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Evaluating the impact of individual leaf traits on atmospheric particulate matter accumulation using natural and synthetic leaves.

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Highlights

- Individual leaf size, shape and micromorphology had a significant impact on capturing and retaining PM₁, PM_{2.5} and PM₁₀ while other influential variables were standardised.
- Smaller leaves and complex leaf shapes showed a greater potential to capture and retain particles.
- Leaf hair/trichomes, epicuticular wax, and surface-ridges are favourable to capture PM and presence of leaf hairs/trichomes found to be most influential.

Abstract

The ability of vegetation to capture and retain atmospheric Particulate Matter (PM) is directly dependent on the interactions between PM and plant surfaces. However, the impact of individual leaf traits in this respect is still under debate due to variations in published findings. This study employed standardised experimental designs with natural and synthetic leaves in three experiments to explore the impact of individual leaf traits on traffic-generated PM accumulation whilst other influential variables were controlled. The impact of leaf size on PM deposition was explored using synthetic leaves of different sizes (small, medium and large) but with the same shape and surface characteristics ($n = 20$ for each category). The impact of leaf shape was examined using another set of synthetic leaves of different shape (elliptical, palmately-lobed and linear) but with same surface area and the same surface characteristics ($n = 20$ for each category). PM accumulation (PM_1 , $PM_{2.5}$ and PM_{10}) on these leaves was quantified using an Environmental Scanning Electron Microscope (ESEM) and ImageJ software. Any differences in PM capture levels due to leaf size and leaf shape were identified using one-way Anova and Tukey's pairwise comparison. In a subsequent experiment, equal-sized, square-shaped leaf sections obtained from four plant species ($n = 20$ for each species) with different micromorphology were exposed to traffic-generated pollution and any PM capture differences due to leaf micromorphology identified employing the same SEM/ImageJ and statistical approach. The results of all three experiments showed significant differences in PM accumulation between different leaf sizes ($p < 0.001$), between different leaf shapes ($p < 0.001$) and between different leaf micromorphology ($p < 0.001$) suggesting that all these characters are influential in the capture and retention of PM on leaves. Smaller leaves and complex leaf shapes (lobed leaves) showed a greater potential to capture and retain PM. Leaf surfaces with hair/trichomes, epicuticular wax, and surface-ridges accumulated more PM compared to smooth surfaces; of these characters, leaf hairiness/ presence of trichomes was found to be the most important. Species sharing most of these important leaf traits are recommended as effective PM filters.

Key words: Traffic-generated pollution; Living walls; Green walls; Green infrastructure; Leaf shape; Leaf size; Micromorphology

1. Introduction

Three different size fractions of Particulate Matter (PM) are of particular concern based upon their ability to be inhaled and toxicity: coarse particles/ PM_{10} (aerodynamic diameter $\leq 10 \mu m$), fine particles/ $PM_{2.5}$ (aerodynamic diameter $\leq 2.5 \mu m$) and ultra-fine particles/ $PM_{0.1}$ (aerodynamic diameter $\leq 0.1 \mu m$) (Chow *et al.*, 2006; Solomon *et al.*, 2012). Long-term exposure to coarse particles diminishes lung function and increases cardiovascular mortality (Gilmour *et al.*, 1996). $PM_{2.5}$ can reach the narrower spaces in lungs (Brunkeef & Holgate, 2002) and cause lung cancer and cardio-vascular mortality associated with acute ischemic events (Solomon *et al.*, 2012). Ultra-fine particles are more dangerous than the other size ranges as they can cross cell membranes and influence intracellular functions (Riddle, 2009). According to Seaton *et al.* (1995), $PM_{0.1}$ can cause systemic inflammatory changes by entering the blood stream

or influence phagocytosis by accumulating in alveolar macrophages. They can also enter the brain via the olfactory nerves (Solomon *et al.*, 2012) which may cause central nervous system disorders (e.g. Alzheimer's disease and Parkinson's disease) depending on their chemical composition and toxicity (Allsop *et al.*, 2008; Maher *et al.* 2013).

Vegetation has been known as a sink for atmospheric PM for some time (Smith, 1975; Zulfacar, 1979) and the PM filtering behaviour of different types of vegetation has been studied using a range of different techniques (Beckett *et al.*, 2000; Dover, 2015; Freer-Smith *et al.*, 2004; Leonard *et al.*, 2016; Maher *et al.*, 2013; McDonald *et al.*, 2007; Ottel  *et al.*, 2010; Sternberg *et al.*, 2011; Terzaghi *et al.*, 2013; Zhang *et al.*, 2017). Vegetation has been found to be more effective in removing PM from air compared to other building/land surfaces due to high air turbulence created by their complex morphology and large surface area (Roupsard *et al.*, 2013; Tallis *et al.*, 2011).

Dry deposition of PM on vegetation takes place via sedimentation under gravity, impaction (via turbulent transfer), interception, and diffusion (Slinn, 1982; Wang *et al.*, 2006); processes antagonistic to deposition include aerodynamic resistance (resistance exerted on particles by the air), boundary layer resistance (reduced ability to cross the laminar air layer immediately adjacent to the deposition surface) and surface resistance (due to the properties of the deposition surface) (Davidson & Wu, 1990). The aerodynamic behaviour of different particle size fractions has been studied comprehensively under different meteorological conditions (Legg and Powel, 1979; Slinn, 1982; Petroff *et al.*, 2008b). Wind speed, wind turbulence, humidity and rainfall all had a considerable influence on PM deposition on vegetation (Litschke & Kuttler, 2008; Tomasevi  *et al.*, 2005). PM deposition is also driven by the interactions between the particles and plant surfaces including the latter's geometrical properties such as shape, size, orientation and surface morphology (Chen *et al.*, 2016; Freer-Smith *et al.*, 2005; Leonard *et al.*, 2016; Litschke & Kuttler, 2008; Petroff *et al.*, 2008a; Tomasevi  *et al.* 2005). Understanding the impact of such leaf traits on particulate deposition is thus crucial in the design and use of vegetation as an environmental control filter of particulate pollution. Legg and Powell (1979) modelled coarse particle impaction and sedimentation using fungal spores, and found that collection efficiency depended on the nature of the deposition surface. When the inertia of particles is too high to follow the wind flow deviations in the mean air flow around an object, they collide with it and deposit via impaction which can be, again, influenced by surface characteristics (Petroff *et al.*, 2008a). When particles with smaller inertia, which thus follow the wind flow deviations in the mean airflow, pass over plant surfaces with less than half a diameter distance between the centre of the particle and the plant surface they can deposit via interception; this process is also influenced by the micro-topography of the plant (Slinn, 1982). Despite several studies that have examined these interactions, there remains much debate on the relative importance of different leaf characteristics on PM capture.

For example, coniferous species with leaf needles have been frequently cited as being good PM filters compared to broad-leaved species (Beckett *et al.*, 2000; Dzierzanowski *et al.*, 2011; Wang *et al.* 2011), and Shackleton *et al.* (2010) found "grass-like" (linear leaved) species to be the best PM filters out of the 16 species they tested. In contrast, Leonard *et al.* (2016) found significantly higher PM levels on lanceolate leaves compared to needle-like or linear leaves. The effects of epicuticular wax and leaf

hairs have also produced mixed results. According to Dzierzanowski *et al.* (2011), the relationship between PM deposition and epicuticular wax does not depend on the amount of wax but on the structure and composition of the wax. Liu *et al.* (2012) found a negative impact of epicuticular wax in capturing particles but identified stomata, deep grooves, and leaf size as critically important characters. In contrast, Sæbø *et al.* (2012) found that PM deposition was a function of epicuticular wax content. Hairy leaves were found to be effective in accumulating PM in several different studies (Beckett *et al.*, 2000; Kardel *et al.*, 2012; Ram *et al.*, 2012); conversely, Perini *et al.* (2017) found a negative impact of leaf hair on PM capture.

These discrepancies in findings could be attributed to various interactive effects of different leaf traits on PM capture. Standardising the influence of non-target leaf variables should facilitate the investigation of the impact of each leaf character on PM accumulation. Assuming that positive characteristics are at least additive, and at best synergistic, in value, species carrying a collection of such leaf traits should result in high PM capture efficiency and hence be most appropriate to employ as PM filters. As these characters are inherent and not manipulatable in natural leaves, use of leaf models or synthetic leaves can help in standardising the influence of non-target variables. We therefore explored the individual impact of leaf size, shape and micromorphology on PM accumulation on leaves using manipulative experimental designs with natural and synthetic leaves whilst controlling for the influence of additional variables. We believe this is the first attempt to use such standardised designs in the evaluation of the impact of individual leaf characters on PM accumulation. This study is a contribution to the optimisation of living walls (vertical, irrigated, greenery systems typically carrying multiple non-climbing or twining plant species) as urban PM filters (Weerakkody *et al.* 2017) and hence the experimental designs employed relate to the configuration of vertical greenery systems. Since traffic-generated pollution has become the major source of PM in the UK (DEFRA, 2015) and categorised as the most toxic class of PM globally (WHO, 2005), PM generated through road traffic was focused on in this study.

2. Materials and Method

2.1 Site description

Stoke-on-Trent is a city located in Staffordshire, United Kingdom with an estimated population of 259,140 and population density of 6,640 persons/km² (ukpopulation, 2017). Leek Road is one of the busiest single carriageways in Stoke-on-Trent, categorised as an A Road (Fig. 1), with a traffic density of 20,251 Average Daily Flow in 2016 (Department for Transport, 2017). Given the continuous pollution generation due to road traffic, a linear, grassed area, 11.5 m in width at Staffordshire University, located parallel to Leek Road (4.5 m distance from the road) (Fig. 1) was selected to erect experimental rigs.

2.2 Manufacturing the synthetic leaves and sampling natural leaves.

Synthetic leaves used in these experiments were hand-made, using stiffened Poplin. A 1.0 m x 1.4 m section of commercially available Poplin (125.0 gm^{-2}), was stiffened by painting a sago solution (15 g of sago boiled in a 1 L of water) on the fabric to ensure synthetic leaves had no pleats or folds. Cardboard templates of different sizes and shapes (sizes and shapes are detailed in sections 2.4 and 2.5) were used to outline and cut the leaves from the fabric as required. Commercially available floral stems (plastic-paper covered stem wire, Handicrafts Ltd.) were stuck on one side of each of the leaves using fabric glue (Hobby glue gun- 230 V, 15 W, Powerbox International Ltd.) dried for 20 minutes. Using the same fabric, without any pleats or folds, produced artificial leaves with exactly the same surface characteristics and roughness (Fig. 2). Natural leaves required for the experiments were obtained from a free-standing living wall system (an experimentally manipulated system designed for our work on PM pollution, manufactured and installed by Nemec Cascade Garden Ltd., Czech Republic) (Fig. 1) located at the same site, facing Leek Road. Sampling and experiments were conducted on several occasions during March and April 2017. The mean temperature, mean humidity and mean wind speed of the study site was recorded as 12.8°C , 68% and 2.3 m s^{-1} during this period.

2.3 Scanning Electron Microscope (ESEM)/ImageJ approach to quantifying PM densities on natural and artificial leaves

PM accumulation on both natural and synthetic leaves used in this study were quantified using an Environmental Scanning Electron Microscope (ESEM) (Model: JSM-6610LV) (Ottele *et al.*, 2010; Sternberg *et al.*, 2010; Weerakkody *et al.*, 2017) and ImageJ image analysis software (Collins, 2007). Sampling numbers and methods of each experiment are detailed in the relevant sections below. In experiments where complete leaves were exposed to pollution all the leaves were synthetic (sections 2.4 and 2.5) and three leaf sections ($5 \text{ mm} \times 5 \text{ mm}$) from every leaf blade were cropped out and mounted on aluminium stubs using double-sided carbon adhesive tabs for microscopic analysis. In the experiment in which only leaf sections were exposed to pollution (all from natural leaves) (section 2.6), whole leaf sections were mounted without cropping. Both natural and synthetic leaves were scanned under a low vacuum in the ESEM at $\times 450$ and $\times 1,000$ magnifications using Back Scattered Electrons, without any conductive coating following the same approach used in Weerakkody *et al.* (2017) to visualise natural leaves. The high carbon content of the fibres used in the synthetic leaves minimises conductive charging, and PM accumulation was also clearly imaged using this approach. Micrographs were taken at three random points on each leaf section (natural or synthetic), maintaining the same working distance and consistent contrast/brightness to help define the threshold in image processing. The smallest particle that could clearly be visualised using this approach was $0.1 \mu\text{m}$ in diameter. Considering their different health effects and different aerodynamics, the number of PM_1 ($\text{PM}_{0.100} - \text{PM}_{1.000}$) $\text{PM}_{2.5}$ ($\text{PM}_{1.001} - \text{PM}_{2.500}$) and PM_{10} ($\text{PM}_{2.501} - \text{PM}_{10.000}$) on all the micrographs were quantified using ImageJ. The most appropriate threshold available in the auto-threshold menu was carefully selected using ten random micrographs to minimise potential human error of using a user-defined threshold. PM accumulation on each leaf section was estimated as PM density (number of PM per 1 mm^2) using the mean PM count on three random micrographs taken for each leaf section. In the

experiments where whole leaves were used, mean PM density on each leaf was estimated taking the mean PM density of three leaf sections (see further, section 2.4 and 2.5).

2.4 The impact of leaf size on PM capture

Synthetic leaves were manufactured for the following three size ranges: small (1.7 cm^2) medium (28.9 cm^2) and large (59.6 cm^2) with 40 leaves in each range (Fig. 3a). The leaves were of the same basic elliptical shape and the same surface characteristics (i.e. same fabric). The size of natural leaves in the Nemec living wall outside the Science Centre were used as an approximate guide for the size ranges. Prior to use, the synthetic leaves were washed using a spray bottle and then dipped and shaken in deionised water in large beakers to remove existing particles. Subsequently, they were dried in a closed drying chamber (with controlled light and temperature regimes), thereby reducing contamination by indoor particulates. Twenty leaves from each size category were then scanned, to visualise any PM remaining on their surfaces, using the SEM/imageJ approach (see section 2.2); any significant variation in baseline PM levels between size ranges were identified using a one-way Anova (R statistical software version 3.2.5: R Core Team, 2016). The remaining 20 leaves of each size category were then exposed to traffic pollution generated from Leek Road by mounting them on a wooden garden trellis. Leaves were attached to the trellis by their petioles using thin wires at 1.0 m -1.5 m height from the Road surface, keeping a similar configuration as vertical greenery systems (i.e. facing the road). Leaf arrangement was random using a Latin Square design to avoid any variation relating to columns and rows. The trellis was erected at the roadside edge of the grassed area, 4.5 m distance from the roadside and leaves were left exposed to traffic pollution for five consecutive dry days. Subsequently, leaves were taken to the laboratory using sealed storage boxes to provide minimal disturbance and PM densities were quantified using the ESEM/imageJ. Identification of baseline PM levels prior to exposure was carried out to identify if there were significant differences in the different leaf categories, and hence allow for adjustment of the experimental data by subtracting the mean baseline levels from the PM densities found on the roadside-exposed leaves (i.e. PM density on exposed leaves – mean baseline PM density) if required. Differences in PM levels captured by different sizes of leaves were identified using a one-way Anova followed by Tukey's pairwise comparison ($n = 20$). The leaf perimeter/leaf surface area ratio in each category was calculated to explain any differences in PM accumulation due to variable edge effects; the leaf perimeter was measured using ImageJ image analysis software.

2.5 The impact of leaf shape on PM capture

Synthetic leaves with exactly the same surface area (28.9 cm^2) and surface characteristics were made in three common leaf-shapes (elliptical, palmately-lobed and linear) (Fig. 3b); 40 leaves were made for each shape category. Leaves were washed using the same approach followed in section 2.4 to remove existing particles and dried in a closed drying chamber avoiding contamination from indoor particulates. Twenty leaves from each shape category were then tested for their baseline PM levels using SEM/imageJ analysis. The remaining twenty leaves of each shape category were simultaneously exposed to traffic pollution on Leek Road for five consecutive days using a wooden garden trellis following the same approach given in section 2.4. Subsequent transfer of leaves to the lab, visualisation

and counting of particulates, and statistical analysis followed the approach in Section 2.4 ($n = 20$). The Leaf perimeter/ surface area ratio in each category was calculated to explain any differences in PM accumulation due to variable edge effects.

2.6 The impact of leaf micro-morphology on PM capture

Sixteen square holes (1 cm x 1 cm) were cropped-out from plastic laminating pouches (ImageLast 125 Micron, Lyreco) in 4 equally spaced parallel rows. Four species of plant with different surface textures (e.g. hairy, smooth, rough and velvety) (*Geranium macrorrhizum* L., *Bergenia cordifolia* (L.) Fritsch, *Helleborus x sternii* and *Heuchera villosa* Michx. var. *macrorrhiza*) used in the Nemec living wall (Fig. 1) were selected for this experiment. The leaf shape and size was standardised in this experiment so that differences in PM capture due to surface characteristics (micromorphology) could be identified. Forty leaves per species were randomly collected from the living wall and carefully washed by dipping and slowly shaking in cold deionised water to remove existing PM. Leaves were dried at 22 °C in a closed drying chamber for one hour. Subsequently, 1.5 cm x 1.5 cm sections were excised from every leaf blade and twenty leaf sections of each species were immediately tested for their baseline PM levels. Remaining leaf sections were attached to the back of the laminating paper replacing the empty squares in a randomised order (Latin Square design) using thin sticky tapes; this resulted in a grid of equal sized leaf squares with different micro-morphology (Fig. 4). Five such rigs, each holding 16 leaf sections (4 leaf sections from each species) were made to hold a total of 80 leaf sections (20 leaf sections per species). The back of the laminating papers were then covered with wet layers of gauze to prevent dehydration of leaves, and inserted into plastic frames with water retentive backing. The plastic frames holding the leaf sections, with supportive material, were then exposed to traffic pollution generated from Leek Road (4.5 m distance from the roadside) for five consecutive days. Gauze layers were kept moist by spraying deionised filtered water through holes in the back of the plastic frames each day to avoid any structural changes in leaf sections due to dehydration. These experimental rigs were taken to the laboratory with minimal disturbance using the same approach followed in section 2.4 and 2.5. Leaf sections were carefully removed from the holding frames and PM densities on leaf sections were quantified using SEM/ImageJ analysis. Statistical analysis followed the approach in Section 2.4 ($n = 20$). Specific leaf micromorphological characters (hairs, trichomes, epicuticular wax, ridges) of the leaf sections were observed using the ESEM at a range of magnifications as appropriate.

3. Results

3.1 Impact of leaf size on PM capture using synthetic leaves of the same shape

Baseline PM levels on synthetic leaves were not significantly different between different size categories for all particle size fractions ($p > 0.05$) hence they were not deducted from the PM levels on synthetic leaves exposed to roadside air pollution. Roadside-exposed leaves showed differential PM densities on leaves of different sizes (Fig. 5) (PM_1 : $F = 114.9$, $p < 0.001$, $PM_{2.5}$: $F = 41.68$, $p < 0.001$, PM_{10} : $F = 76.53$, $p < 0.001$) in all particle size fractions. The highest mean PM densities in all particle size fractions ($PM_1 = 2,233 \text{ mm}^{-2}$, $PM_{2.5} = 1,122 \text{ mm}^{-2}$ and $PM_{10} = 303 \text{ mm}^{-2}$) were found on the smallest leaves and these levels were significantly higher than the PM levels in both medium and large-sized leaves ($p < 0.001$).

The second highest mean PM densities in all PM size fractions were found on medium-sized leaves; the density of PM₁₀ on medium sized leaves was significantly higher compared to larger leaves ($p < 0.001$). However, densities of PM₁ and PM_{2.5} on medium sized leaves were not significantly different from larger leaves ($p = 0.06$ and $p = 0.22$ respectively). The leaf perimeter/surface area ratio was 0.2, 0.07 and 0.04 for small, medium and large sized leaves respectively (ratio between sizes, small: medium: large = 27 : 7 : 4).

3.2 Impact of leaf shape on PM capture using synthetic leaves of the same area

Baseline PM densities on leaves of different shape categories were not significantly different ($p > 0.05$) and hence were not deducted from the PM densities on synthetic leaves exposed to roadside air pollution. There were differential PM levels on roadside-exposed leaves with different shapes (Fig. 6) in all PM size fractions (PM₁: $F = 21.06$, $p < 0.001$; PM_{2.5}: $F = 31.09$, $p < 0.001$; PM₁₀: $F = 37.9$, $p < 0.001$). The highest PM densities in all particle size fractions (PM₁ = 2,226 mm⁻², PM_{2.5} = 652 mm⁻² and PM₁₀ = 211 mm⁻²) were found in palmately-lobed leaves and these densities were significantly higher than for both elliptical and linear leaves. The second highest PM densities were found on linear leaves in all particle size fractions and the density of PM₁₀ on linear leaves was significantly higher compared to PM density on elliptical leaves ($p = 0.01$). However, these levels were not significantly different for smaller PM sizes (PM₁: $p = 0.37$ and PM_{2.5}: $p = 0.88$). The leaf perimeter/surface area ratio was 0.07, 0.09 and 0.16 for elliptical, lobed and linear leaves respectively (ratio between shapes:- elliptical : lobed : linear = 7 : 9 : 16).

3.3 Impact of micromorphology on PM capture by the adaxial surface of natural leaves with leaf area and shape held constant

Leaf micrographs of four species of plants (Fig. 8) showed their different surface micromorphologies and are detailed in Table 1. Baseline PM densities on leaf sections of different species were not significantly different ($p > 0.05$) after washing-off and hence were not deducted from the PM densities of leaf sections subsequently exposed to roadside air pollution. There were differential PM levels on roadside-exposed leaf sections of different species (Fig. 7) in all PM size fractions (PM₁: $F = 10.38$, $p < 0.001$; PM_{2.5}: $F = 12.27$, $p < 0.001$; PM₁₀: $F = 140.9$, $p < 0.001$). Leaf sections of *G. macrorrhizum* showed the highest mean PM densities in all particle size fractions (PM₁ = 7,424 mm⁻², PM_{2.5} = 1,902 mm⁻² and PM₁₀ = 383 mm⁻²). PM₁ densities on *G. macrorrhizum*, *H. villosa* and *H. sternii* were significantly higher than on *B. cordifolia* ($p < 0.001$) but not significantly different ($p > 0.05$) from one another. The density of PM_{2.5} on *G. macrorrhizum* was significantly higher than *B. cordifolia* ($p < 0.001$) and *H. villosa* ($p = 0.03$) but not significantly different from *H. sternii* ($p = 0.13$). *B. cordifolia* showed the lowest mean PM density in all particle size fractions (PM₁ = 3,539 mm⁻², PM_{2.5} = 790 mm⁻² and PM₁₀ = 69 mm⁻²) which were significantly lower ($p < 0.05$) than the rest of the species. PM levels on *H. villosa* and *H. sternii* were not significantly different in the smaller PM size fractions ($p > 0.05$). PM₁₀ levels were more varied between species compared to other size fractions and the densities of PM₁₀ on leaf sections of each species were significantly different from each other.

4. Discussion

4.1 Impact of leaf size on PM accumulation on leaves

As synthetic leaves with the same leaf shape and same surface characteristics were arranged randomly, at the same height, and with the same pollution exposure levels and timings, the different PM capture levels that resulted (Fig. 5) can only be attributed to their size. In contrast to the findings of Sæbø *et al.* (2012), large differences in PM densities were found in all PM size fractions which demonstrates that size is an important trait to consider in assessing PM accumulation on leaves. Similar to the findings of Freer-Smith *et al.* (2005) and Leonard *et al.* (2016), and in contrast to Liu *et al.* (2012), high PM accumulation on smaller-sized leaves demonstrates their greater potential to capture and retain PM. A pilot study we carried out (unpublished data) on the distribution of particulates on leaf surfaces found elevated PM levels on leaf edges and tips except for linear-shaped leaves where there was no apparent change in density with distance from the leaf edge. The relatively larger edge effect present in smaller sized leaves (due to their high perimeter/surface area ratio) may have resulted in high levels of PM impaction on leaves. However, this pattern was only significant for PM₁₀ and not for the smaller particles size fractions (PM₁ and PM_{2.5}); this difference may result from the different aerodynamic behaviour of different particle sizes (Slinn, 1982). Increased turbulence in the boundary layer around a deposition surface increases PM accumulation via turbulent transfer which is more important for smaller particle size fractions (Petroff *et al.*, 2008a; Slinn, 1982). More turbulence around leaf edges can result from leaves by swaying with the wind flow. There is a substantial difference in perimeter/surface area ratios between small and medium-sized leaves but this is much smaller between medium and larger leaves (small : medium : large = 27 : 7 : 4). This might explain the large differences in PM levels between small and medium sized leaves and the smaller differences evident between medium and larger leaves.

4.2 Impact of leaf shape on PM accumulation on leaves

The shape of the deposition surface can directly influence the airflow pattern around the surface and hence, has a marked influence on PM deposition (Davidson & Wu, 1990; Petroff *et al.*, 2009), and this experiment with synthetic material showed that there were large differences in PM accumulation on leaves with different shapes. As additional factors (e.g. surface area, surface characteristics, exposure to weather and PM exposure levels) were held constant, the different levels of PM accumulation can only be attributed to the shape of the leaves. Although the different leaf shapes had different perimeter/surface area ratios (elliptical : lobed : linear = 7 : 9 : 16), they did not explain the differential PM accumulation by different shapes. Variable shapes generate different drag forces on them due to wind (Gemba, 2007), changing their influence on surrounding air flow patterns; the response of leaves to such forces can be by swaying, bending or fluttering (Gillies *et al.*, 2002) and hence different levels of turbulence can result. The pattern of PM accumulation between species was consistent for all PM size fractions (Fig. 6); palmately lobed leaves showed a greater potential to capture and retain PM, probably due to their complex shape. Lobed leaves create more than one "leaf-tip-like" area creating a more complex morphology; results of an unpublished pilot study (see section 4.1) showed elevated PM

accumulation at leaf tips. Similar to the results of Leonard *et al.* (2016), PM accumulation on our elliptical and linear leaves was relatively poor. Even though linear leaves can be predicted to be good PM filters on the basis of their larger perimeter, they can also bend more readily with the wind flow (as these lengthy leaves are connected to a petiole with a narrow leaf base) without swaying with the wind currents, potentially resulting in lower levels of turbulence. However, despite having a linear shape, leaf needles are frequently cited as good PM filters (Beckett *et al.*, 2000a; Freer-Smith *et al.*, 2005; Mori *et al.*, 2015; Räsänen *et al.*, 2013), in addition to their epicuticular wax helping to provide a sticky surface to retain PM, this may also be attributed to their rigid/stiff nature compared to “grass-like” linear leaves. Further research in this area is clearly warranted.

4.3 Impact of leaf micro-morphology on PM accumulation on leaves

Significant variations in PM levels on real leaf sections with different micromorphology but with area and shape held constant clearly demonstrated that leaf micromorphology has an impact on PM capture and retention and hence is an important leaf trait to consider where vegetation is being proposed for PM removal. Variation in PM₁₀ levels were particularly interesting, where differential PM capture abilities were evident between all the species examined. In addition to turbulent deposition, capture by sedimentation is more important for PM₁₀ compared to smaller particles (Petroff *et al.*, 2008a) and surface texture is known to be very important in preventing heavy PM rebounding from surfaces (Davidson & Wu, 1990). The greater potential of *G. macorrhizum* to capture and retain PM in all size fractions can be attributed to its complex micromorphology, with densely arranged trichomes and glandular hairs (Fig. 8) (Table 1). *H. villosa* is also slightly hairy and also showed relatively high PM accumulation supporting this argument. Leaf hairs are known to be important in increased PM accumulation (Beckett *et al.*, 2000; Leonard *et al.*, 2016; Räsänen *et al.*, 2013; Ram *et al.*, 2012; Sæbø *et al.*, 2012) by increasing surface area (for capture) and by preventing re-suspension of captured PM (Prusty *et al.*, 2005; Qiu *et al.*, 2009). Having such protruding structures can also create complex micro-topography on leaf surfaces which also facilitates capture and retention of PM compared to smooth leaves. In addition, the hydrophobicity of some leaf hairs is known to have a positive impact on attracting metal-based charged particles (Fernández *et al.*, 2014).

The poor ability of leaves of *B. cordifolia* to capture and retain PM can probably be attributed to their glossy smooth leaf surface. PM can readily rebound from smooth surfaces resulting in the retention of only a small fraction of captured particles (Davidson & Wu, 1990). *H. sternii* also showed relatively high PM levels compared to *B. cordifolia* which could probably be attributed to their deep surface ridges and epicuticular wax. While a positive impact of epicuticular wax on PM capture is frequently cited in the literature (Barima *et al.*, 2014; Räsänen *et al.*, 2013; Sæbø *et al.*, 2012), several studies found reduced PM levels on waxy surfaces due to their variable chemical structure and composition (Faini *et al.*, 1999; Kardel *et al.*, 2012; Leonard *et al.* 2016) and also due to the self-cleansing ability of wax tubules (Wang *et al.*, 2011). However, rough leaf surfaces with a large number of ridges were found to be effective in PM capture (Kardel *et al.*, 2012; Ram *et al.*, 2012; Zhang *et al.*, 2017) and Barima *et al.* (2014) found significantly higher PM levels on ridged leaf surfaces compared to waxy surfaces. The relatively high

levels of PM on *H. sternii* could either be attributed to their epicuticular wax or ridges or to the collective impact of both these characters. However, these levels did not significantly exceed the PM levels found on hairy leaves for any particle size fraction, highlighting the importance of leaf hairs in the capture and retention of PM. Nevertheless, the actual impact of these species of plants under field conditions, may be quite different when they stand as leaves instead of leaf sections. For example, irrespective of their ridged surface, Weerakkody *et al.* (2017) found very low PM accumulation on leaves and plants of *H. sternii* citing their wide leaves and low leaf area index as potential reasons. Therefore, the potential of leaf surfaces for the capture and retention of PM can be enhanced or limited by other influential variables such as leaf size, shape, location, configuration and leaf area index (Freer-Smith *et al.*, 2005; Leonard *et al.*, 2016; Weerakkody *et al.*, 2017). However, selecting species sharing at least a few of these micro-morphological characters (e.g. ridged hairy leaf surfaces) would probably be beneficial to increase PM accumulation.

4.4 Implications of the findings

This study showed that there was a considerable impact of all tested leaf characteristics on PM capture and retention when they act alone. The discrepancy we find in the literature on the impact of these characteristics are probably attributable to their collective impact which can enhance or limit the ability of particular plants to capture and retain PM. As the ability of variable leaf surfaces to capture and retain PM can be enhanced or limited by other influential variables it is important to recognise that those variations in PM capture did not represent their particular species, but the specific micro-morphological features. As smaller leaves, lobed shapes, hairy and rough leaf surfaces were found to have a positive impact on PM capture and retention, using a collection of species that share most of these characters (e.g. smaller leaves with a complex shape and a rough, hairy surface) combined with a high leaf area index (Weerakkody *et al.* 2017) would probably provide maximised benefits where vegetation is intended to be used as a PM filter.

Conclusion

Size, shape and micromorphology of individual leaves showed a significant impact on capturing and retaining all particle size fractions tested. Smaller leaves showed a greater capacity to capture and retain particles probably due their larger edge effect. Palmately-lobed leaves showed high PM levels compared to elliptical or linear leaves as they may create more turbulence in the boundary air layer with their complex shape and “tip-like” areas. Hairy leaves and leaves with rough ridged surfaces with epicuticular wax were good at capturing and retaining particles in all size fractions compared to leaves with a smooth surface. The real-world effectiveness of these traits to capture and retain PM can be enhanced or limited by other influential variables. A selection of species sharing these characters is likely to maximise the benefits of vegetation as PM filters.

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Fig. 1: Upper image: map showing the location of the experiment (circled) on Leek Road, Stoke-on-Trent, UK Contains OS data ©Crown copyright and database right (2017). Lower image: the grassed area adjacent to the Science Centre at Staffordshire University facing the Leek Road and the living wall system. The location and direction of the photo taken is marked by the arrow in the (above) map

Fig. 2: ESEM images of synthetic leaves in different sizes showing their same surface structure (x50)

Fig. 3: (a) an image of synthetic leaves designed with different leaf sizes but with the same shape and micromorphology, and (b) an image of synthetic leaves designed with different shapes but with the same surface area and same micromorphology.

Fig. 4: Schematic diagram of an experimental rig holding equally sized and shaped leaf sections with different micro-morphology arranged in a random order.

Fig 5: Mean \pm 1SE PM densities on leaves of different sizes (small: 1.7 cm², medium: 28.9 cm², and large: 59.6 cm²) but with the same shape and micromorphology (see further Fig. 3a). Species labeled with the same letter are not significantly different from each other within the given particle size fraction (PM₁: a,b; PM_{2.5}: c,d; PM₁₀: e,f,g).

Fig. 6: Mean \pm 1SE PM densities on leaves of different shapes but with the same surface area (28.9 cm²) and micromorphology (see further Fig. 3b). Species labeled with same letters are not significantly different from each other within the given particle size fraction (PM₁: a,b; PM_{2.5}: c,d; PM₁₀: e,f,g).

Fig. 7: Mean \pm 1SE PM densities on real leaf sections with different micromorphology but with same surface area and shape. Species labeled with the same letters are not significantly different from each other within the given particle size fraction (PM₁: a,b; PM_{2.5}: c,d,e; PM₁₀: f,g,h,i).

Fig. 8: Scanning Electron Microscope images (450x) of leaf micromorphology of the adaxial surface of a) *B. cordifolia* b) *H. sternii* c) *H. villosa* (*Heuchera macrorrhiza*) and d) *G. macrorrhizum*



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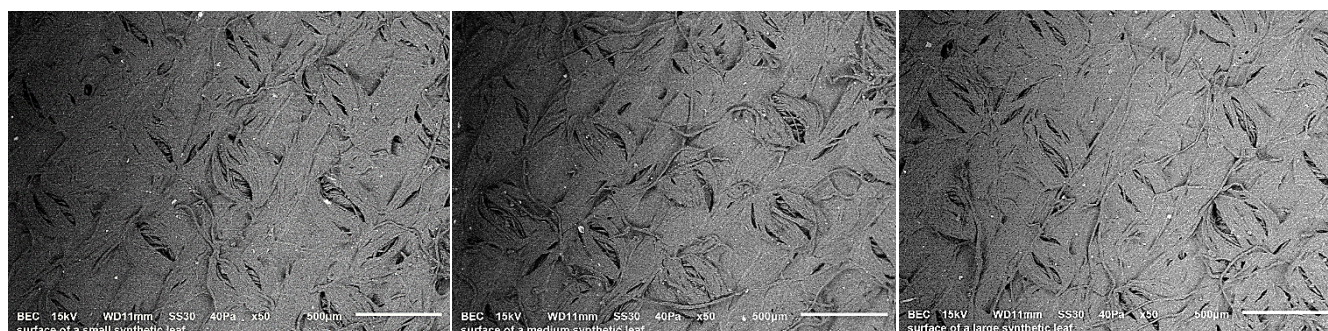


Fig. 2: ESEM images (x50) of synthetic leaves used in manipulative experiments where size or shape were varied whilst holding other variables constant (see further Fig. 3). In this series of images, the surface characteristics of small, medium, and large leaves (left to right) are visualized to demonstrate their similar surface characteristics.

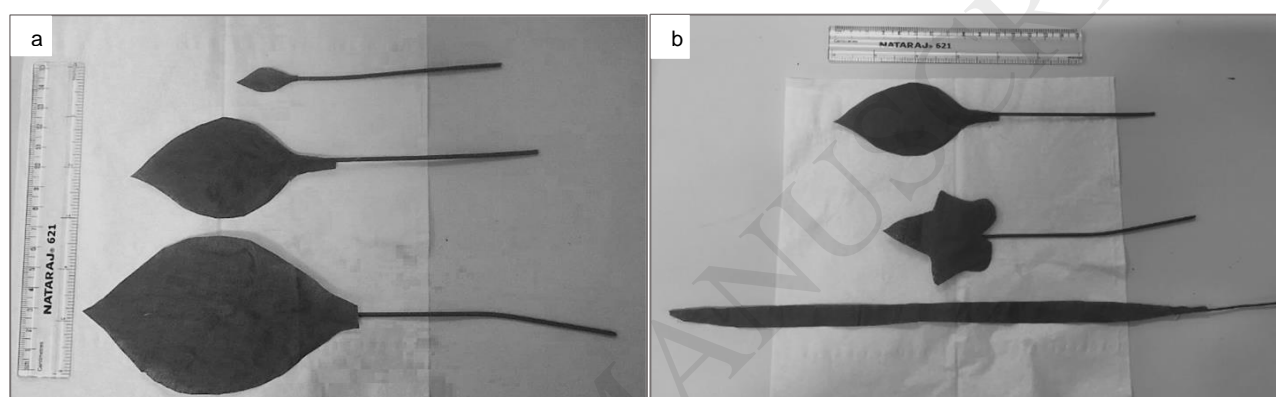


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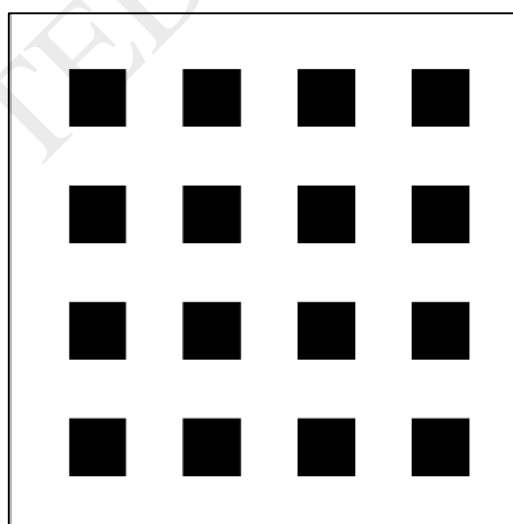


Fig. 4: Schematic diagram of an experimental rig holding equally sized and shaped leaf sections with different micro-morphology arranged in a random order.

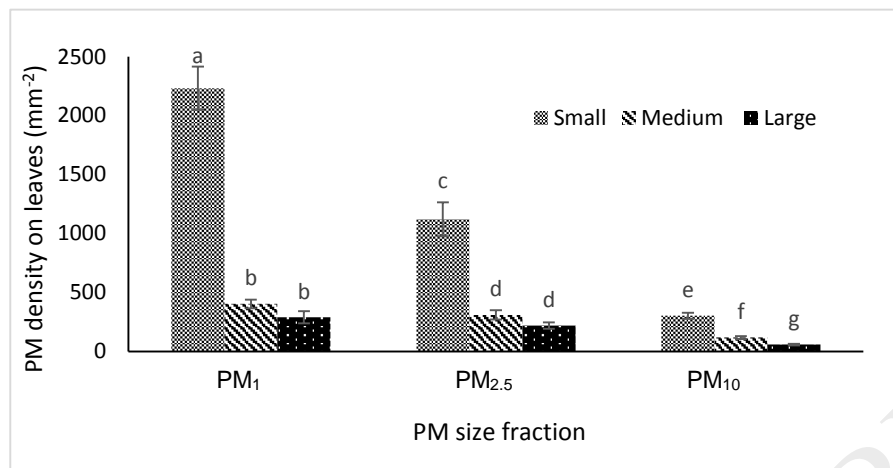


Fig 4. Mean ± 1 SE PM densities on leaves of different sizes (small: 1.7 cm², medium: 28.9 cm², and large: 59.6 cm²) but with the same shape and surface characteristics (see further Fig. 3a). Species labeled with the same letter are not significantly different from each other within the given particle size fraction (PM₁: a,b; PM_{2.5}: c,d; PM₁₀: e,f,g).

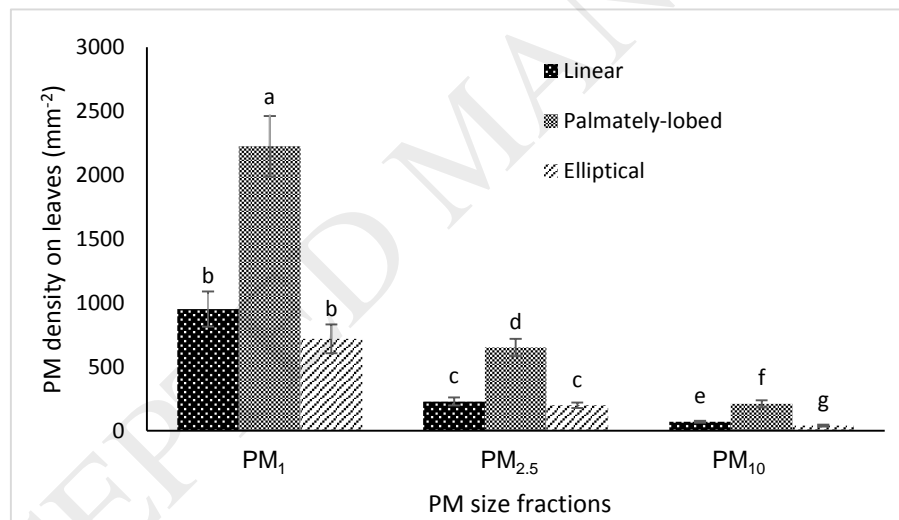


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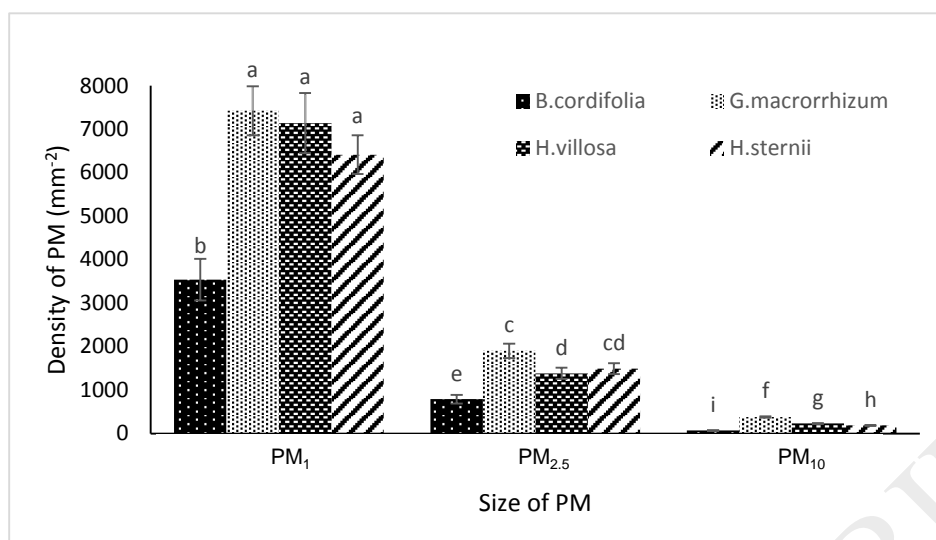


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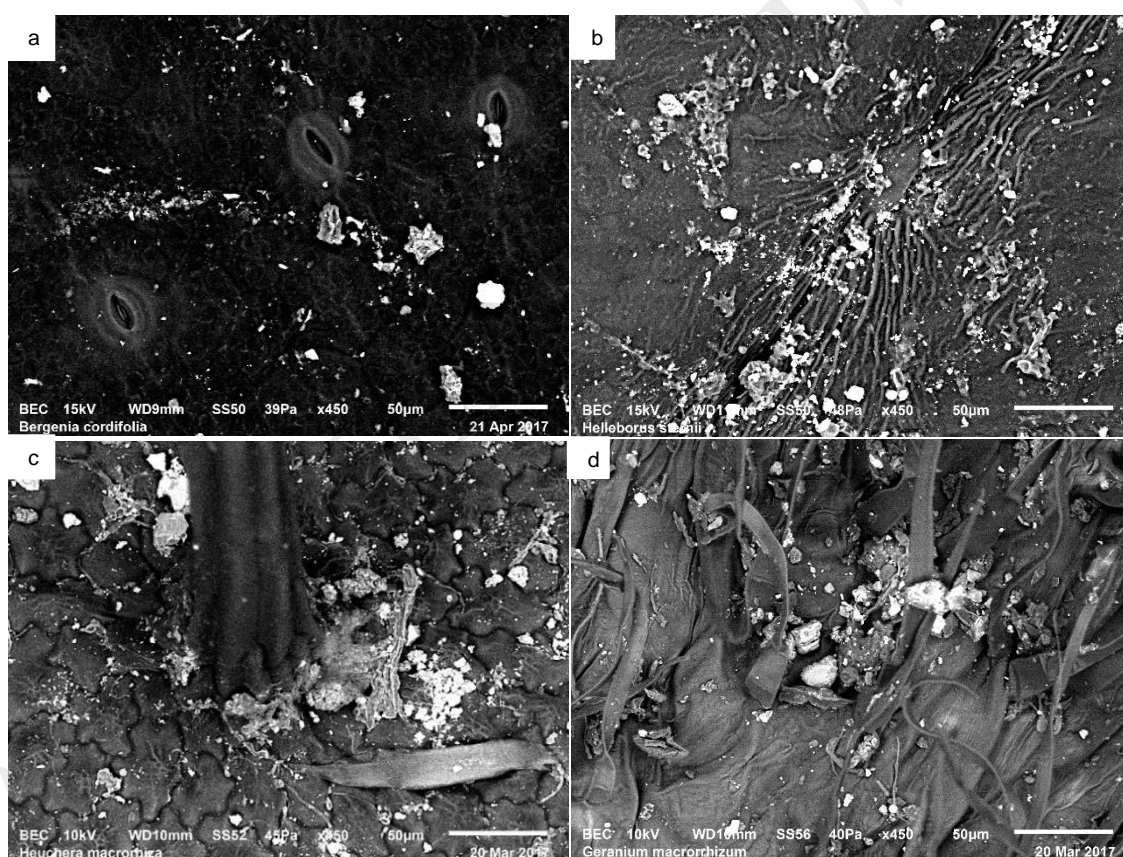


Fig. 8 Scanning Electron Microscope images (450x) of leaf micromorphology of the adaxial surface of a) *B. cordifolia* b) *H. sternii* c) *H. villosa* (*Heuchera macrorrhiza*) and d) *G. macrorrhizum*

Table 1: Micromorphological characteristics of different species of plants used in the study

| Species | Common name | Description of leaf micro-morphology |
|--|------------------------------------|--|
| <i>Bergenia cordifolia</i> (L.) Fritsch | Heart-leaf bergenia | Leaf surfaces were glossy and smooth. Sparsely arranged wax glands were present. |
| <i>Helleborus x sternii</i> Turrill | Hellebore, blackthorn strain | Leaf surfaces were leathery but rough due to densely arranged ridges and grooves. Epicuticular wax layers were slightly prominent. |
| <i>Heuchera villosa</i> Michx. var. <i>macrorhiza</i> (<i>Heuchera macrorhiza</i>) | Autumn Bride | Leaf surfaces were velvety and slightly hairy (49 hairs per 1 mm ²). Epicuticular wax was not prominent. |
| <i>Geranium macrorrhizum</i> L. | Geranium macrorrhizum | Leaf surfaces were covered with densely arranged hairs (135 hairs per 1 mm ²) and glandular trichomes. Epicuticular wax was localised and not prominent. |