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How Much Does Weather Matter? Effects of Rain and Wind on PM Accumulation by Four Species of Australian Native Trees

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Abstract: As interest in improving urban air quality grows, phytoremediation—amelioration through plants—is an increasingly popular method of targeting particulate matter (PM), one of the most harmful pollutants. Decades of research has proven that plants effectively capture PM from air; however, more information is needed on the dynamics of PM accumulation. Our study evaluated the effects of meteorological conditions on the dynamics of PM deposition, wash off and resuspension using four Australian tree species growing under natural conditions near a busy highway. Accumulation of PM on foliage was analyzed over the short term (daily changes) and over a longer time period (weekly changes). The results obtained were correlated with ambient concentrations of PM_{2.5} and PM₁₀, rain intensity and wind strength. The highest accumulation of PM was recorded for *Eucalyptus ovata* (100.2 µg cm⁻²), which also had the thickest wax layer while the lowest was for *Brachychiton acerifolius* (77.9 µg cm⁻²). PM accumulation was highly changeable, with up to 35% different PM loads on the foliage from one day to the next. Importantly these dynamics are hidden in weekly measurements. Changes in PM deposition on the leaves was mostly affected by rain and to a lesser extent by wind, but the extent of the effect was species specific. The large PM fraction (10–100 µm) was the first to be removed from leaves, while the smallest PM fraction (0.2–2.5 µm) was retained for longer. Precipitation affects also PM retained in waxes, which until now were believed to be not affected by rain. This work demonstrates important interactions between PM load and weather, as well as adding to the small inventory of Australian native tree PM accumulation data.

Keywords: air quality; urban; sustainability; vegetation; PM; phytoremediation

1. Introduction

With 68% of the world's population predicted to live in urban areas by 2050, city planners worldwide are increasingly concerned about the declining air quality that comes with such increased population density. Among mechanisms used to improve air quality is phytoremediation: using various characteristics of living plants to reduce ambient pollutant concentrations. One of the most important targets of these phytoremediation efforts is particulate matter (PM) [1].

These microscopic pollutants consist of particles with diameters between 0.001 and 100 µm in a heterogeneous solid-liquid mixture [2]. Depending on size, PM can be suspended in the air for weeks [3] and travel great distances, as illustrated by the discovery in 2006 that 11% of black carbon found in the western United States was produced in China [4]. In terms of source, we distinguish between PM of natural origin such as forest fires, volcanic activity or dust storms and anthropogenic causes such as

incomplete combustion of fuels in car engines, abrasion of roads and tyres, and industrial or construction activities [5–7]. Chemically, PM may be composed of several trace metals, black carbon, dibenzofurans, polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons [8,9]. Chronic exposure to PM is the cause of numerous health problems including cardiopulmonary diseases and lung cancer [10,11] and is responsible for around 2.1 million premature deaths annually over the world [12].

Urban plantings such as trees and shrubs with dense twigs and leaves can effectively capture PM from ambient air [13,14] or be a physical barrier preventing its spread [15,16]. It was estimated that deciduous trees and shrubs forming street canyons can reduce PM concentration in the air by as much as 60% [17]. However, species differ in their ability to accumulate PM [14,18,19]. Plants with rough leaf surfaces are more effective in capturing PM than those with smooth leaves [20]. Accumulation of PM on the leaves can be also increased by leaf surface structures such as hairs and a wax layer [21–23]. While it is well established that species may vary in their entrapment efficiency, only a handful of species have been tested, and many of these are in selected countries and regions only. For instance, in Australia, only 16 species, all found in the Greater Sydney Region, have been investigated for PM capture [24].

Another area ripe for investigation is the variation of PM accumulation over time. Most studies, for instance, Sæbø et al. (2012) [25], Popek et al. (2013) [26], and Xu et al. (2018) [27], demonstrate the ability of trees and shrubs to accumulate PM by measuring PM deposition at a given time, usually at the end of the growing season. However, PM deposition, its runoff and resuspension from foliage is a complicated and dynamic process, greatly affected by weather conditions, especially precipitation and wind [28]. How much PM is lost from foliage depends on rain intensity and duration, wind speed and direction and other meteorological factors, as well as season and plant species, PM concentration in the air and type of PM [28–32]. The overall effect of weather depends on intensity: heavy rain and strong wind may remove PM from leaf surfaces, while lighter rain and weaker wind may increase PM load on vegetation. This is because leaves can become more sticky, and more likely to trap PM on first wetting, but heavy rain will effectively wash a leaf and clean off PM [33–35]. Schaubroeck et al. (2014) [35] used a model to demonstrate that a Scots pine (*Pinus sylvestris* L.) stand accumulated 31.43 kg ha^{−1} year^{−1} PM_{2.5} via dry deposition, of which 23.93 kg was resuspended, 7.38 kg was considered as definitely removed by dripping off the canopy to the forest floor and only 0.11 kg remained in the tree canopy at the end of the year. Removal of PM from foliage surface by rain and wind is however not necessarily an undesirable phenomenon because leaf surfaces may become saturated with PM over time and their phytoremediation capacity would thus be greatly reduced [36].

Precipitation and wind not only impact deposition of PM, but also the concentration of PM in the air [20,37]. Greater wind speed creates turbulence, resulting in dispersion of PM in the air [38], while wind direction affects the spatial distribution of PM and the direction it is transported [28]. Precipitation is one of the primary natural processes reducing PM in the ambient air [39]; its scavenging effects result from wet deposition onto surfaces such as foliage and wet removal from the air [40]. Zhang et al. (2018) [28], showed that exactly how wind and precipitation impact PM concentration depends on PM size. Fine PM (0.2–2.5 µm) decreased gradually with increasing wind speed, while coarse PM (2.5–10 µm) increased in concentration due to resuspension in strong wind. The purifying efficiency of precipitation on coarse PM was over twice that of fine PM. It is also important to note that other meteorological conditions will affect PM concentration in the air, and these include air temperature, humidity and vertical inversions in the lower troposphere [41].

While it is appreciated that PM deposits change in concentration due to runoff and resuspension, most investigations into the effects of rain and wind have been carried out in controlled environments, rather than in the field. So although this is a growing field of research, knowledge about the dynamics and life cycle of PM remains scattered and incomplete.

This work aims to evaluate the effect of meteorological conditions (precipitation, wind, concentration of PM in ambient air) on the dynamics of PM deposition on four Australian woody species over long and short time periods under natural conditions. We tested the following hypotheses:

(i) Under natural conditions, the amount of PM deposited on foliage changes significantly over short time periods (one day) compared to over longer periods (one week), (ii) atmospheric conditions (rain and wind) affect the amount of PM on the leaf surface, and (iii) PM capture varies across the four plant species.

2. Methods

2.1. Study Area and Plant Material

The study took place in April–May 2018 at the University of Wollongong's main campus on the south east coast of Australia (Figure 1).

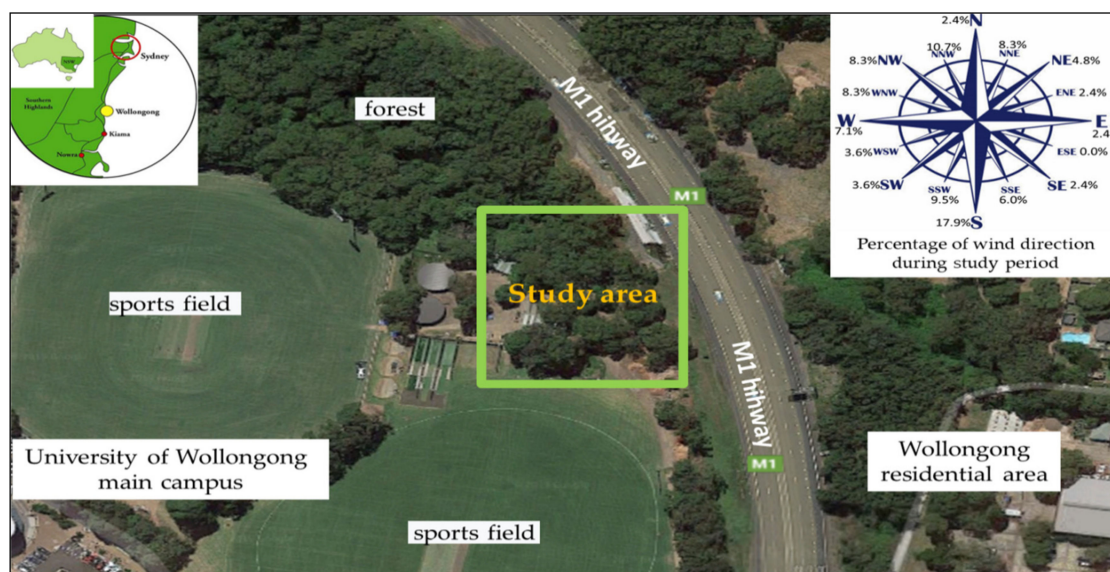


Figure 1. Map showing location of the study area with a wind rose showing percentage of wind direction during the study period. Satellite image from Google Maps; Google [42].

Wollongong is the third largest city in New South Wales. According to the World Health Organization, its annual average PM_{10} concentration is $17 \mu g m^{-3}$ while $PM_{2.5}$ is $7 \mu g m^{-3}$, positioning Wollongong among medium polluted Australian cities, but among the least polluted cities when compared to Asia and Europe (data from 2016) [43]. The study site was located in the north-west of the campus, 50 m from the M1 highway (Princes Motorway). According to a station three km to the north, traffic volume in 2018 was 53,083 cars per day (54,553 on weekdays and 48,832 at weekends) [44]. In our study, the M1 highway was considered to be the major source of pollution, because 71.4% of time the wind was blowing from the direction of the highway. Selected plants were located in the lower part of a forest remnant. On the west side of study area were warehouses and two sport fields, while the nearest university buildings were about 400 m away. On the other side of the highway were residential buildings. The plants selected for this study were four Australian native woody species common to NSW. The Illawarra Flame Tree (*Brachychiton acerifolius* [A. Cunn. ex G. Don] Macarthur) has large, 3-5-lobed leaves which are 15 cm long and 12 cm wide. The Swamp Gum (*Eucalyptus ovata* Labill.) has rounded leaves, 19×8.5 cm. The Large Mock-olive (*Notelaea longifolia* Vent.) has narrow lanceolate leaves (16×6 cm) and Sweet Pittosporum (*Pittosporum undulatum* Vent.) has smooth and glossy leaves (15×5 cm) with wavy margins. The leaves of all selected species were hairless with non-serrated edges. All plants had been growing in vivo in the selected location and were approximately the same age (about 30 years old) and size., except for *E. ovata*, which was about five years old. All trees used were in good condition, i.e., healthy and free from pests.

2.2. Sample Collection

For the experiment, three samples of leaves of each species were harvested every week from 12 April 2018 to 10 May 2018 (five weeks) and daily from 27 April 2018 to 3 May 2018 (seven days) at 12 pm. Samples were harvested from the side of the tree that faced the road, at a height of 1.0 to 1.8 m above the ground, which corresponds to the height at which most air is inhaled by humans, and stored in paper bags at ambient temperature until analysis. Each sample consisted of three to seven leaves, depending on size, (for weeks $n = 4$ plant species \times 3 samples \times 5 weeks; for days $n = 4$ plant species \times 3 samples \times 7 days). Leaves were harvested at the same time of the day, regardless of weather conditions.

2.3. Quantitative Analysis of PM and Leaf Waxes

We followed a standard protocol for PM isolation by filtration [18]. For each sample, PM of three size fractions and two categories were determined. Each leaf sample was washed with water to obtain surface PM (sPM), which in natural conditions can be washed off by rain. Next, samples were washed with chloroform to obtain the PM contained in the epicuticular waxes (wPM). Both liquids were then passed through a 100 μm mesh sieve, before being filtered through membrane filters (Whatman, UK) with three pore sizes 10, 2.5 and 0.2 μm , respectively. This yielded three size fractions of PM: large (10–100 μm), coarse (2.5–10 μm) and fine (0.2–2.5 μm). The quantity of waxes was weighed after the evaporation of chloroform and collected (after filtration) in pre-weighed beakers. The area of the leaves in samples was measured with a LI-3000C Portable Area Meter (Lincoln, NE, USA). Finally, PM amounts and waxes were calculated to $\mu g \cdot cm^{-2}$ of leaf area.

2.4. Measurement of PM Concentration in Air and Ambient Weather Conditions

Measurements of ambient PM_{10} and $PM_{2.5}$ concentrations were made using a TSI DustTrak DRX 8533 particulate monitor (Keison Products, UK). The measurements were made at three-minute intervals over one month. The instrument was placed 1.2 m above the ground, 50 m from the road, near the trees from which leaves were collected. Rainfall, wind speed and direction statistics were obtained from the nearest Bureau of Meteorology station [45], situated 10 m above sea level, in Bellambi about 4 km from the study area.

2.5. Statistical Analysis

One and two-way Analysis of Variance (ANOVA) with random effects were used to compare measurements of particulate matter (total PM, sPM , wPM in three size fractions as well as amount of waxes). Non-parametric Spearman's correlation coefficients were calculated between the amounts of different types of PM on leaves, in the air and also weather conditions. Tukey's honest significance (HSD) test ($P = 0.05$) was employed to assess the significance of differences among variants. The data are given as means with standard errors of the mean (\pm SE). Statistical analyses were conducted in JMP 13.0 Pro (SAS Institute Inc., Cary, NC, USA).

3. Results

3.1. Particulate Matter Accumulation

3.1.1. General PM Accumulation and Amount of Waxes

All species examined have the ability to accumulate PM on the leaf surface (Figure 2). On average, the highest amount of PM was accumulated by *E. ovata*. The amount of PM on the foliage of *E. ovata* exceeded by 10%, 15% and 20% PM accumulation on *P. undulatum*, *N. longifolia* and *B. acerifolius* respectively. All tested species accumulated most PM as sPM (Figure 2), the percentage of sPM was 55% in *B. acerifolius* and *P. undulatum*, while in *N. longifolia* it was 53% and, at 52%, slightly lower in *E. ovata*. The tree species examined also differed in the amount of waxes on the leaf surfaces (Figure 2).

The heaviest wax layer was found on the leaves of *E. ovata*, it was higher by 88%, 73% and 59% than on leaves of *B. acerifolius*, *P. undulatum* and *N. longifolia* respectively.

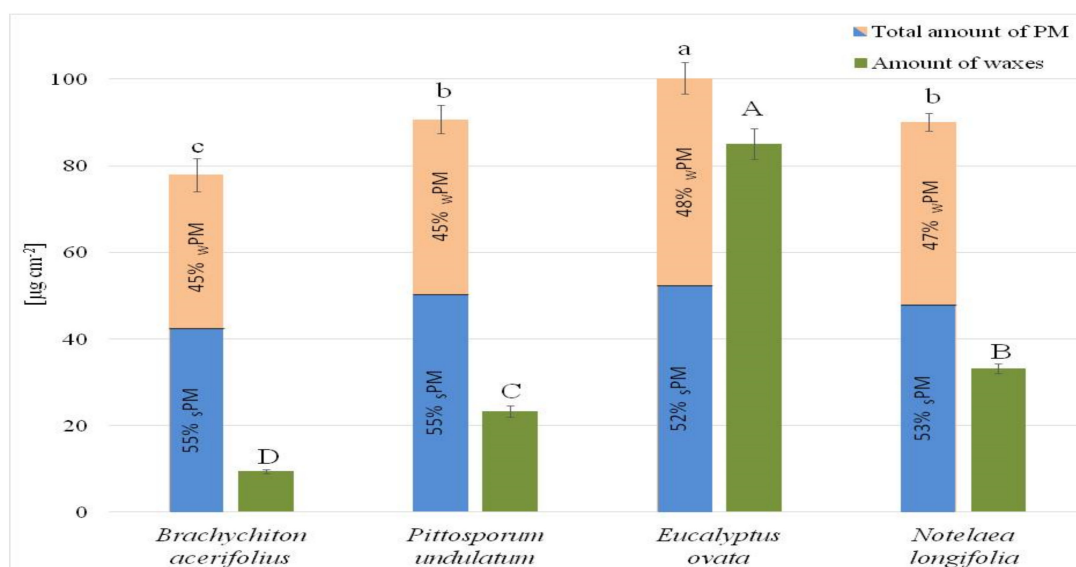


Figure 2. Average amount of PM (divided into sPM and wPM) and waxes on leaves of *B. acerifolius*, *P. undulatum*, *E. ovata* and *N. longifolia* over five weeks. PM and waxes values are expressed as µg per cm² of leaf. Data are means ± SE, *n* = 15. Different letters (lowercase for PM and capital for waxes) indicate statistical significance (*P* < 0.05).

3.1.2. Accumulation of Different PM Fractions during the Sampling Period

The amounts of all size fractions deposited on plant foliage changed significantly over the short term (days) for all species except for fine PM fraction on *E. ovata* leaves. Only large- and coarse-sized PM varied significantly in leaf deposition over the long term (weeks, Figures 3–5). For large PM size, *P. undulatum*, *B. acerifolius*, and *E. ovata* all varied over the five weeks (Figure 3b), while for coarse PM, only *P. undulatum* and *B. acerifolius* varied significantly (Figure 4b).

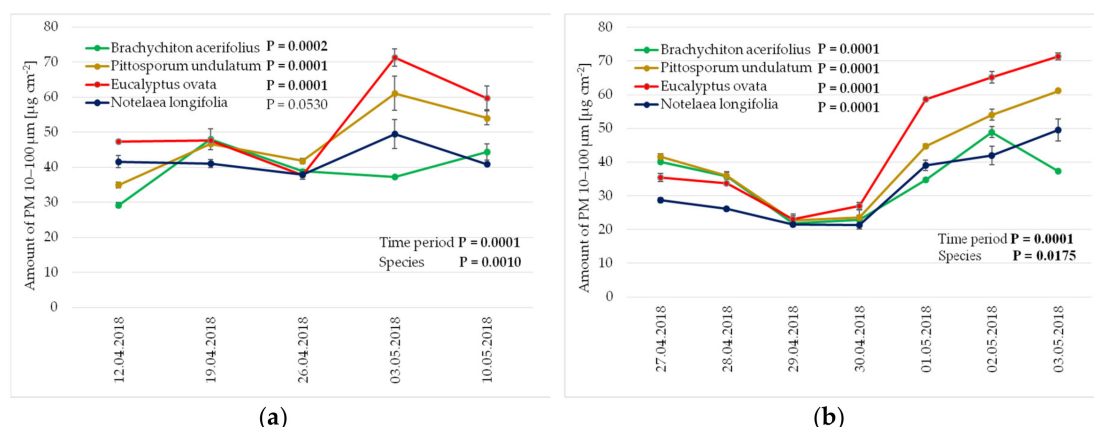


Figure 3. Amount of large PM fraction (10–100 µm) PM deposited on leaves of *B. acerifolius*, *P. undulatum*, *E. ovata* and *N. longifolia* during (a) five weeks and (b) seven days of measurements. Data are means ± SE, *n* = 3. *P* values in bold indicate statistical significance.

Two clear peaks, when the level of large PM increased considerably compared to previous weeks, were recorded on 19 April 2018 (increases of 39%, 25% and 2% compared to the previous week in *B. acerifolius*, *P. undulatum* and *E. ovata* respectively) and on 3 May 2018 (an increase of 48% and 32% compared to the previous week in *E. ovata* and *P. undulatum* respectively). These two peaks were

followed by decreases in large PM loads on the foliage in most species (Figure 3a). A seven-day intensive period of daily monitoring of large PM deposition on foliage, within the five-week period, revealed finer detail of the significant differences in accumulation of PM fraction 10–100 μm during the second increase in PM described above (Figure 3b). These revealed a similar higher content of large PM in different leaf samples over the weekly interval (more large PM on 3 May 2018 than 26/27 April 2018), but also showed a gradual decrease (43% on average) in large PM for the first three days after 27 April 2018 followed by a rapid increase between 30 April 2018 and 3 May 2018 (Figure 3b).

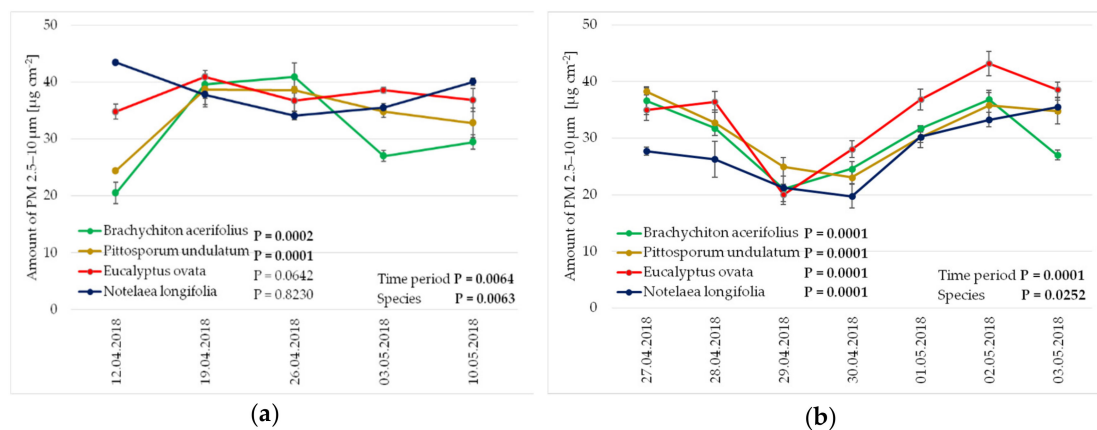


Figure 4. Amount of coarse PM fraction (2.5–10 μm) PM deposited on leaves of *B. acerifolius*, *P. undulatum*, *E. ovata* and *N. longifolia* during (a) five weeks and (b) seven days of measurements. Data are means \pm SE, $n = 3$. P values in bold indicate statistical significance.

Coarse PM amounts on *B. acerifolius* and *P. undulatum* leaves reached a maximum between 19 April 2018 and 26 April 2018 (a 48% and 37% increase over the previous week in *B. acerifolius* and *P. undulatum* respectively), and then dropped in both species in the following weeks (Figure 4a). During the seven-day period, significant changes in the amounts of coarse PM were recorded in all species (Figure 4b). As in the case of weekly measurements, when measured daily, amounts of PM 2.5–10 μm deposited on foliage on 26/27 April 2018 and 3 May 2018 were similar, just slightly higher on the last date. Daily measurements, however, showed a significant decrease in coarse PM on foliage of all species between 28 April 2018 and 29 April 2018 (decreases between 35% and 49%, *P. undulatum* and *B. acerifolius*, respectively), with the load increasing from 1 May, reaching a peak on 2 May 2018 (Figure 4b).

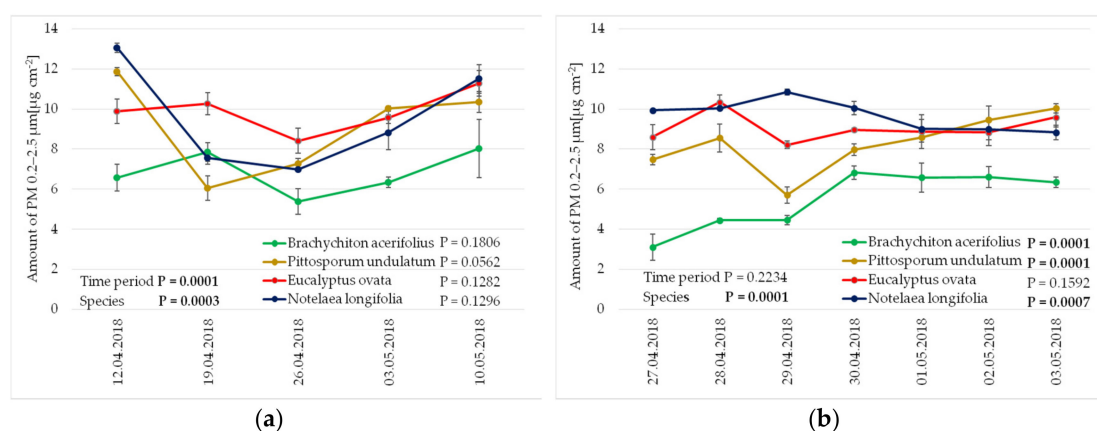


Figure 5. Amount of fine PM fraction (0.2–2.5 μm) PM deposited on leaves of *B. acerifolius*, *P. undulatum*, *E. ovata* and *N. longifolia* during (a) five weeks and (b) seven days of measurements. Data are means \pm SE, $n = 3$. P values in bold indicate statistical significance.

Accumulation of fine PM (0.2–2.5 μm) did not differ significantly between species during weekly measurements (Figure 5a). Daily measurements only showed a decrease in PM deposition on *P. undulatum* (33%), followed by an increase, a gradual increase up to 30 April for *B. acerifolius*, a slight increase in *N. longifolia* (8%) on 29 April 2018 (Figure 5b) and no change for *E. ovata*.

3.1.3. Accumulation of $s\text{PM}$ and $w\text{PM}$ during Time Period

The amounts of $s\text{PM}$ and $w\text{PM}$, measured both weekly and daily (Figure 6a–d), followed similar trends as recorded for large PM (Figure 3).

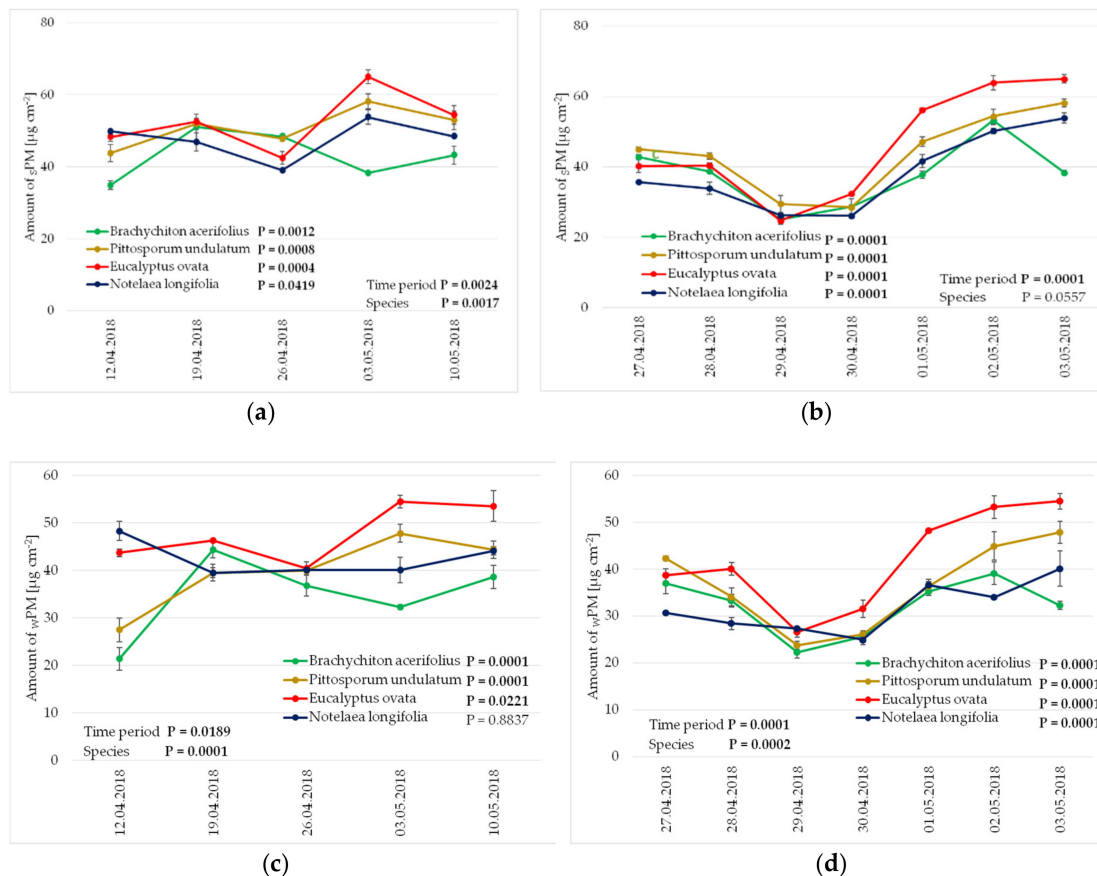


Figure 6. Amount of $s\text{PM}$ in (a) a five-week period and (b) a seven-day period, and $w\text{PM}$ in (c) a five-week period and (d) a seven-day period on leaves of *B. acerifolius*, *P. undulatum*, *E. ovata* and *N. longifolia* during measurement period. Data are means \pm SE, $n = 3$. P values in bold indicate statistical significance.

Accumulation of $s\text{PM}$ and $w\text{PM}$ measured in weekly intervals on leaves of *E. ovata* and *P. undulatum* peaked twice (on 19 April 2018 and 3 May 2018), and in both cases this was followed by decreased PM load (Figure 6a,c). In the case of *N. longifolia* and *B. acerifolius* recorded changes in foliar PM deposition were less clear and not always significant (Figure 6a,c). Amounts of $s\text{PM}$ and $w\text{PM}$ obtained 26/27 April 2018 and 3 May 2018 were similar regardless of whether the measurements were made on a weekly or daily basis. Trends recorded during measurements taken in the seven day period were the same for all species (Figure 6b,d). During the first three days of measurements, $s\text{PM}$ and $w\text{PM}$ gradually decreased and reached the lowest accumulation on 29/30 April 2018. Subsequently $s\text{PM}$ and $w\text{PM}$ increased until a peak on 2/3 May 2018. The average difference between lowest and highest measurements of $s\text{PM}$ and $w\text{PM}$ was 53% and 43% for *B. acerifolius*; 51% and 50% for *P. undulatum*; 62% and 51% for *E. ovata*; 51 and 27% for *N. longifolia* (Figure 6b,d).

3.2. Amount of PM in Air and Weather Conditions during the Study Period

Monitoring of air pollution on location showed a highly variable concentration of PM₁₀ and PM_{2.5} during the study period (Figure 7).

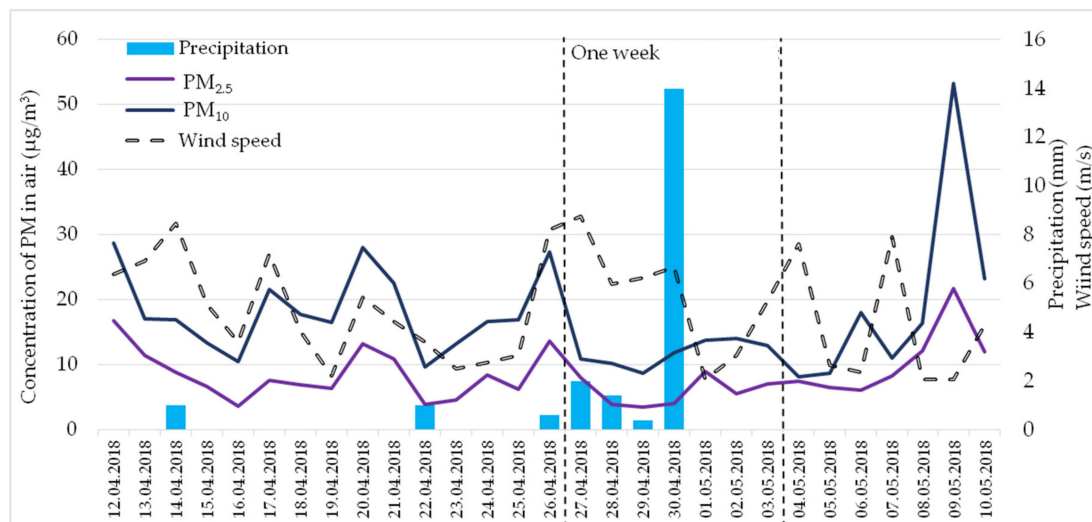


Figure 7. Meteorological conditions (rainfall and wind speed) and concentration of PM₁₀ and PM_{2.5} during the five-week period. Dashed lines show the seven-day period subject to daily analysis.

The air concentrations of PM₁₀ (from 8.2 $\mu\text{g m}^{-3}$ to 53.2 $\mu\text{g m}^{-3}$) were always higher than those of PM_{2.5} (3.5 $\mu\text{g m}^{-3}$ to 21.8 $\mu\text{g m}^{-3}$), but both were affected by the level of precipitation and, to some degree, by wind speed. During rainy periods, the concentration of both PM fractions tended to decrease (e.g., 21–22 April; 26–30 April) followed by their gradual increase. Wind strength was not consistently related to PM concentrations. During periods of low rain and wind the air level of PM₁₀ and PM_{2.5} increased (e.g., 17–20 April; 23rd–26 April; 5–9 May). The highest concentrations of PM₁₀ and PM_{2.5} were recorded on 9 May, after a week without rain and after a few days with low wind speeds (Figure 7).

3.3. Correlation Analysis

Air concentrations of PM₁₀ and PM_{2.5} were significantly positively correlated with all PM size fractions (10–100, 2.5–10 and 0.2–2.5) and categories (sPM and wPM), with only three exceptions recorded for fine PM (*B. acerifolius* for both PM₁₀ and PM_{2.5}, and *N. longifolia* for PM_{2.5}). With only one exception *P. undulatum* and PM₁₀, correlations obtained for wPM were higher than sPM (Table 1). Correlation analysis also revealed that the accumulation of large PM (10–100 μm), sPM and wPM was significantly negatively correlated with amounts of rainfall in all examined species, with coarse PM (2.5–10 μm) in *P. undulatum* and *N. longifolia* also significantly negatively correlated with rainfall amount, but there were no correlations with precipitation for fine PM (0.2–2.5 μm) (Table 1).

Wind speed was also significantly negatively correlated with accumulation of large PM in *P. undulatum*, *E. ovata* and *N. longifolia*, with coarse PM for *E. ovata* and *N. longifolia*, with fine PM (*P. undulatum*), sPM (*P. undulatum*, *E. ovata* and *N. longifolia*) and wPM in *E. ovata* and *N. longifolia*.

Table 1. Correlation coefficients between meteorological conditions (rainfall and wind speed) and concentration of PM₁₀ and PM_{2.5} in the air and the amount of PM (in three size fractions—fine, coarse and large—and the two types of PM—_sPM and _wPM) accumulating on leaves of four species of Australian trees. Results in bold are statistically significant.

Factor	Species	PM Fraction			Type of PM	
		PM 10–100 µm	PM 2.5–10 µm	PM 0.2–2.5 µm	_s PM	_w PM
Rainfall	<i>Brachychiton acerifolius</i>	−0.56	−0.37	−0.15	−0.49	−0.48
	<i>Pittosporum undulatum</i>	−0.58	−0.60	−0.03	−0.65	−0.54
	<i>Eucalyptus ovata</i>	−0.48	−0.40	−0.21	−0.46	−0.50
	<i>Notelaea longifolia</i>	−0.57	−0.62	−0.04	−0.58	−0.60
Wind speed	<i>Brachychiton acerifolius</i>	−0.33	−0.18	−0.16	−0.35	−0.31
	<i>Pittosporum undulatum</i>	−0.45	−0.06	−0.57	−0.45	−0.27
	<i>Eucalyptus ovata</i>	−0.63	−0.44	−0.39	−0.61	−0.59
	<i>Notelaea longifolia</i>	−0.59	−0.47	−0.14	−0.53	−0.52
PM ₁₀ in air	<i>Brachychiton acerifolius</i>	0.60	0.52	−0.25	0.63	0.65
	<i>Pittosporum undulatum</i>	0.54	0.48	0.67	0.57	0.54
	<i>Eucalyptus ovata</i>	0.45	0.54	0.57	0.47	0.57
	<i>Notelaea longifolia</i>	0.63	0.81	0.22	0.57	0.85
PM _{2.5} in air	<i>Brachychiton acerifolius</i>	0.57	0.46	−0.12	0.51	0.65
	<i>Pittosporum undulatum</i>	0.64	0.54	0.47	0.64	0.66
	<i>Eucalyptus ovata</i>	0.54	0.50	0.50	0.52	0.62
	<i>Notelaea longifolia</i>	0.65	0.76	0.25	0.59	0.84

4. Discussion

Native trees can perform a valuable service by reducing PM concentration in roadside air, but as our work shows, their interaction with PM is highly dynamic, altering in large amounts from day to day due to weather conditions such as rain and wind. Additionally, we confirm that not only does PM capture vary across species, but also the dynamics of deposition and wash off also vary and should be taken into consideration when choosing species for urban plantings. In this work, deposition of PM on leaves of four Australian native tree species was positively correlated with ambient air concentrations of PM_{2.5} and PM₁₀, proving that plants remove PM pollution from the air. Regardless of the species, PM accumulated on the foliage in three size fractions (0.2–2.5, 2.5–10, 10–100 µm) and two categories (water-washable (_sPM) and wax-embedded (_wPM)). Significantly, the highest accumulation of total PM (0.2–100 µm) and simultaneously the greatest wax layer (_wPM) were recorded for *E. ovata*, while the lowest, by one-fifth of total PM and an eighth of wax was *B. acerifolius*. These findings are in line with numerous other studies that demonstrate high positive correlation between PM accumulation and amount of wax on foliage [15,25]. However, in another Australian study, Leonard et al. (2016) [24], found that a *Eucalyptus* species (unidentified) was in the bottom two of 16 tree and shrub species in terms of total amount of PM accumulated on leaves. The high PM accumulation by *E. ovata* might be explained by the presence of oil glands on the leaf surfaces, supporting the hypothesis that leaf shape and morphological structures i.e., greater amounts of pubescence and rougher surfaces increase PM deposition on the foliage [16,20,23,25,46]. However, there is clearly much more work to do, and trait combinations may be more important than single characteristics such as oil glands or leaf hairs [24]. In contrast, Weerakkody et al. (2018) [47], found no correlation between PM accumulation and leaf surface characteristics. They concluded that PM load on foliage depends largely on individual leaf size irrespective of micro-morphology, but this may have been due to the particular leaf surface features of the species they studied. Many studies have shown that PM entrapment is greater in species with more leaf waxes, but in the current study *P. undulatum* and *N. longifolia* accumulated only slightly less PM than *E. ovata*, despite a much lighter wax layer. This contrasting pattern suggests that not only total wax content, but also its composition or structure, may be important. In the case of *P. undulatum*,

high PM deposition may result from its leaf structure (wrinkled and folded inwards, resembling a small basin and therefore acting as a natural trap for PM). Similar leaf characteristics were previously noted in *Syringa meyeri* 'Palibin' C.K. Schneid., which also has curled edges and accumulates high quantities of PM [26]. *B. acerifolius* accumulated the least PM and also had least waxes. However, this species should be considered for air phytoremediation, because unlike most other trees grown in Australia, it sheds leaves in autumn. These leaves can be collected, or decay on the ground and PM can be permanently removed from the atmosphere.

Foliar PM accumulation did not appear to increase uniformly between rain or wind events. One reason for this may be that after a certain number of days or weeks without precipitation, leaves reach their maximum PM loading capacities [19]. The amount of PM on leaves at a given time is therefore the net result of its accumulation from the atmosphere, less runoff and/or resuspension [27], and is also a function of recent rain intensity and duration, wind speed and direction, leaf characteristics and PM concentration in the air [29–32,34]. In this study, the amount of PM on foliage of *B. acerifolius*, *P. undulatum*, *E. ovata* and *N. Longifolia* was analyzed over a long time period (when measurements were made weekly) and a short time period (when measurements were made daily). PM load on leaves of all tested species changed very dynamically, regardless of the frequency of measurements. The relatively big daily differences in PM deposition on foliage (33–35%) were particularly interesting and to some extent surprising. This is a new finding and demonstrates that the assessment of species' potential for PM accumulation based on a single measurement (usually at the end of growing season) could be misleading. This is even more likely when plants are growing in different locations and are affected by different microclimatic conditions. Correlation analysis proved that the amount of PM on plant foliage is affected by rain and wind, with rain being more significant. Heavy rain and strong wind are likely to remove PM from foliage, while lighter rain and weaker wind may result in an increase PM load [32–34].

Rain and wind did not affect PM size fractions and categories in the same way. Large particles were the easiest to wash off and be resuspended from the leaf surface, while the fine PM fraction was almost unaffected by weather conditions. Przybysz et al. (2014) [29], also showed that the largest PM fraction is most easily removed by simulated precipitation and that the fine PM fraction seemed to adhere most strongly to the foliage surface. A more permanent accumulation of PM with a diameter of 0.2–2.5 μm is beneficial, because this PM fraction causes the greatest threat to human health [10–12]. Surprisingly, in all four species examined, precipitation had a negative effect on the retention of $w\text{PM}$ as well as $s\text{PM}$. Previously, it was believed that unlike $s\text{PM}$, $w\text{PM}$ was strongly immobilized in the waxes [15,18,23] and its resuspension was only possible after the desquamation (shedding) of waxes over the lifetime of a leaf, due to leaf aging, adverse weather conditions and exposure to contaminants [48,49]. This conclusion can be explained by the fact that in laboratory experiments (simulated rainfall), distilled water is used, which has a different composition and pH than natural rain. Additionally, in natural conditions, plants are affected simultaneously by several weather factors (e.g., precipitation, wind, changing temperature) during a rain event. Therefore, in our opinion, experiments with simulated rain do not accurately demonstrate the dynamics of PM accumulation, and wash-off results are therefore underestimates. Our observations to some extent confirm estimations of Schaubroeck et al. (2014) [35], who calculated that only 0.11% of annual dry deposition is deposited on plant foliage at the end of the growing season. Future experiments should take into the account the potential for significant inter-species variations in PM wash-off/resuspension [19], due to different micromorphologies of leaves. In addition, models should allow for water-insoluble particles that are dissolved in rainwater and removed early [50].

The effect of wind speed on PM retention by leaves was usually significantly negative, but in contrast to precipitation, plants differed in their responses to wind. The greatest impact of wind was recorded in *E. ovata* and *N. longifolia* (negative correlation with all PM fractions and categories, except the fine PM fraction), while *P. undulatum* was least affected and in *B. acerifolius*, there was no significant correlation. As all individuals of the examined species were growing in close proximity, the effect of

wind was most probably determined by a combination of plant morphological characteristics and the composition and structure of wax layers. Wang et al. (2015) [33] showed that the resuspension of PM from *Ligustrum lucidum* by a wind of 11 m s^{-1} was small compared with total deposition and they predict that only wind stronger than this can have a significant impact on PM deposition. Our results suggest that effect of wind on PM resuspension cannot be averaged for all species. A lower effect of wind than rain on $w\text{PM}$ is another interesting result of our work. Until now, it was believed that $w\text{PM}$ were protected against precipitation, but could be resuspended by strong wind. This may be an interpretation bias, resulting from using distilled water in experiments with simulated rain. A factor that should be accounted for in future experiments, is wind direction. According to Sgrigna et al. (2015) [51], it was not the wind speed, but its direction relative to main roads, that had most impact on the increase in PM load on studied plants.

The dynamics of accumulation, wash-off and resuspension of the accumulated PM are critical to the effect of plants on air quality. Wash-off and resuspension should be considered as a process akin to cleaning the filter and preparing the leaves for more deposition. These processes should be considered as beneficial in the planning of air phytoremediation in an urbanized area. This is especially critical for species that do not shed leaves during the year, such as those in this study: *P. undulatum*, *E. ovata* and *N. longifolia*. Additionally, as recorded in this work, accumulation of PM on foliage after rain is usually very high (e.g., high PM accumulation on 1 May after the rain event on 30 May). It can be explained by increased leaf 'stickiness' after precipitation. Rain also reduces the toxic effect on trees of contaminants accumulated on the foliage. Bell et al. (2011) [2], Gratani et al. (2000) [52], and Hanslin et al. (2017) [53] have recognized species-specific responses, mostly including growth inhibition, negative changes in gas exchange and leaves performance in urban plants exposed to air pollution. Moreover, when evergreen species retain foliage for more than a year, they may become so efficient in the retention of pollutants that they die because of an excessively heavy load of contaminants [54]. After rainfall, most of the runoff will be deposited on the ground. If the surface is open soil, or covered with vegetation, then the PM in the runoff will probably be immobilized there, whereas the runoff from paved areas is more likely to be resuspended by wind. However, the runoff of pollutants from leaf surfaces may cause environmental problems, such as leaching of bioavailable contaminants into soil and underground water, disturbing soil microflora, limiting plant growth and potentially posing a risk to human health [55].

5. Conclusions

Our study showed that the process of PM accumulation by vegetation is very dynamic, and differences in the PM load on the foliage can be large, even after one day. PM deposition on leaves was primarily affected by rain and to a lesser extent by wind, the latter was also more species-specific. The main fraction removed from leaf surfaces was the large PM fraction ($10\text{--}100 \mu\text{m}$), while the smallest PM fraction ($0.2\text{--}2.5 \mu\text{m}$) was retained more permanently. Surprisingly, precipitation also affected PM retention in waxes ($w\text{PM}$), which until now was believed to be not affected by rain. In our opinion, PM wash off and resuspension should be recognized as positive process, essential to recover the ability of plants to accumulate PM and to reduce phytotoxic effects of PM accumulated on leaf surfaces. This study also adds to the inventory of PM accumulation data for Australian tree species which might be considered for phytoremediation. The findings enhance the understanding of the complex interaction between plants, pollution and the atmosphere by adding a fourth variable, the weather, which, as demonstrated, has a strong influence on PM accumulation.

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