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The influence of plant species, leaf morphology, height and season on PM capture efficiency in living wall systems

#### Reference:

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- 1 The influence of plant species, leaf morphology, height and season on PM
- 2 capture efficiency in living wall systems

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#### Abstract

Green infrastructure (GI) is already known to be a suitable way to enhance air quality in urban environments. Living wall systems (LWS) can be implemented in locations where other forms of GI, such as trees or hedges, are not suitable. However, much debate remains about the variables that influence their particulate matter (PM) accumulation efficiency. This study attempts to clarify which plant species are relatively the most efficient in capturing PM and which traits are decisive when it comes to the implementation of a LWS. We investigated 11 plant species commonly used on living walls, located close to train tracks and roads. PM accumulation on leaves was quantified by magnetic analysis (Saturation Isothermal Remanent Magnetization (SIRM)). Several leaf morphological variables that could potentially influence PM capture were assessed, as well as the Wall Leaf Area Index. A wide range in SIRM values (2.74 - 417 µA) was found between all species. Differences in SIRM could be attributed to one of the morphological parameters, namely SLA (specific leaf area). This suggest that by just assessing SLA, one can estimate the PM capture efficiency of a plant species, which is extremely interesting for urban greeners. Regarding temporal variation, some species accumulated PM over the growing season, while others actually decreased in PM levels. This decrease can be attributed to rapid leaf expansion and variations in meteorology. Correct assessment of leaf age is important here; we suggest individual labeling of leaves for further studies. Highest SIRM values were found close to ground level. This suggests that, when traffic is the main pollution source, it is most effective when LWS are applied at ground level. We conclude that LWS can act as local sinks for PM, provided that species are selected correctly and systems are applied according to the state of the art.

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### 1 Introduction

Particulate matter (PM) pollution is one of the major challenges in urban environments nowadays. Even though PM<sub>2.5</sub> concentrations in urban environments decreased by approximately 20% between 2005 and 2015, EU air quality standards have not been met (EEA, 2017a; EEA, 2017b). Epidemiological studies show a strong correlation between PM pollution and adverse health effects (Pope III and Dockery 2006; Solomon et al. 2012; Van Dyck et al. 2019). Besides severe health effects and premature mortality (Brunekreef 1997), PM pollution also causes damage to the environment, crops and infrastructure (Jimoda et al. 2012; Rai et al. 2016). Urban green infrastructure (UGI) such as trees, hedgerows, green roofs and green walls can act as a natural filter for PM and contribute to air pollution mitigation by deposition of particles on plant leaves (Beckett, Freer-Smith, and Taylor 1998; Litschke and Kuttler 2008; Nowak, Crane, and Stevens 2006; Pugh et al. 2012; Shao et al. 2021). Deposition effects related to vegetation were studied by Gallagher et al. (2015) and Janhäll (2015), among others. Moreover, Abhijith et al. (2017) discussed the comparison of different UGIs on air quality improvement. However, due to the street canyon effect, models suggest that trees can sometimes have a negative impact on local air quality (Vos et al. 2012) by hampering air flow. In contrast, green walls (vertical structures covered by vegetation) do not inhibit free air flow through the street canyon and thus can also have a positive impact on urban air quality, when applied appropriately. Moreover, green walls provide many more ecosystem services than PM mitigation, such as cooling the ambient air and underlying walls (Pérez et al. 2011; Wong et al. 2010; Koch et al. 2020), increasing biodiversity (Madre et al. 2015), water retention and purification (Prodanovic et al. 2017; van de Wouw et 45 al. 2017; Hussain Lakho et al. 2021), carbon sequestration (Tallis et al. 2015) and social and cultural benefits 46 (White and Gatersleben 2011). 47 Recently, some interesting studies have been published regarding green walls and their potential to 48 capture PM, on which a review was published by Ysebaert et al. (2021). Literature mainly considered soil-49 bound Ivy (Hedera helix) (Ottelé et al. 2010; Sternberg et al. 2010; Przybysz et al. 2014), although (Perini 50 et al. 2017) studied several other climber plant species and (Weerakkody et al. 2017, 2018a, 2018b and 51 Viecco et al. 2018) reported about living wall systems (LWS). LWS are systems in which plants that normally 52 grow in the soil are rooted in a substrate material, directly attached to the wall (Koch et al. 2020). The 53 substrate can be pre-cultivated panels, bags, textiles or other materials, and can be of natural composition 54 (soil, moss) or synthetic. There is always an irrigation system that also includes nutrient distribution over 55 the plants. This kind of system facilitates the use of a wide variety of plant species, from small herbs to 56 medium sized shrubs. 57 Plant species, all of which have their own unique morphological and physiological characteristics, are 58 typically characterized by different PM capture qualities according to this review paper. The importance 59 of selecting plants with certain traits is also highlighted by Litschke and Kuttler (2008), Weerakkody et al. 60 (2017), Muhammad et al. (2019) and others. It was found that plant leaves with a higher wax content 61 (Perini et al. 2017), higher wettability (Muhammad et al. 2019; Wang et al. 2013) or with a higher stomatal 62 density (Weerakkody et al. 2018b) collect more particles. The influence of trichomes is still considered 63 inconclusive for green wall studies. Hairy leaves were less effective in PM capture according to Perini et al. 64 (2017) while hairiness was considered insignificant according to Weerakkody et al. 2018b). This is in 65 contrast to other studies on more than hundred tree, shrub and climber species (Sæbø et al. 2012; Kardel

et al. 2012; Muhammad et al. 2019) and herbaceous plants (Weber et al; 2014), which report important

differences between hairy and non-hairy leaves and significant positive relationships between PM

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accumulation and trichome density. To summarize, we assume that the amount of LWS species considered is too small to make decent comparisons with the large amount of tree species considered in literature. Leaf shape was also considered an important variable, with small and needle-like leaves being better PM accumulators (Beckett et al. 2000; Dzierzanowski et al. 2011). The specific leaf area (SLA, the amount of one-sided leaf surface divided by its dry mass) appears to be a good predictor of PM accumulation and immobilisation for trees, as a proxy for the complex interactions of various leaf morphological characteristics (Muhammad 2019, 2020). In contrast, a recent study conducted by Paull et al. (2019) on 11 common green wall plant species found minimal evidence for the influence of leaf morphology on PM accumulation, but nevertheless reported differences between species. Pollution level is another important factor to be considered. Several studies suggest that the load of captured particles on the leaves is higher when the pollution level is higher (Ottelé et al. 2010; Sternberg et al. 2010; Kardel et al. 2012; Przybysz et al. 2014). In our study, however, pollution level is constant due to the nature of the experimental setup; a common garden with similar outdoor conditions and sources, which implies that differences between species can be attributed to other parameters. Also wind speed, turbulence, relative air humidity and precipitation can influence particle deposition and accumulation on plants (Litschke and Kuttler 2008; Weerakkody, John W Dover, et al. 2018). For living wall systems (LWS) in particular, insufficient research has been conducted in terms of their PM mitigation capacity, and the few studies conducted have produced inconsistent results concerning relevant parameters. Furthermore, the effects of height on PM deposition are generally poorly understood due to the lack of studies. However, green walls offer the opportunity to capture PM at lower heights than urban trees, e.g. they can be applied at pedestrian height, while tree canopies are situated higher above the ground. Furthermore, the seasonal dynamics and height variation of PM accumulation on leaves in

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LWS were studied.

In this study, it is examined to which extent PM accumulation on plants in LWS is determined by plant species and their leaf characteristics, utilizing biomagnetic analysis (SIRM; *Saturation Isothermal Remanent Magnetization*) (Hofman et al. 2013; Matzka and Maher 1999). We aim to provide suggestions to optimize the design of living wall systems in terms of plant species selection to maximise PM mitigation. Moreover, the influence planting height on PM load and seasonal variation within these plant species are included in the analysis. We hypothesize that smaller, hairier leaves with lower SLA and at lower height perform better in terms of PM accumulation capacity. Furthermore we expect an increase in PM load on the plants over the growing season.

## 2 Materials & methods

#### 2.1 Test location

Plant samples were collected at the Testing Center for Floriculture (Proefcentum voor Sierteelt, hereafter referred to as PCS) in Destelbergen, Belgium (lat. = 51.071744° N, lon. = 3.812062° E) (Appendix Figure 1). This site has a large amount of LWS which are all exposed to the same external conditions. Local pollution sources originated from the background concentrations of the city of Ghent, the Port of Ghent and the steel manufacturer Arcelor-Mittal, located west of the test site. The impact of the Arcelor-Mittal metallurgic factory on airborne pollution was discussed by (Declercq et al. 2020) Considering the main southwest wind direction in Belgium, the test site is located more or less downwind of these urban sources. Furthermore, a calm street and a railroad are located in close proximity of the test site; approximately 10 m westward and 100 m northward, respectively. The busy ring road R4 is located approximately 400 m west of the test area.

#### 2.2 Sample collection

The test facility contains 13 southwest-facing living wall systems (LWS, wall-ID 1-13), constructed in 2016 and developed by different manufacturers from Belgium and the Netherlands (Appendix Figure 2). The LWS contained a wide variety of plant species, an overview of which is given in Appendix table 1. A total of 11 species were sampled and only species that were sufficiently present for sampling without visibly damaging the system were selected for the experiment. However, not every plant species was present on each testing wall and some testing walls (wall 1 to 5 and 11) were removed during the experiment (Appendix table 2). Samples were collected in spring (28/04, sampling moment 1) and summer (24/08, sampling moment 2) of 2017 and spring (20/03, sampling moment 3) of 2018.

Since individual plants had been present for a year at the time of first sampling, environmental pollution level was equal for all systems (common garden) and the plants had developed fresh leaves since then, so that differences in plant origin were considered negligible. For magnetic analysis, from each species 4 to 10 healthy leaves (depending on the availability and to have a sufficient leaf area for testing) were collected at three different heights: (2.5 ± 0.2) m (A), (1.5± 0.2) m (B) and (0.5 ± 0.2) m (C). In order to

#### 2.3 Meteorological conditions

up to maximum 48 hours before further analysis in the lab.

Meteorological data for the entire sampling period and four extra months before were obtained from the Flanders Environmental Agency (VMM) for the nearby station M701 at Tolhuiskaai, Gent. Appendix Figure 3 shows that temperatures never fell below -5°C and never rose above 27°C. According to Köppen Climate Classification, our study area is situated in zone Cfb which is a temperate oceanic climate.

avoid contamination, the leaves were put into paper bags immediately after cutting and stored at 7°C for

#### 2.4 Leaf morphological variables

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for at least 160h at 40°C.

All leaf analyses were performed in the Laboratory of Environmental and Urban Ecology of the Bioscience Engineering Department of the University of Antwerp, Belgium. For each species considered, a series of leaf morphological variables was determined with the aim of determining key characteristics for PM capture. This was done at one sampling moment, taking only mature leaves into account. A set of morphological parameters were determined for 5 leaves per species (Appendix table 4) unless mentioned otherwise. The drop contact angle (DCA, degrees) describes the internal angle between the perimeter of a water droplet and the leaf contact surface, and is a measure for the hydrophobicity of the leaf (Muhammad et al. 2020). Leaves with a higher DCA have a higher hydrophobicity and are less wettable. To assess this variable, two fresh leaves collected on the same day and treated within 6 hours of harvesting were used. The studied leaf was cut in half and the two parts were fixed to a surface with either the adaxial or the abaxial leaf side facing upwards using double-sided tape. A 7.5 μL droplet of distilled water was put on both the adaxial and abaxial side of a leaf. A picture was taken with a Canon EOS 550D camera with a macro lens (MP-E 65 mm 1:2.8) with 3× magnification. For image processing, the software ImageJ was used with the plugin 'Low-Bond Axisymmetric Drop Shape Analysis' (LB-ADSA) (Stalder et al. 2010)(Stalder et al. 2010) to determine the mean of the left and right drop contact angles. Some examples of different DCA's are shown in Appendix Figure 4. The specific leaf area (SLA, m² kg-1) is the ratio between the one-sided fresh leaf area (Aleaf) and the leaf dry mass (M<sub>leaf</sub>). For this, 10 fresh leaves per plant species were scanned maximum 24 hours after sampling with an HP Scanjet G3100 scanner and leaf area was measured in ImageJ. Dry mass of the leaves was measured with an analytical balance (S-234, Denver Instruments, NY, USA, accuracy= 0.1 mg), after drying

Specific leaf area (SLA) = 
$$\frac{A_{leaf}}{M_{leaf}}$$

The *leaf dissection index* (LDI, dimensionless)) describes leaf shape and is the ratio of the leaf circumference (P<sub>leaf</sub>) to the root of the leaf area (VA<sub>leaf</sub>) (McLellan and Endler 1998). The lowest value is 3.545 which is for a circular leaf, while a high LDI means long or serrated leaf shape (Koch, Samson, and Denys 2019). For species with compound leaves, LDI is taken for one leaflet only.

Leaf dissection index (LDI) = 
$$\frac{P_{leaf}}{\sqrt{A_{leaf}}}$$

The *functional leaf size* (FLS) is determined by dividing the largest circular area in the leaf ( $A_{circle}$ ) by the leaf area ( $A_{leaf}$ ). This is a good measure for leaf boundary layer size, which is an important factor for PM deposition (Koch et al. 2019, 2020). For species with compound leaves, FLS is taken for one leaflet instead of the whole leaf as we assume this is aerodynamically more correct.

Functional leaf size (FLS) = 
$$\frac{A_{circle}}{A_{leaf}}$$

Finally, *hairiness* of leaves was determined by visual examination of the DCA images. Three classes were determined: no visible hairs (0), visually short hairs (1) and visually long hairs (2) (Appendix Figure 4) as viewed through the macro lens. This was based on the amount and the length of trichomes.

#### 2.5 Magnetic analysis

PM accumulation on leaves was assessed using biomagnetic analysis as described by (Kardel et al. 2011, 2012; Hofman et al. 2014a, b). Hofman et al. (2017) published a review covering all knowledge upon the publication date about PM deposition on biological surfaces. In this method, dried leaves were first tightly wrapped in cling film and placed in small plastic containers of 10 cm<sup>3</sup>. Next, the leaf samples were

magnetized (*Molspin pulse* magnetiser) in a magnetic field of 800 mT and the remanent magnetisation (in A/m) was measured in a *Molspin Minispin* magnetometer. In this way, the SIRM (*saturation isothermal remanent magnetisation*) was determined by multiplying the volume of the container for the remanent magnetization and normalizing for leaf area or mass. This variable is an indicator of atmospheric trafficand industry-related particulate matter and metals (Kardel et al. 2012; Maher et al. 2013). The SIRM values were normalized both for mass (SIRMm, A .  $m^2$  .  $kg^{-1}$  .  $10^6$ ), measured with a scientific balance (S-234, Denver Instruments, NY, USA, accuracy= 0.1 mg) and for fresh leaf area (SIRMa,  $\mu$ A) which was determined with a Li-3100 leaf area meter (Li-cor Inc., Lincoln, NE, USA). In total, 578 leaf samples were magnetically analyzed.

## 2.6 Wall Leaf Area Index and upscaling

The wall leaf area index (WLAI) is a dimensionless number that describes the amount of one sided leaf area compared to the amount of covered wall surface (Koch, et al. 2020; Ysebaert et al. 2021). Often the leaf area index is used to describe tree canopy cover, but for vertical green elements, the leaf area index has been expanded to the wall leaf area index. This is an important parameter, as it can be used to calculate the total amount of PM capture per unit wall surface covered by LWS, and is calculated as:

WLAI = 
$$\frac{leaf\ surface\ (m^2)}{wall\ surface\ (m^2)}$$

To assess this parameter, for every plant species a 50x50 cm square was cut out of one of the experimental green walls. This was done during the pruning season in August. Leaf areas were determined for fresh leaves.

SIRMa data provide information about how much magnetic particulate matter is accumulated on a certain amount of leaf surface of a certain plant species (Hofman et al. 2014). But plant species themselves also

differ in how much leaf surface they actually have. Therefore, the SIRM was scaled up with WLAI to create a variable called totalSIRM (total normalized SIRM), to give an idea of the total PM capture per unit of wall surface area.

$$totalSIRM (\mu A) = SIRMa (\mu A) \times WLAI$$

#### 2.7 Statistical analysis

Data were analyzed in R version 3.3.1 (https://cran.r-project.org/). In this study, a difference is considered significant if the p value is ≤ 0.05. Because of non-normality according to a Shapiro-Wilk normality test, the SIRM data were In transformed. A mixed model (Ime) was used to test SIRM values against species, sampling time, height and wall-ID (as a random factor). Some LWS have been analyzed more often than others and because some species have been measured multiple times on the same LWS. This means that SIRM values of species from the same wall can be expected to be more similar than on different walls, because they are in a slightly different location. In order to know the significant differences in SIRMm and SIRMa values between species, the order of species names in the model was changed until all combinations were made. The same was done for the variables sampling time and height. Differences between sampling times per species for both SIRM analyses regarding seasonal variation were retrieved with ANOVA. Furthermore, correlations of all morphological parameters were tested against each other. Because the morphological parameters were not taken from the same leaves as the SIRMm and SIRMa values, the average SIRM values per plant species were tested with a linear model against all leaf morphological parameters. Values that were almost significant were isolated and retested against SIRM parameters.

## 3 Results

#### 3.1 Difference in PM accumulation between species

The SIRM values per species normalized for mass (SIRMm), averaged for all sampling moments and all heights, ranged between 74.17 and 3543.22  $\mu$ A m<sup>2</sup> kg<sup>-1</sup> and are plotted in ascending order of log values in Figure 1. Figure 2 shows the surface area normalized SIRM (SIRMa) (values of 2.74 up to 417  $\mu$ A). There were significant differences in SIRM signal between species, of which *Bergenia cordifolia* scored lowest for mass normalization while *Santolina chamaecyparissus* scored highest. *Santolina chamaecyparissus* also scored highest for area normalization, but *Campanula* sp. scored the lowest.



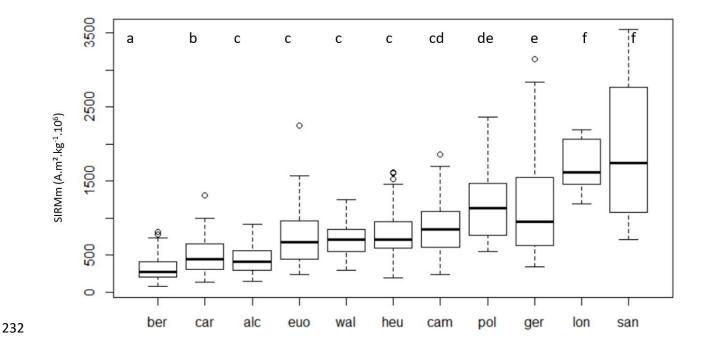


Figure 1: Boxplot of the leaf dry mass normalized SIRMm-values for all species, with indication of the mean, the first and third quartile, minimum and maximum values. Species abbreviations can be found in Appendix table 1. Species with different letters (a-f) are significantly (p < 0.05) different.

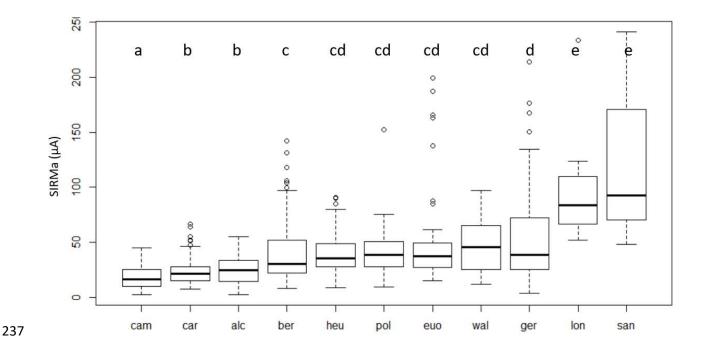


Figure 2: Boxplot of the leaf area normalized SIRMa-values for all species, with indication of the mean, the first and third quartile, minimum and maximum values. Species abbreviations can be found in Appendix table 1. Species with different letters (a-e) are significantly (p < 0.05) different.

### 3.2 TotalSIRM: average SIRMa normalized for WLAI

As mentioned earlier, in order to understand how much PM can be captured by a certain amount of foliage, it is important to know how much leaf surface is available per unit of wall surface area. In order to obtain comparable numbers, only SIRMa values of the first sampling campaign were considered, as this was the sampling with the most samples of most species (see Appendix table 2). Figure 3 shows totalSIRM values of species in ascending order.

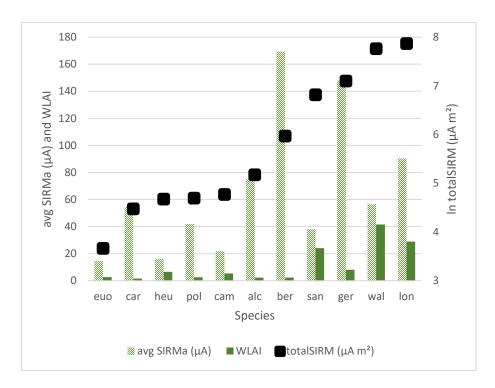


Figure 3: Total normalized In transformed SIRM values (totalSIRM) of species in ascending order (right axis). Average SIRMa and WLAI are both shown on the left axis. Species abbreviations can be found in Appendix table 1.

#### 3.3 Effect of sampling time – seasonal variation

When all species and heights are considered in the model, variation in SIRMm and SIRMa values can be found between sampling moments (Fig. 4). For SIRMm, sampling 2 was significantly different from 1 and 3. For SIRMa, sampling time 3 was significantly different from the others.

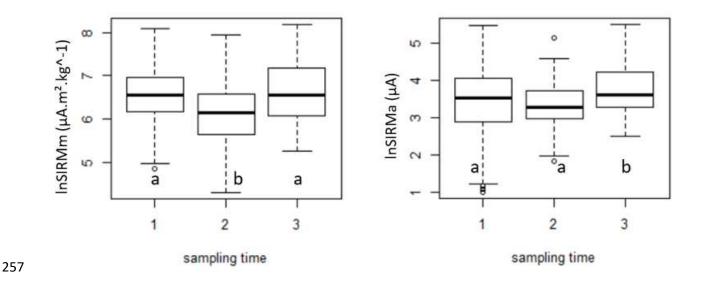


Figure 4: Boxplots of InSIRMm values at the three sampling moments (1, 2 and 3). Values with different letters (a-b) are significantly (p < 0.05) different.

Temporal patterns appear to be very different between species, both for SIRMm (Figure 5) and SIRMa (Figure 6). We categorized these patterns into 2 categories. For *Alchemilla mollis* (SIRMm and SIRMa), *Campanula* sp. (SIRMa), *Geranium* sp. (SIRMa), *Heuchera* sp. (SIRMm), *Polypodium vulgare* (SIRMa) and *Walsteinia ternata* (SIRMm and SIRMa), we observed a general increase of values in time, hereafter referred to as Category 1. Furthermore, *Bergenia cordifolia* (SIRMm and SIRMa), *Campanula* sp. (SIRMm), *Carex* sp. (SIRMm and SIRMa), *Euonymus fortunei* (SIRMm), *Geranium* sp. (SIRMm), *Heuchera* sp. (SIRMa), *Polypodium vulgare* (SIRMm) and *Santolina chamaecyparissus* (SIRMm and SIRMa) showed values that decreased or remained the same during the growing season, hereafter referred to as Category 2. Note that not every species could be sampled at every sampling moment. *Lonicera nitida* has been omitted from this analysis because it was only sampled the first time due to later removal of the walls that contained this species.

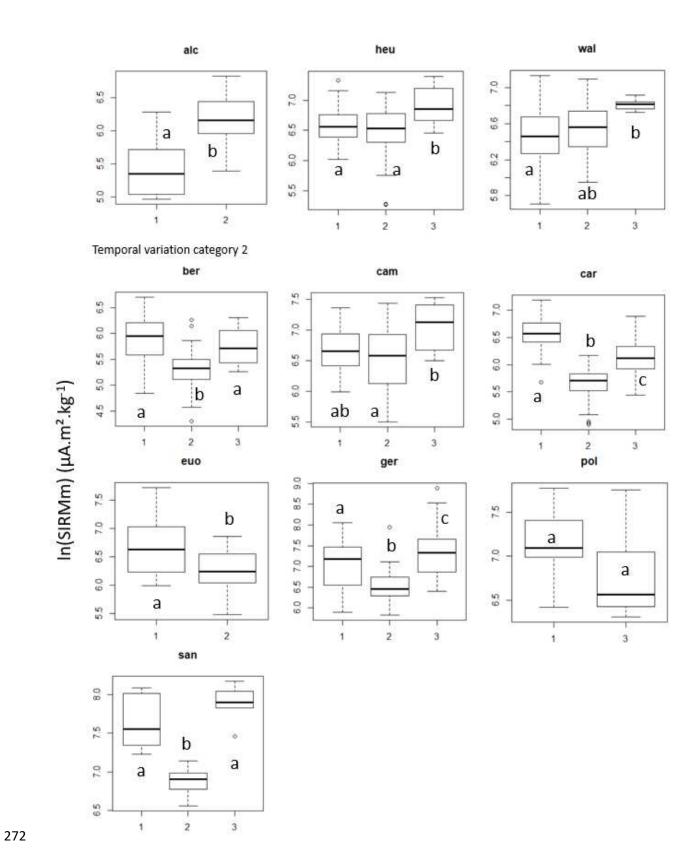
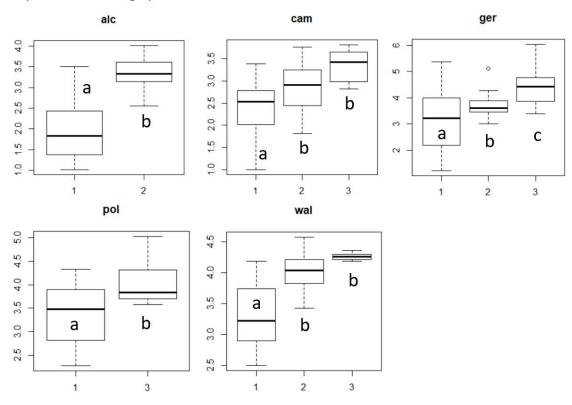
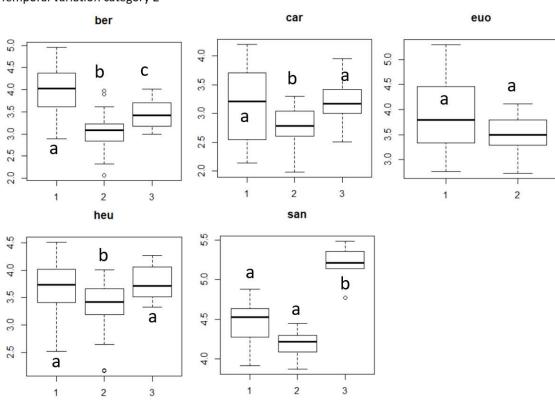


Figure 5: Boxplots of temporal variation of the mass normalized SIRM values (SIRMm) for all species (abbreviations are clarified in Appendix table 1) for the three sampling dates: sampling 1 (28-04-2017), sampling 2 (24-08-2017), sampling 3 (20-03-2018). Species with different letters (a-c) are significantly (p < 0.05) different.

#### Temporal variation category 1



Temporal variation category 2



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In(SIRMa) (μA)

Figure 6: Boxplots of temporal variation of the area normalized SIRM values (SIRMa) for all species (abbreviations are clarified in Appendix table 1) for the three sampling dates: sampling 1 (28-04-2017), sampling 2 (24-08-2017), sampling 3 (20-03-2018). Species with different letters (a-c) are significantly (p < 0.05) different.

#### 3.4 Influence of height

Vegetation was also sampled at different heights above the ground and this parameter was considered in the same model as the parameters species, sampling time and wall-ID. A indicates the highest sampling point  $(2.5 \pm 0.2 \text{ m})$  on a LWS while C is the lowest  $(0.5 \pm 0.2 \text{ m})$ . Significant differences between heights A and B vs. C were found (Fig. 7). For both SIRMm and SIRMa the highest values were found at the lowest sampling height.

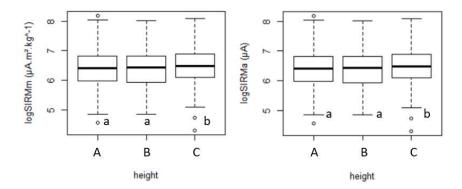
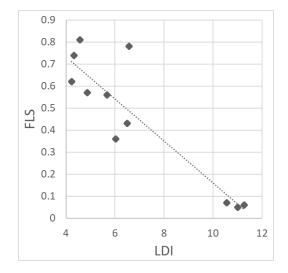


Figure 7: Boxplots of the three sampling heights (A:  $2.5 \pm 0.2$  m), (B:  $1.5 \pm 0.2$  m) and (C:  $0.5 \pm 0.2$  m). Left: SIRM normalized for mass (SIRMm), right: SIRM normalized for area (SIRMa). Species with different letters (a-b) are significantly (p < 0.05) different.

#### 3.5 Effect of morphological parameters

All mean values of the morphological parameters are shown in Appendix table 4. Before testing against SIRM values, correlations between all morphological parameters were tested against each other. Only LDI and FLS showed to be negatively correlated (Fig. 8). Furthermore, morphological parameters were statistically tested against mean SIRMm and SIRMa values using a linear model. For SIRMm, no variation could be explained by any of the morphological parameters. However, SIRMa showed almost interaction with SLA in the linear model considering all parameters. Therefore, SLA was isolated and significantly negatively contributed to the variation in SIRMa (p < 0.05, Fig. 8). For *Santolina chamaecyparissus*, no drop contact angle could be determined due to its complex leaf shape.





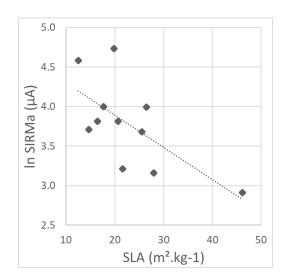


Figure 8: Left: Correlation between leaf dissection index (LDI) and functional leaf size (FLS). FLS = -0.0953.LDI + 1.1147.  $R^2 = 0.8137$ , p < 0.05. Right: Correlation between specific leaf area (SLA, in  $m^2.kg^{-1}$ ) and the ln of the saturation isothermal remanent magnetization normalized for leaf area (ln SIRMa, in  $\mu$ A). In SIRMa = -0.0407.SLA + 4.7048.  $R^2 = 0.4455$ , p < 0.05.

## 4 Discussion

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#### 4.1 Difference in PM accumulation between species

This study is the first to apply the SIRM technique to green wall plant species specifically. Magnetic biomonitoring of plant leaves has been proposed as a good indicator of ambient traffic-derived PM (Castañeda-Miranda et al. 2020; Chen et al. n.d.; Fusaro et al. 2021; Hofman et al. 2013; Matzka and Maher 1999; Winkler et al. 2022). The LWS in this common garden experiment were all exposed to the same source, which allowed the researchers to make use of SIRM as a proxy for leaf deposited PM. When looking at species and their PM load per leaf area (SIRMa) and per mass (SIRMm), large relative differences were found (SIRMa values of 2.74 up to 417 μA). Compared to the range found by Muhammad et al. (2019) (0.7 to 31.6 µA) our SIRMa values are higher. We believe this is partly because of the difference in location of the test site; our common garden had a more open character and was in proximity of a rail road, while the common garden of Muhammad et al. (2019) was in a more secluded area, surrounded by high buildings and trees and further away from a direct pollution source. We must point out that area measurement of S. chamaecyparissus may be underestimated, due to the three dimensional shape of the leaves (unlike other species that had more flattened leaves). This could result in an overestimation of SIRMa. However, for SIRMm, S. chamaecyparissus also scored highest so it can be assumed that it is generally a good PM accumulator. L. nitida and Geranium sp. also scored high for both methods. In general, we suspect that relative differences between species are due to differences in morphologies.

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#### 4.2 Influence of morphological parameters

In many studies, differences in PM accumulation capacity between species are believed to be caused by the species-specific morphological characteristics (Litschke & Kuttler 2008; Perini et al. 2017; Weerakkody et al. 2017; Muhammad et al. 2019). In this study, seven morphological parameters were assessed

(Appendix table 4). A significant negative correlation was found between SIRMa and SLA (Fig. 8, right). This means that leaves with a larger two-dimensional leaf area compared to their dry mass accumulate less PM than e.g. smaller, more rigid leaves. On the left of Figure 8 (right) there are the more 'fleshy' leaves and on the right we see the thinner leaves. This is in accordance with Muhammad et al. (2019, 2020), who suggested that SLA (together with LDI) is a good predictor of PM accumulation and immobilisation by vegetation, as a proxy for the complex interactions of various leaf morphological characteristics. This is valuable for urban managers in terms of optimizing LWS design in terms of urban air quality; smaller, rigid leaves are more desirable.

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For the other morphological parameters, no correlation with SIRMm or SIRMa values was found. This is in accordance with the findings of Chen et al. (2016), who also found differences in PM capture between species, without demonstrable species-specific criteria. Paull et al. (2019) also found no evidence for the influence of leaf morphology on PM accumulation. It is possible that the total amount of environmental PM was not well enough represented by its magnetic fraction, which usually represents the anthropogenic metallic fraction of PM, in urban contexts often dominated by vehicular traffic (Winkler et al. 2021) in order to determine differences caused by plant morphology. Other literature suggests that grass-like species (like Carex sp.) often do not show strong PM accumulation due to their soft or bendable structure that causes less turbulence than over leaves of other vegetation species (Leonard et al. 2016; Weerakkody et al. 2018a). This was also found in our study; Carex sp. was situated at the lower end of the net PM accumulation capacity distribution. Moreover, Weerakkody (2018a) found that palmately lobed leaves were the best PM accumulators, regardless of size. In our study however, LDI and FLS, which are measures of leaf shape, had no significant effect on PM capture efficiency. However, they are both correlated with each other (Fig. 8). This is in accordance with Weerakkody et al. (2018a) who also found LDI to be an insignificant parameter. In some literature a rigidity factor is suggested (Koch et al. 2020) and air flow and turbulence may be more important than plant morphology itself (Chen et al. 2016; Koch et al. 2019). Therefore, we suggest wind tunnel experiments where aerodynamic properties (i.e. leaf boundary layer, vegetation porosity, turbulent air flow, etc.) can be monitored and where an aerodynamic indicator can be determined under controlled conditions.

Similar to the findings of Perini et al. (2017), Weerakkody et al. (2018b) and Paull et al. (2019), we found no interaction between amount and length of trichomes and PM accumulation. Other researches contradict this (Beckett et al. 2000; Kardel et al. 2012; Saebo et al. 2012; Leonard et al. 2016; Weerakkody et al. 2018a; Muhammad et al. 2019), stating that hairy leaves can capture more PM because the hairs inhibit resuspension by wind or rain, and they increase the surface area available for capture. On the other hand, a high abundance of trichomes makes a leaf hydrophobic, altering leaf wettability, which can result in an opposite effect (Muhammad et al. 2019). We assume this is the case for A. *mollis*. Furthermore, there is also a wide variation of trichome size, length and texture (Weerakkody et al. 2018b) which can result in very different effects. Some of the plant species examined here can be compared with plant species from other manuscripts. In this study *Geranium* sp. has moderate to high PM capture, comparable to *G. macrorrhizum* in the study by Weerakkody et al. (2018a). *B. cordifolia* scored low to medium in this study, which was also the case according to Weerakkody et al. (2018a), who attributed this to the smooth surface of the species.

Drop contact angle is often seen as a measure for the hydrophobicity or wettability of the epicuticular wax layer, but was in this study also inconclusive with regard to PM accumulation. This is in accordance with the research by Saebo et al. (2012), Kardel et al. (2012) and Leonard et al. (2016). On the other hand, Muhammad et al. (2019) found lower PM accumulation on leaves that were more hydrophobic. We suspect that general microtopography is more important than the presence of hair or wax layer properties such as ridges and grooves. This is also suggested by Kardel et al. (2012).

#### 4.3 Seasonal variation

The mixed model showed significant differences in PM load on the LWS over the growing season. However, the course between SIRMm and SIRMa is different (Fig. 4), making it necessary to analyze each species separately. The graphs in Figures 10 and 11 show the seasonal variation of individual species. These were divided into two categories. The first category includes all species that have the most obvious result for both PM measuring methods, i.e. an increase over time. This is in accordance to the findings of Wang et al. (2013), among others, who used the gravimetric method for tree species and found that mature leaves had a higher PM load than younger leaves. Our species, evergreen or not, had not changed their leaves yet at the third sampling in the next Spring, which explains the high PM load. *A. mollis* leaves were not yet present at the time of the third sampling as they also emerge later in the season.

The second category consists of species whose PM load decreases or remains the same during the growing season and after winter, while new leaves had not yet emerged during the third sampling. For these species, we believe that the decrease or stagnation in PM load during the growing season is mainly due to PM dilution due to rapid leaf expansion. In other words, leaves expand faster than PM is deposited during the growing season (sampling 1 to 2). Because the green walls have controlled watering systems with added nutrients and are therefore in fairly ideal conditions, rapid growth is likely. Hofman et al. (2014) found similar results for *Platanus* leaves when only the leaf encapsulated fraction of PM was considered (when wash-off was excluded), further supporting our hypothesis. Subsequently, when the leaves reach maturation in late summer, PM accumulation per surface or mass will increase again until senescence. An additional explanation for this phenomenon could be the reduction of pollution input at the sampling site during the growing season, similar to the observations by Castanheiro et al. (2020). They observed an increase in PM load during the first six sampling weeks for strawberry, after which PM levels decreased again. Furthermore, resuspension by rainfall or wind can differ greatly between species due to micromorphology and plant porosity (Currie & Bass 2008; Przybysz et al. 2014). In contrast, Ottelé et al.

(2010) and Perini et al. (2017) found that seasonal variation did not affect PM accumulation. To further examine seasonal variation and the influence of meteorological conditions, laboratory experiments with controlled precipitation are recommended. It should also be noted that PM assessment techniques differ between our study and some of the literature we refer to (SIRM versus gravimetric or electron microscopic analyses), which makes direct comparison difficult, but general trends should be the same.

We believe that the different temporal variation categories are also influenced by leaf age uncertainty; *E. fortunei, L. nitida* and *S. chamaecyparissus* are evergreens, while the other species change leaves, but also in different time frames. This makes it very difficult to estimate the actual age of the leaf. In order to counter this, although very labor-intensive, labeling leaves would be a good option.

#### 4.4 Effect of WLAI

Since SIRMa is a PM measure normalized for leaf area, it can be derived from this value how much the PM collection capacity is for a given amount of leaf material. Obviously, WLAI changes throughout the season, but given the large amount of leaves needed and not to visually damage the systems, samples for leaf area determination were only taken during the pruning season. Therefore, each species has only one value for WLAI, which is at least a good representation of the differences between species, and consequently also only one value for totalSIRM. When SIRMa is multiplied with WLAI, the order of species from low to high PM capture capacity changes slightly, but generally remains the same; *S. chamaecyparissus, Geranium* sp. and *L. nitida* again perform the best, together with *W. ternata*. In general, a higher WLAI is believed to be better in terms of PM capture (Weerakkody et al. 2017). However, it should be noted that WLAI only expresses the total amount of leaf area per wall area and not the three-dimensional plant configuration.

#### 4.5 Vegetation height

In general, biomagnetic monitoring has been proven useful for assessing the three-dimensional spatial distribution of particulates (Mitchell & Maher 2009; Kardel et al. 2011; Hofman et al. 2013). Hofman et al. (2013) found that SIRM values decrease exponentially with tree leaf sampling height and tree crown height, respectively. SIRM is a grainsize dependent magnetic parameter; thus, the use of SIRM as a proxy for the concentration of airborne PM must be done under the assumption that particle size is constant. This could not be the case for different sampling heights, because heavier particles gravitationally stay closer to the ground than finer particles and thus particle size is not equally distributed over the walls vertically. Moreover, the finest ultrafine particles, that are superparamagnetic at room temperature, do not contribute to SIRM. So, for determining the magnetic properties according to the grainsize, the combined used of magnetic susceptibility, hysteresis loops and electron microscopy techniques should be applied.

The highest values for both SIRMm and SIRMa were found at the lowest sampling height, i.e. 0.2-0.5 m above the ground (Fig. 7). This is in accordance with the majority of literature on the spatial difference in SIRM values. This could mean that, when traffic is the main pollution source, it is most effective when green walls are applied from ground level and upwards for pedestrian health then when they are only applied from a certain building wall height. However, because SIRM is dependent on particle composition and size and heavier particles tend to stay closer to the ground, there is still some uncertainty regarding height differences.

#### 4.6 Consequences for human health

The bioaccumulation of PM in this study was solely defined with SIRM. This technique has proven to be very useful when it comes to biomonitoring analyses and big data collection, in the lab of the researchers

and many others (Hofman et al. 2017). However, it has to be noted that this method only works for health assessment when harmful PM is well represented by its magnetic fraction (Winkler et al. 2021). This Is not necessarily the case in this study, as ultrafine particles probably settle at the highest heights, but do not contribute to SIRM, and therefore no suggestions can be given health-wise. In exceptional cases there can even be a negative correlation between SIRM and airborne PM concentrations (Petrovský et al. 2020). However, because our study was a common garden experiment with the same PM sources and meteorological conditions, we assume this very unlikely. The fact that SIRM does not take into account the smallest magnetic PM fraction (magnetite < 35nm), which also has the most implications for health, also deserves more attention.

#### 5 Conclusion

In this study, 11 commonly used living wall system (LWS) plant species were assessed in a common garden experiment via SIRM (*Saturation isothermal remanent magnetization*, both normalized for leaf area and mass) in terms of relative particulate matter (PM) accumulation and its change over time and in height. Large differences in total SIRM values between species were found. *S. chamaecyparissus*, *Geranium* sp., *L. nitida* and *W. ternata* were the best PM capturers. In general, we can conclude that green wall plant species are definitely able to capture PM out of the environment. However, careful species selection and implementation is important. This study shows how complex interactions can be between an engineered vegetation system and its environment and demonstrates the need for optimal plant composition and system design. Specific leaf area (SLA) showed a negative correlation with SIRM normalized for leaf area (SIRMa). This suggests that SLA is a good representative for the leaf morphological complex. Hereby it is interesting for LWS designers, professionally or self-builders, to select plant species based solely on their fleshy or thin appearance, in order to have an idea of their relative PM accumulation capacity, without the

need for PM analysis on the leaves, which is much more time consuming and expensive. Furthermore, species that perform poorly in PM accumulation per unit area cannot make up for this by simply making lots of leaves. It is important to include wall leaf area index (WLAI) because the species that perform best taking the WLAI into account are not necessarily the species that score highest in terms of PM accumulation (SIRM).

For future research we suggest additional laboratory experiments under controlled circumstances: correct leaf age assessment by tagging, leaf expansion, senescence, monitored PM deposition, resuspension and controlled precipitation and irrigation. These 'ideal' conditions could be a proxy for making estimates for outdoor situations, which can then provide a comparison with this experiment. Furthermore it would be useful to asses further magnetic parameters, e.g. sensitive to ultrafine particles or grainsize independent for the total concentration. Taking foliage permeability and porosity into account would also greatly contribute to a better understanding of how air flows through vegetation and its three-dimensional structure. Nevertheless, field experiments examining the systems under their real conditions remain as important.

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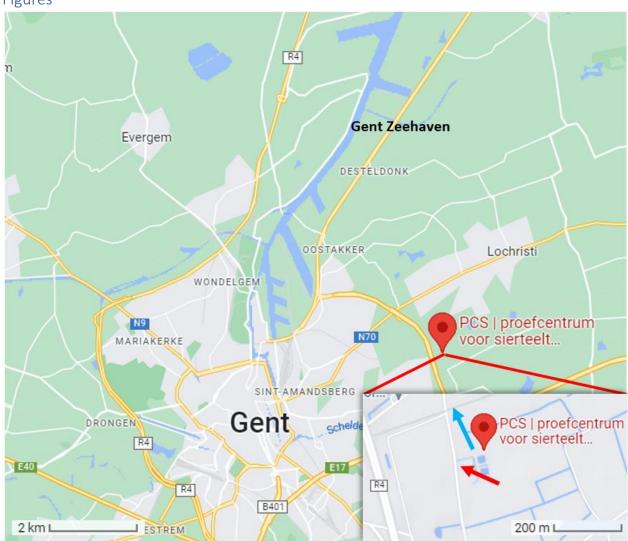
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The influence of plant species, leaf morphology, height and season on PM capture efficiency in living wall systems

## **Appendix**

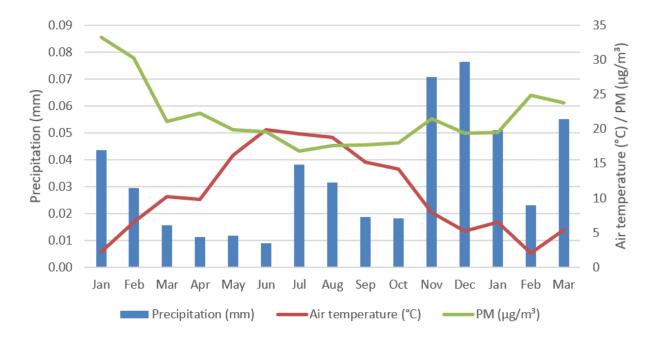
**Figures** 



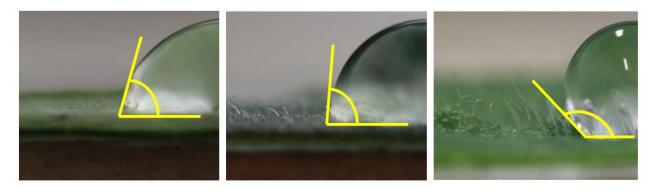
Appendix Figure 1: Location of the Testing Center for Floriculture (Proefcentum voor Sierteelt) in proximity of Ghent city and the Port of Ghent (Gent Zeehaven) in the North. Inset: Red arrow indicates the nearest road; blue arrow indicates the rail road. (Source: Google Maps)



Appendix Figure 2: Several individual test walls on the facility at PCS, Destelbergen. Wall X was placed later, and therefore not tested.



Appendix Figure 3: Average monthly air temperature (red line), average monthly PM concentration (green line) and monthly precipitation (blue bars) from 1/01/2017 until 20/03/2018 (station M701, Tolhuiskaai, Gent and station 44R710) (source: Flanders Environmental Agency (VMM)).



Appendix Figure 4: drop contact angle: different angles are shown in the pictures. Hairiness: from left to right; no visible hairs (class 0), short or few visible hairs (class 1) and many or long visible hairs (class 2) as viewed through the macro lens.

#### Tables

Table 1: Overview of all investigated plant species, their leaf shape and the abbreviations used in the text are mentioned between brackets. Photos are not actual size, nor relative size to each other.

Alchemilla mollis (alc)	Bergenia cordifolia (ber)	Campanula sp. (cam)	Carex sp. (car)
Euonymus fortunei (euo)	Geranium sp. (ger)	Heuchera sp. (heu)	Lonicera nitida (lon)
	樂		
Polypodium vulgare (pol)	Santolina chamaecyparissus (san)	Waldsteinia ternata (wal)	
***	***		

Table 2: Plant species that were sampled on the different living wall systems throughout the experiment. Numbers indicate wall-ID. Species that were sampled at all three sampling dates at a certain wall are indicated with a '\*'.

Species	28 April 2017	24 August 2017	20 March 2018
A. mollis	8	8, 11, 13	
B. cordifolia	1, 2, 3, 4, 6*, 8*, 11, 13	4, 6*, 8*, 11, 13	6*, 8*, 13
Campanula sp.	2, 5, 6*, 12,	5, 6*, 12	6*
Carex sp.	7*, 9*, 12, 13*	7*, 8, 9*, 12, 13*	7*, 8, 9*, 13*
E. fortunei	2, 3, 4, 5	5	
Geranium sp.	1, 7*, 8*, 12, 13	7*, 8*, 12, 13,	7*, 8*
Heuchera sp.	3, 4, 6, 9, 11, 12	4, 6, 9, 11, 12	6, 9
L. nitida	1, 11		
P. vulgare	6, 9		6
S. chamaecyparissus	7*	7*	7*
W. ternate	2, 3, 4, 5, 6 *	4, 5, 6*	6*

Table 3: Average wind directions per month prior to and during the sampling period. N = north, E = east, S = south, W = west (station 44R701, Baudelohof, Gent (source: Flanders Environmental Agency (VMM)).

	2017									2018					
Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar
Wind direction	S	S	S	WSW	S	S	SW	SSW	SSW	SW	WSW	SW	SSW	SE	S

Table 4: Average morphological leaf variables +/- standard deviation for all species. DCA = drop contact angle (ad = adaxial, ab = abaxial)(degrees), SLA = specific leaf area ( $m^2$  .  $kg^{-1}$ ), LDI = leaf dissection index (dimensionless), FLS = functional leaf size (dimensionless), hairiness categories: no visible hairs (0), short hairs (1) and long hairs (2). NA = not applicable.

Species	WLAI	DCA (ad)	DCA (ab)	SLA	LDI	FLS	Hairines	
							S	
A. mollis	2.37	149 (±	146 (± 1.16)	21.63 (± 6.58)	5.68 (± 0.33)	0.56 (± 0.11)	2	
		0.12)						
B. cordifolia	2.32	96 (±1.21	56 (± 0.75)	14.70 (± 6.44)	4.32 (± 0.12)	0.74 (± 0.06)	0	
Campanula sp.	5.41	77 (± 0.59)	76 (± 1.90)	46.11 (± 11.90)	4.87 (± 0.20)	0.57 (± 0.06)	1	
Carex sp.	1.61	70 (± 3.06)	107 (± 1.52)	27.96 (± 4.43)	10.56 (± 0.92)	0.07 (± 0.01)	1	
E. fortunei	2.66	76 (± 0.64)	92 (± 0.42)	17.71 (± 0.71)	4.57 (± 0.28)	0.81 (± 0.05)	0	
Geranium sp.	8.19	93 (±1.10)	105 (±	26.50 (± 6.41)	6.04 (± 1.04)	0.36 (± 0.17)	2	
			0.80)					
Heuchera sp.	6.60	98 (± 0.94)	120 (± 0.74)	25.51 (± 5.04)	6.50 (± 0.51)	0.43 (± 0.09)	2	
L. nitida	28.94	103 (±	86 (± 1.40)	12.49 (± 0.43)	4.23 (± 0.33)	0.62 (± 0.05)	0	
		1.52)						
P. vulgare	2.61	98 (± 1.34)	99 (± 1.15)	16.49 (± 4.78)	11.02 (± 4.03)	0.05 (± 0.03)	0	
S.	24.04	NA	NA	19.79 (± 2.78)	11.28 (± 2.09)	0.06 (± 0.03)	2	
chamaecyparissu								
S								
W. ternata	41.54	112 (±	92 (± 0.88)	20.66 (± 6.20)	6.57 (± 0.39)	0.78 (± 0.08)	2	
		2.51)						