periods) and its washing off (after treatment with simulated rain) from foliage varied between species and different time periods during the vegetative season. During the spring accumulation period, the deposition of total PM on foliage increased by 41%, 44% and 58% respectively for *B. pendula*, *T. cordata* and *Q. robur*. The first artificial rain washed off

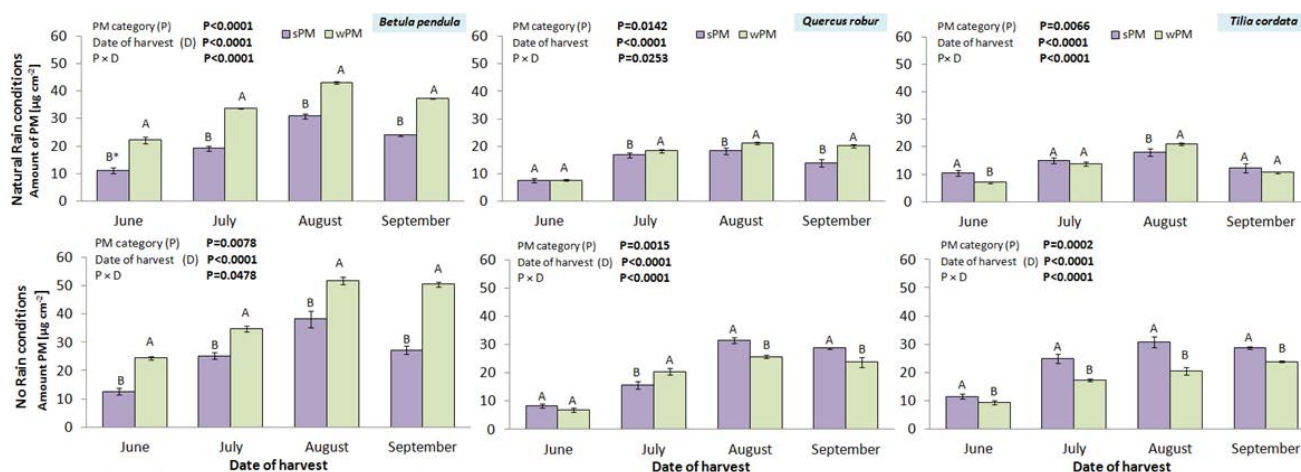


Fig. 2. Amount of $s\text{PM}$ and $w\text{PM}$ on the foliage of *B. pendula*, *Q. robur* and *T. cordata* growing in Natural Rain and No Rain conditions. Data are means \pm SE, $n = 12$. *Different lowercase letters indicate a significant difference between Natural Rain and No Rain conditions in the individual month at $P < 0.05$ by Tukey's HSD test.

the greatest amount of total PM from the leaves of *T. cordata* (37%), followed by *Q. robur* (28%) and lastly *B. pendula* (24%). The summer accumulation period resulted in the largest increases of total PM on foliage during the growing season. Deposition of total PM on *B. pendula* increased by 55% compared with the amount of PM after the first artificial rain. In *Q. robur* and *T. cordata*, this increase amounted to 60% and 65% respectively. The second simulated rain event caused the greatest reduction in the amounts of total PM on foliage. Furthermore, the recorded decrease was greater than the amount of total PM accumulated during the second accumulation period, reducing by 61%, 67% and 69% for *B. pendula*, *Q. robur* and *T. cordata* respectively. During the autumn accumulation period, the increase in total PM deposition on plants was smaller than during the earlier accumulation periods, and was similar for all species (35%).

In all the examined species, the increase in the accumulation of large (10–100 μm) and coarse (2.5–10 μm) PM was greatest during the summer accumulation period, rising by 57%–67% and 54%–58% for large and coarse PM respectively (Fig. 3A). The smallest increase in deposition of large and coarse PM in *Q. robur* (44% for large PM and 51% for coarse PM) and *T. cordata* (38% for large PM and 35% for coarse PM) was recorded during the autumn accumulation period. On the foliage of *B. pendula*, the smallest increase (40%) of large and coarse PM was noted in the spring accumulation period. Accumulation of fine (0.2–2.5 μm) PM increased (by 52%, 72% and 81% for *B. pendula*, *T. cordata* and *Q. robur* respectively) to the greatest extent in the spring accumulation period, while the smallest increase of this PM fraction was recorded in the autumn accumulation period at 14% and 10% for *B. pendula* and *T. cordata* respectively. The only species in which PM deposition decreased (by 49% in the last accumulation period) was *Q. robur*. The amount of PM belonging to different size fractions washed off foliage by artificial rain differed between species and time periods. Irrespective of the species and PM size fraction, most of the PM was washed off plants by a second treatment with

artificial rain. In *B. pendula* and *Q. robur* plants, the second simulated rain event removed mostly large PM (70% of previously accumulated PM) from the foliage and the smallest amount removed was fine PM (45% of the previously accumulated PM). A different trend was recorded in *T. cordata*, from which all PM fractions were washed off foliage to a similar extent (67%, 70%, and 75% for large, coarse and fine PM respectively). The first treatment with artificial rain washed less PM off the plants. In *T. cordata*, the amounts of large, coarse and fine PM decreased after the first simulated precipitation by 52%, 10% and 10% respectively. In *B. pendula* and *Q. robur*, the deposition of large and fine PM was lower after the first artificial rain by 26% and 37% and 40% and 46% respectively, while surprisingly deposition of coarse PM increased by 12% and 20%.

Deposition of surface PM ($s\text{PM}$) and in-wax PM ($w\text{PM}$) on plant foliage was affected differently by accumulation periods and treatments with artificial rain (Fig. 3B). The greatest increase in accumulation of both PM categories was recorded during the summer accumulation period and amounted to 68%–77% and 41%–60%, respectively, for $s\text{PM}$ and $w\text{PM}$. During the two remaining accumulation periods (spring and autumn), the amounts of $s\text{PM}$ and $w\text{PM}$ also increased, but to a lesser extent. This was especially evident in the autumn accumulation period for $w\text{PM}$, which increased in *T. cordata* by 5%, while it increased in *Q. robur* by 20%. It was easier for the artificial rain to wash off $s\text{PM}$ than $w\text{PM}$. On average, artificial precipitation removed 61%, 64% and 72% of $s\text{PM}$ and 30%, 46% and 26% of $w\text{PM}$ from plant foliage in *B. pendula*, *T. cordata* and *Q. robur* respectively. At the end of the experiment, the amount of $s\text{PM}$ had increased by 2.5-fold in *B. pendula* and *T. cordata*, and by 5.5-fold in *Q. robur*, while the amount of $w\text{PM}$ was 2–2.5-fold higher in all species compared with the amounts at the start of the experiment.

The amount of epicuticular waxes on *B. pendula* and *Q. robur* foliage did not differ significantly, but it was evident that over time the amount of waxes slightly increased in *B. pendula* and decreased in *Q. robur* (Fig. 3B). In contrast,

Table 2

Amount of large PM (10–100 μm), coarse PM (2.5–10 μm) and fine PM (0.2–2.5 μm), and amount of epicuticular waxes on the leaves of *B. pendula*, *Q. robur* and *T. cordata* growing in Natural Rain and No Rain conditions. Data are means \pm SE, $n = 12$

Species	Treatment	Date of harvest	PM size fraction ($\mu\text{g cm}^{-2}$) (mean \pm SE)			Epicuticular waxes ($\mu\text{g cm}^{-2}$) (mean \pm SE)				
			10–100 (μm)	2.5–10 (μm)	0.2–2.5 (μm)					
<i>Betula pendula</i>	Natural Rain	June	16.5 (± 1.6)	9.1 (± 1.1)	9.2 (± 0.6)	908.0 (± 36.4)				
		July	28.1 (± 0.3)	11.7 (± 0.5)	13.1 (± 0.2)	846.4 (± 28.2)				
		August	27.9 (± 1.1)	22.6 (± 0.1)	23.5 (± 0.6)	897.5 (± 37.2)				
	No Rain	September	32.1 (± 1.0)	17.2 (± 1.2)	12.1 (± 0.2)	871.8 (± 14.0)				
		June	15.9 (± 1.0)	8.6 (± 0.4)	11.2 (± 0.5)	932.2 (± 31.0)				
		July	25.3 (± 0.1)	18.7 (± 0.8)	16.0 (± 0.5)	853.7 (± 39.9)				
<i>Quercus robur</i>	Natural Rain	August	40.6 (± 1.9)	23.2 (± 1.9)	26.2 (± 1.0)	907.8 (± 32.2)				
		September	39.9 (± 2.3)	18.4 (± 0.8)	19.6 (± 0.8)	910.6 (± 54.6)				
		June	8.4 (± 1.4)	5.6 (± 0.5)	1.2 (± 0.1)	86.6 (± 4.4)				
	No Rain	July	20.7 (± 0.6)	11.4 (± 0.6)	3.0 (± 0.4)	87.6 (± 0.1)				
		August	22.6 (± 1.7)	11.8 (± 1.0)	5.1 (± 0.5)	87.2 (± 2.1)				
		September	18.1 (± 0.6)	11.1 (± 0.5)	4.9 (± 0.1)	59.5 (± 4.8)				
<i>Tilia cordata</i>	Natural Rain	June	6.0 (± 1.1)	7.7 (± 0.2)	1.6 (± 0.2)	89.6 (± 9.2)				
		July	18.4 (± 1.8)	12.0 (± 1.2)	5.8 (± 0.5)	92.0 (± 9.3)				
		August	29.2 (± 0.8)	19.8 (± 0.6)	8.2 (± 0.4)	83.8 (± 0.6)				
	No Rain	September	28.3 (± 1.6)	19.3 (± 0.1)	5.0 (± 0.5)	63.2 (± 2.9)				
		June	9.2 (± 0.4)	5.7 (± 0.4)	2.5 (± 0.2)	48.0 (± 2.6)				
		July	15.6 (± 0.5)	7.4 (± 0.3)	5.8 (± 0.2)	55.0 (± 1.3)				
ANOVA			<i>F</i>	<i>P</i>	<i>F</i>	<i>P</i>	<i>F</i>	<i>P</i>		
	Species		196.7	<0.0001	84.01	<0.0001	1634	<0.0001	2582	<0.0001
	Treatment		108.7	<0.0001	220.3	<0.0001	108.9	<0.0001	0.266	0.6156
Harvest		187.5	<0.0001	157.9	<0.0001	235.3	<0.0001	1.151	0.2283	
Species \times treatment		6.222	0.0140	20.61	0.0001	19.83	0.0002	0.482	0.6285	
Species \times harvest		5.899	0.0002	4.840	0.0010	43.58	<0.0001	2.222	0.0632	
Treatment \times harvest		19.85	<0.0001	11.60	<0.0001	12.15	<0.0001	0.137	0.9376	
Species \times treatment \times harvest		8.027	<0.0001	8.628	<0.0001	9.425	<0.0001	0.087	0.9972	

a significant gradual increase in the amount was recorded in *T. cordata* (Fig. 3B).

4. Discussion

4.1. Effect of natural rain on PM deposition on foliage

Vegetation is essential for the proper functioning of urban areas [16,46]. Of particular interest are trees, which have a large biologically active biomass acting as “green livers” in cities [15–17]. In this work, the ability of trees to accumulate PM was confirmed [23,44]. Birch turned out to be the species that accumulated the most PM, which can be explained by the large amount of wax on its leaves [14,44]. Moreover, leaves of *B. pendula* are smaller than those of *Q. robur* and *T. cordata*, which may also increase their phytoremediation potential [32,47]. The key role of

leaf morphology in PM accumulation processes has been emphasised by many authors [20,21–23]. PM accumulation is influenced by leaf type, size and surface complexity, the amount of wax and the presence of trichomes [32]. These properties also result in greater PM retention on foliage [26].

The accumulation of PM on plants is an extremely dynamic process. The amount of PM on plants can vary over the course of individual days [37] and even hours [48]. Nguyen et al. [48] demonstrated that under specific experimental conditions the accumulation of $\text{PM}_{2.5}$ on foliage is greatest in the morning and decreases in the evening due to human activities and environmental factors. PM deposition on plants is largely dependent on weather conditions [26,27,37]. First, wind can disperse PM, while rain can wash away PM present in the air [37], which makes the pollutants impossible to neutralise by plants. Wind and

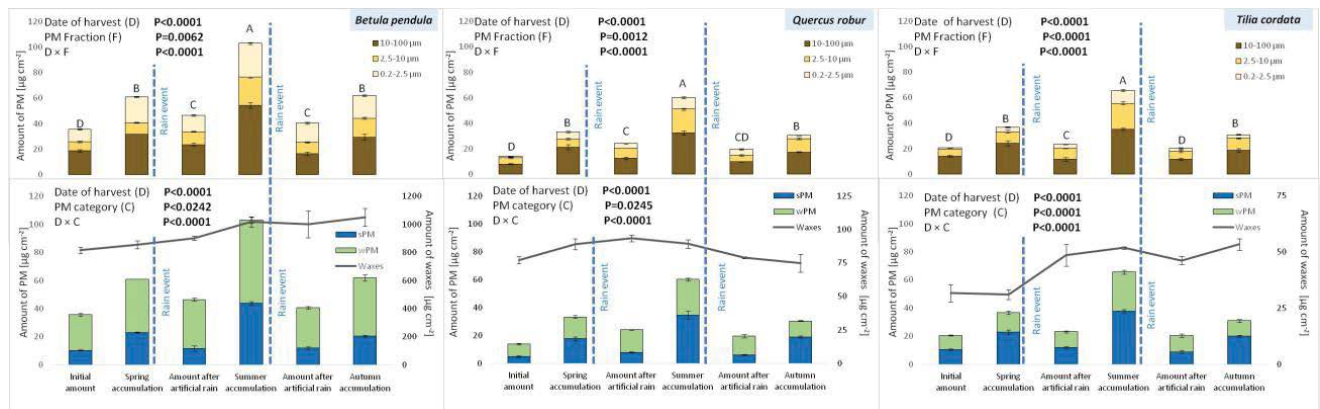


Fig. 3. Amount of (A) large PM (10–100 μm), coarse PM (2.5–10 μm) and fine PM (0.2–2.5 μm), and (B) surface PM ($s\text{PM}$), in-wax PM ($w\text{PM}$) and epicuticular waxes on leaves of *B. pendula*, *Q. robur* and *T. cordata* during three accumulation periods interrupted by artificial rain events. Different uppercase letters indicate a significant difference between harvest dates at $P < 0.05$ by Tukey's HSD test. Analysis of variance (ANOVA) assesses the statistical significance and " P " values < 0.05 are in bold. Data for both PM and waxes amount are means \pm SE, $n = 12$.

rain also have a significant effect on PM already accumulated on plants. A large proportion of the PM deposited on plant foliage is accumulated only temporarily and can be re-suspended by wind or precipitation [31,32,37,41]. Up to 27%–36% of PM deposited on plants can be blown off by strong winds [27]. Each rain event washes PM off the plant surface, but the intensity of this process depends on many factors, for example, the intensity and duration of the rainfall, the size and chemical composition of the PM, and the morphological structure of the plant [27,29,31,35–38]. Rain can remove 20%–70% of previously accumulated PM (mainly large PM with a diameter of 10–100 μm) from plants [38,39]. Also in the present study, rain affected the amount of PM deposited on the plants. Trees of all the examined species protected against the rain accumulated more PM (throughout the growing season) than plants exposed to precipitation. It should be emphasised that rain was not simulated with distilled water in this study and PM was washed off by natural rainfall, which better demonstrates the phenomenon taking place under realistic conditions. PM washed off trees by rain may be deposited on vegetation below [37,39,46,48,49] or settle on a paved surface from which it can be re-suspended in the air, continuing to threaten people and the environment [31,48]. In this study, the influence of rain on PM accumulation was investigated on leaf samples harvested from the entire cross-section of the crown, therefore the results obtained do not allow for clarification of whether the amount of PM washed away from the top of the tree differed from that from the depth of the crown, for example, or whether PM from the top was accumulated by the leaves underneath.

PM can be accumulated on the surface of leaves or permanently retained in the waxes [14,15,44,47]. Surface PM is only loosely attached to the foliage, and can be relatively easily removed and re-suspended [38] if the leaf is not irregular and/or wrinkled or is not covered by various morphological structures (trichomes and fungal hyphae) [23,26,50]. In contrast, lipophilic PM is permanently bonded to the wax and is very difficult for rain to wash off [26,37,44,49]. Moreover, PM embedded in wax cannot

aggregate into larger particulates that can be easier to remove from foliage [37,41]. The positive effect of wax on the retention of PM on leaves decreases with the end of the growing season, when the wax gradually deteriorates [51]. In this experiment, regardless of the species examined, the amount of surface and in-wax PM was higher on the trees protected from rain. It is worth emphasising that the greatest difference between plants protected from rain and those exposed to rainfall was found for surface PM. In-wax PM is less dependent on rain and, regardless of the weather conditions, its amount on the leaves fluctuates less and grows systematically until the end of the growing season. The amount of PM on the plants increased in the following months independently of the rain conditions (No Rain and exposure to natural precipitation). The increase in PM deposition on plants exposed to rain during the growing season has previously been demonstrated by other authors [27,37,41]. In the present study, the amount of surface PM increased more, especially in July (*B. pendula*, *Q. robur*) and September (*T. cordata*), than in-wax PM. This is a very interesting result because the increase in surface PM deposition was not dependent on the amount of rainfall (July: high, September: low). To some extent, this can be explained by the fact that in the summer period, the leaves are fully developed and have the greatest phytoremediation capacity (they accumulated the most PM). These results suggest that in order to make full use of plants for air purification, urban greenery should be organised in such a way that plants are fully developed for as long as possible or that individual species develop at different times.

The amount of PM size fractions (10–100, 2.5–10 and 0.2–2.5 μm), regardless of the species and harvest date, was usually higher on plants protected from the rain. The fraction ratio (large PM:coarse PM:fine PM) differed only slightly between plants exposed and not exposed to natural precipitation. This suggests that rain washes PM of every size fraction off leaves. However, most of the washed-off PM was large PM, and the least was fine PM. This has previously been presented by other authors [27,32,41]. Large PM is not only greater in diameter, but usually also heavier. For this

reason, large PM can be removed from the plant surface by rain and wind more easily. Fine PM may also be more likely to be stuck between hairs or different wax structures. According to Schaubroeck et al. [31], a maximum of only 24% fine PM can be washed off foliage.

4.2. Dynamics of PM accumulation

The assessment of a plant's ability to accumulate PM is usually undertaken once, mostly at the end of the growing season [12–15,45]. This allows an estimation of how much PM has been permanently removed from the environment along with fallen and disposed leaves. Unfortunately, this approach does not allow the processes of PM accumulation, retention and re-suppression to be fully understood. The air-cleaning potential of plants is also underestimated since PM that is deposited on plants during the growing season but removed from foliage (e.g., washed off by rain or wind-blown) is not included in the estimate. As a result, the real effectiveness of air phytoremediation is unknown. The present study showed that PM accumulation by plants is carried out from the outset of the growing season, even on the youngest, not fully developed leaves. The largest amount of PM throughout the experiment was found on the foliage of *B. pendula*, therefore on a species that starts the growing season early (the earliest of the species studied) and accumulates PM for longer than the other plants examined. In all species, the accumulation of PM was interrupted by the artificial precipitation treatment. Even though rain washes off up to 70% [38] of accumulated pollutants, this PM is not taken into account when assessing the phytoremediation potential of plants. In this study, after taking into account the amount of total PM that was washed from the plant foliage, the amount of PM that plants have actually removed from the air was 55% (*B. pendula*), 66% (*T. cordata*) and 62% (*Q. robur*) greater than the amount deposited on leaves at the end of growing season. When analysing individual PM size fractions, the underestimation of PM accumulation was at the level of 61%, 45% and 51% (*B. pendula*), 66%, 61% and 74% (*T. cordata*), and 64%, 53% and 72% (*Q. robur*) for large, coarse and fine PM respectively. The amount of surface PM was reduced by 67% (a similar value for all species), while in-wax PM was reduced by 45% (*B. pendula*), 50% (*Q. robur*) and 63% for (*T. cordata*). It should be noted that under natural conditions these differences will be much greater, because in this experiment the plants were treated with simulated rainfall only twice.

In this study, the amount of PM washed off the plants depended more on the amount of accumulated PM than on the plant species. Rain duration and intensity were not examined. The rain washed off the most PM in summer when the amount of PM on foliage was highest. The smaller amount of PM washed off leaves in spring could also be due to the greater stickiness of young leaves. From *B. pendula* and *Q. robur*, simulated precipitation washed off mostly large PM and washed off fine PM least. These results are in line with the findings of Przybysz et al. [41] and Mo et al. [52] who suggest that fine PM can permanently stick to the surface of leaves and cannot be washed off by rain. Different results were recorded for *T. cordata*,

where the reduction in the amounts of all PM size fractions was similar after rain. The key role of plant morphological features in PM retention during rain events has previously been demonstrated by Wang et al. [27]. Morphological structures present on leaves (hairs, wax structure) not only hold on to PM, but also decrease the kinetic energy of raindrops striking the leaf surface and thus are not enough to wash off PM [53]. An interesting phenomenon was recorded on the leaves of *B. pendula* and *Q. robur*, on which the amount of coarse PM increased after the first treatment with artificial rain. This can be explained by the fact that rain droplets may contain dissolved salts, which will become the new PM after the water has evaporated [27]. This phenomenon did not occur in *T. cordata*, probably because this species did not retain the droplets on the leaf surface. However, this was not investigated in the present study. Furthermore, small water-insoluble particles (smaller than PM) can also stick together and form larger particles in the presence of water [54]. As expected, simulated rain was much easier to wash off surface PM than PM embedded in wax.

After each artificial rain treatment, new PM accumulation was observed. This is in line with the results of Popek et al. [37] who also found that plants accumulate PM efficiently after rain. This may be due to the fact that the leaves are wet and stickier after rain [27,37,41]. In this work, the efficiency of PM re-accumulation depended on the season. The largest amount of PM (total, large, coarse, surface and in-wax) was deposited on leaves in the summer, probably because in addition to common pollutants (from transport, industrial or construction sources) the air also contains sand particles, pollen, fungal spores and fragments of plants and animals. Moreover, in summer, the leaves are mature, with fully developed hairs and wax on their surfaces. Wang et al. [55] showed that immature leaves retain a smaller amount of PM, and mature (but not too old) leaves have a greater ability to capture PM. The smallest accumulation of total PM was recorded in the autumn, when the leaves were getting older and losing their properties. In contrast to total, large, coarse, surface and in-wax PM, the highest accumulation of fine PM (the most dangerous PM) was in the spring. Early spring is still the heating season, which means that the main source of fine PM is active, and there is also more car traffic than in the summer. In spring, the smallest PM may also clog up empty spaces on leaves, leaving no room for new PM accumulation later. At the end of growing season, the accumulation of fine PM was negligible, while in *Q. robur* a decrease in the smallest PM deposited on foliage was recorded. It is likely that the leaves underwent a slow aging process, losing their stickiness and degrading the wax. Chen et al. [56] also found that leaves had a decreased phytoremediation capacity at the end of the growing season. It is noteworthy that the amount of surface PM at the end of growing season increased to a greater extent than in-wax PM compared with the beginning of experiment. This is a surprising result because surface PM proved to be easier to wash off. This can be explained by the fact that surface PM is also accumulated more quickly by plants than in-wax PM, and its deposition on foliage undergoes significant fluctuations during the growing season [37].

4.3. Dynamics of wax content on foliage

The amount of wax on the plants did not change greatly during the experiment. Only in the case of *Q. robur* the amount of wax decreased slightly at the end of growing season. It can be assumed that in this study, apart from its probable degradation at the end of the growing season, wax did not play a decisive role in the processes of accumulation, retention and re-accumulation of PM in the species studied. It should be noted that this work did not study the chemical composition and morphology of waxes. Plant species may differ greatly in the type of waxes [44,51]. Wax also changes over the growing season and may be affected differently by meteorological conditions. For instance, high temperatures in summer can partially melt wax and increase its viscosity, leading to the higher PM accumulation recorded in this study in July.

5. Conclusions

Weather conditions, particularly rain, are of great importance in the accumulation of PM, which is an extremely dynamic process. PM deposited on foliage can be retained for a long period of time or can be washed off foliage and re-suspended. After PM re-suspension, new PM accumulation is possible. However, not all PM is removed from plants by rain. Although there are many rainfall events during the growing season, PM is present on leaves all the time. Therefore, the assessment of the efficiency of PM accumulation by plants on the basis of the amount of PM deposited on leaves at the end of growing season only is very much an underestimation and does not reflect the true ability of trees to clean the air. It is very important to include PM washed off foliage in calculations in order to understand the realistic potential of plants in air purification.

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