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Phytoremediation Effectiveness of Marginalized Vegetation as Green Bio-Filter of Particulate Matters in The Southwestern of Japan (Fukuoka)

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Abstract: Spectacularly, particulate matter pollution influences human health through aggravating numerous diseases and causes premature deaths. In order to eliminate its concentration in the ambient air, plants species can act as natural bio-filters and capture, degrade and metabolize air pollutants inside their foliage. In a study carried out at three sites with different pollution levels and plant types. The immobilization efficiency of Particulate Matter (PM) was investigated in the leaves of three species (shrub, climber, and herb) in the southwestern of Japan with a time gap of 14 days. Two healthy mature leaf samples of each specimen were carefully collected and analyzed using gravimetric analytical method. Significant quantity of PM in three size fractions with aerodynamic dimeter ranged between (0.1-100 μm) were captured inside the leaf foliage of the analyzed species. Fine particles (2.5 -10 μm) dominated the highest portion of the PM deposition captured by the analyzed species with 70.6 $\mu g.cm^{-2}$(39.5%). Shrub species represented with Elaeagnus pungens proved to be the most efficient species among the analyzed species. Leaf traits such as such as grooves, trichomes, roughness and margin are considered as key factors associated with positive PM deposition, whereas total surface area of the leaves had no direct correlation to PM deposition.

Keywords: pollution; phytoremediation; plants; PM mitigation; deposition; foliage; morphological traits; particulate matter

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1. Introduction

Human anthropogenic activities are considered as the main source of diverse toxic substances for human health, especially, Particulate Matter (PM), which is a mixture of fly ash, dust, black carbon, organic and non-organic compounds, trace elements (TE) and other substances that can be suspended in the air for a long time and transported to long distances from the pollution sources in urban air. PM classified according to aerodynamic dimeter into four categories, total suspended particles (TSP) with a diameter ≤100 μm, coarse particulate matter (PM10) with an aerodynamic diameter ≤10 microns, fine particulate matter (PM2.5) with an aerodynamic diameter equal to or less than 2.5 microns and the most harmful component, ultra-fine particulate matter (PM1) with a diameter of ≤1.0 μm [1,2]. Unfortunately, long-term exposure to these particles with such small dimeter can create systemic inflammatory vicissitudes and exaggerate noncommunicable diseases (NCDs), respiratory diseases, fatal childhood diseases including post-neonatal infant mortality, sudden infant death syndrome (SIDS) cardiopulmonary diseases, lung cancer, atherosclerosis and cardiovascular diseases which may furthering to the so-called 'sick building syndrome'(SBS) and 'building-related illnesses'(BRI) [3–8].

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As a result, it causes approximately 2.1 million worldwide premature deaths annually [9,10]. Particulate matter pollution in Japan has acute health effect, as trace elements, such as Se, Pb, As, Zn, Cu, Cr, Ni, and Cs which distribute homogenously within the particles are associated with nasal, ocular, skin symptoms and severe non-carcinogenic risk to moderate carcinogenic risk for healthy school students [11]. Furthermore, Takeda et al., [12] demonstrated that sulfate long term exposure (component of PM) can decrease the placental weight in the third trimester with a difference of -6.7g from the normal birth weight.

Therefore, several policies, standards and techniques were authorized and applied to eradicate the environmental and health effects of particulate matter. World Health Organization Air Quality Guidelines which are based on systematic reviews and subsequent evaluation methods, serve as global target for local and international governments to work towards improving life quality through eliminating air pollution and constraining their rates. On the other hand, there are innumerable controlling, monitoring and remediation methods that were used to deteriorate the PM pollution values, for instance, chemical methods, which depend on wet, dry and occult deposition [13–15]. Phytoremediation as a natural plant-based environmentally friendly biotechnology approach, involves transferring, removing, detoxifying, degrading, or sequestrating, partially metabolizing and accumulating air and soil contaminants such as the particulate matter, by adhesion to vegetation leaves foliage or absorbing inside roots (plant's rhizosphere) can be used as alternative for complicated and costly techniques [16–18].

Various empirical studies reassured the efficiency of PM removal capacity in different plant types via deposition onto leaves such as Sgrigna et al., [19] and Viecco et al., [20] which evaluated the impact of biodiverse Green Roofs (GRs) and Green Walls (GWs) on PM2.5 capturing, through analyzing seven species using gravimetric dry deposition method. The study concluded that the higher the biodiversity of plants in GRs and GWs, higher the PM2.5 quantity can be captured. Whereas Dzierżanowski et al., [21] proved that four species of roadside trees (Acer campestre L., Fraxinus excelsior L., Platanus × hispanica Mill. ex Muenchh. 'Acerifolia', Tilia cordata Mill), three species of shrubs (Forsythia × intermedia Zabel, Physocarpus opulifolius L. Maxim., Spiraea japonica L.), and one climber species (Hedera helix L.) were able to filtrate the ambient air and capture significant quantity of particulate matter on their leaves. However, plants species differed in their removal capacity of PM. Due to large leaf area and crowns which promote vertical transport by enhancing turbulence [22]. Trees are believed to be the most efficient plant group in scavenging particulate matter according to Sæbø et al., [23] and Muñoz et al., [24]. However, there is an increasing hypothesis that trees can affect air quality negatively, through emitting toxic compounds, such as volatile organic compounds (VOCs), which may contribute to ozone formation [25]. Nevertheless, contributions by other plant species to PM removal was underestimated, and there is still a lack of knowledge about the deposition of PM on other types of leaves, especially the marginalized plants that grow and interact with trees while having the possibility to collect the same quantity of PM or even more [16,26]. The present study aims to investigate and address the capturing ability of PM in shrubs, climbers, and herbs, while considering the substantial associates among PM deposition and leaf size, leaf compositional morphology, structure, and plant type with hypothesis that the plants with large leaf areas are supposed to capture more particles than smaller plants.

2. Materials and Methods

2.1. Leaves samples and experimental sites

Three species types of broad shaped leaves represented by Elaeagnus pungens (shrub), Dioscorea japonica (climber) and Cirsium vulgare (herb) were analyzed in the present study (more details shown in Table 1). These were all commonly planted in urban areas, alongside the roads and streets at Kyushu University's campus (Fukuoka, Japan) (shown in Figure 1) with a distance between the road and the analyzed shrub and herb ranges from 1 to 2 m, while there is no significant distance between the road and the climber species. With gap of 14 days (experimental period), two mature healthy leaf samples (n = 20 leaves/ sample) were collected, when there was no previous rainfall for

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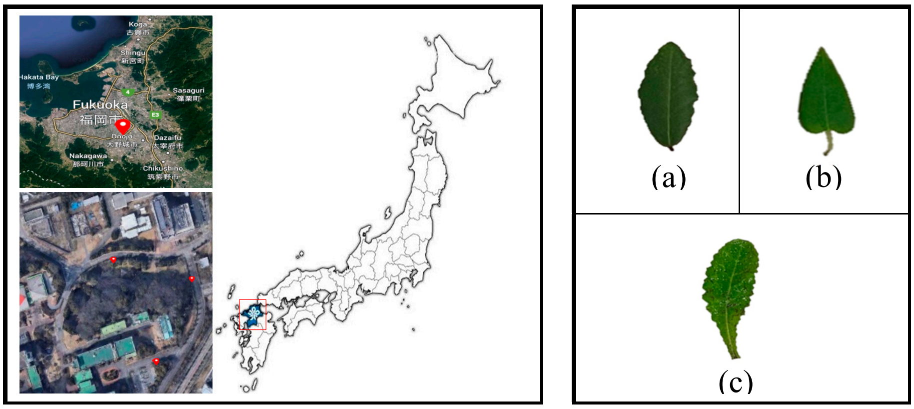
more than 3 days. Sampling height varied from 0.7 to 5 m above the ground level depending on plant structure. The sampled leaves were individually placed and sealed into pre-labelled sample paper bags since PM is susceptible to static electricity and stored at 4°C in a clean laboratory refrigerator before conducting the analysis [27].

Table 1. Analyzed species leaf characteristics according to North Carolina university plant box.

Species	Botanical name	Habit	Leaf length	Leaf shape	Hair	Leaf margin	Leaf arrangement	Leaf surface
Thorny Olive	Elaeagnus pungens	Shrub	3-6 inches	Ovate	-	Undulate	Alternate	Leathery
Japanese mountain Yam	Dioscorea japonica	Climber	3-6 inches	Cordate, heart shape with 3 lobes	-	Lobed	Opposite	sub-pleated
Bull Thistle	Cirsium vulgare	Herb	6 inches	Lanceolate	+	Lobed	rosulate	Hairy

The weather conditions were standard for the summer season in Fukuoka prefecture, sunny, very hot, humid with a middle level rainfall, and the monthly temperature ranged from 26.8-33.8 °C. Fukuoka WMO on-site station (ID:47807) recorded monthly total precipitation in August 2022 with 266.5 mm and 2.7 m/s monthly mean wind speed in this location, according to Japan Meteorological Agency [29].



(A) (B)

Figure 1. (A) Samples site locations in Kyushu university, Fukuoka, Japan. (B) Selected leaf Images (B(a); Elaeagnus pungens, (B(b); Dioscorea japonica, B(c); Cirsium vulgare.

2.2. Quantitative Gravimetric analysis of PM

Extracted PM quantity was analyzed according to Dzierżanowski et al. [21] dry gravimetric method. Two standard solutions are supposed to wash the sampled leaves, (i) Distilled water (DW) and (ii) Chloroform which was used previously as a solvent to dissolve non-polar molecules and the soluble fraction of Poly Aromatic Hydrocarbons (PAHs), as a result, there is a possibility to affect the actual weight of PM. Therefore, Sodium dodecyl sulfate (SDS)($CH_3(CH_2)_{11}OSO_3Na$) (Merck, Germany) with a 5% concentration was used to deep washing of plants leaves and fully extraction of PM as it is a strong anionic surfactant that can attract charged particles and decrease the water surface tension without affecting the chemical composition of the particles [30]. A trial experiment with 10% concentration was applied, however high concentration of SDS produced more foams and impeded the filtration process as its bubbles can occupy the filter membrane gaps instead of extracted particles that were enforced to fall with filtrated solution. Both adaxial and abaxial surfaces of the leaves were washed for 2 min with 250 mL of distilled water and after with 150 ml of SDS for 1 min. Thereafter, the washing solutions were applied to four filtration tools through (i) polypropylene sieve (with

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retention 106 µm, Ø100mmϕ, H40, Itoh seisakusho Manufacturing, Japan) then (ii) 10 µm Omni pore membrane filter (Merck millipore Ltd, Ireland). (iii) 2.5 µm filter paper (Whatman filter, Cytiva, Japan) and finally (iv) 0.2 µm PTFE membrane filter (Merck Millipore Ltd, Ireland). Filtration system setup consisted of a 25 mm vacuum filter holder with stopper support assembly connected to a vacuum pump. Based on this setup, three fractions of PM: (i) 10–100 µm (coarse), (ii) 2.5–10 µm (fine) and (iii) 0.2–2.5µm (ultra-fine) were successfully extracted. The filters were dried before and after filtration for 30min at 60 °C, stabilized in the weighing room for 30 min, and weighed using electronic balance (HT124R, ViBRA, Japan). The total leaf area of plant leaves was measured using ImageJ software (U. S. National Institutes of Health [31]) depending on the same method of Osunkoya et al. [32]. Allowing the amount of PM to be expressed as $\mu g.cm^{-2}$. Further analysis was conducted using LEXT OLS4000 3D Laser Measuring Microscope (Olympus, Japan) to test the relationships between leaf traits in each selected species and PM accumulation.

Calculations

The particulate matter deposition in $\mu g/cm^{-2}$ was measured through this equation according to [21].

$$\text{PM deposition} = W_{F+PM} - W_F/A, \quad (1)$$

W_{F+PM}: Weight of filters and PM after filtration in µg.
W_F: Weight of blank filters before filtration in µg.
A: Total surface area of the analyzed leaves in cm^2.

3. Results

There was a substantial interaction among the analyzed species and ambient PM emerged with a difference in the removal efficiency of all PM fractions in both samples and period. A notable quantity of PM was found on the foliage of the anlyzed species throughout the experiment period with total of 179.02 $\mu g.cm^{-2}$.

Considering the findings of Dzierżanowski et al. [21] and Sæbø et al. [23], large size fractions dominated the highest proportion of all deposited PM. Similarly, in the present study, the majority of PM deposited on plants` foliage was large-size particles represented by the fine size fraction (2.5 - 10µm) which recorded total of 70.6 $\mu g.cm^{-2}$(39.5%), followed by the coarse fraction (10 -100µm) with 69.8 $\mu g.cm^{-2}$ (39%), whereas the ultra-fine fraction (0.1 -2.5 µm) was the lowest with 38.5 $\mu g.cm^{-2}$ (21.5%) as it shown in Figure (1, a). The miniscule size of ultra-fine size fraction makes them more susceptible to weather conditions which disrupts, diminutions the continuous stable deposition and allows to be washed off by precipitation or resuspended with air turbulence.

Shrubs with large leaf areas similar to trees were proved to accumulate substantial quantities of particulate matter [33]. In the present work, we found that the analyzed shrub (Elaeagnus pungens) showed to be the most efficient plant species in PM accumulation, while Cirsium vulgare (herbaceous species) recorded the lowest deposition (shown in Figure (1, b). Similar conclusions were confirmed by Dzierżanowski et al., [21] who found largest Quantity of PM on shrubs leaves (Spiraea). On other hand, Ottelé et al. [34] did not observe any difference in PM accumulation among sampling heights over the range of 0.75–2.0m.

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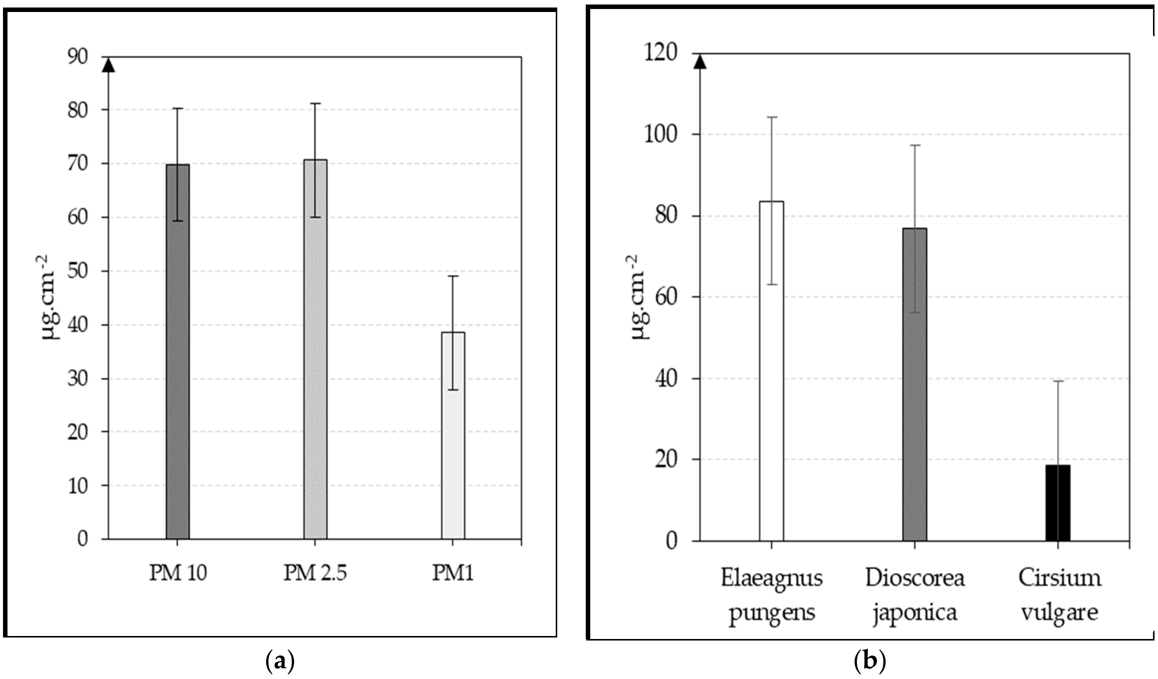

Figure 2. (a) Total PM deposition(µg.cm^{-2}) in the analyzed species, (b) Total PM fractions(µg.m^{-2}) deposition in the analyzed species.

Among the tested plants species, Dioscorea Japonica captured the highest quantity of the coarse size fraction (10-100 µm) with 40.04 µg.cm^{-2}, while Elaeagnus pungens has recorded the greatest values of fine size fraction (2.5 -10 µm) and ultra-fine size fraction (0.1-2.5 µm) with 41.4 µg.cm^{-2} and 19.3 µg.cm^{-2}, respectively (Table 2).

Table 2. Accumulation of particulate matter (µg.cm^{-2}) within three different size fractions on leaves of the analyzed species.

Species	PM size fractions accumulation(µg.cm^{-2})					
	Coarse (10-100µm)	(%)	Fine (2.5-10 µm)	(%)	Ultra-fine (0.2-2.5 µm)	(%)
Elaeagnus pungens	22.8	12.7	41.4	23.1	19.3	10.8
Dioscorea japonica	40.04	22.4	21.5	12.03	15.2	8.5
Cirsium vulgare	6.9	3.9	7.7	4.3	3.9	2.2

4. Discussion

Shrubs presented the major percentage of low height strata that being planted on roadside in urban areas, due to small, occupied spaces and large, biologically active biomass to filtrate ambient air from toxic gases and particulate matter, in particular, fine size particles which considered, besides ultra-fine size fraction as the most harmful PM components. According to Stapleton et al., [35], shrubs can lessen to 5.9% of PM2.5 concentration in small spaces. Moreover, plants species growing near to the ground, such as shrubs, can capture more PM on their leaves which are exposed to different types of pollutions such as, soil splash [36].

The analyzed shrub, Elaeagnus pungens captured the largest portion of all accumulated PM with total of 83.6 µg.cm^{-2}. Fine size fraction (2.5-10 µm) made up the greatest proportion of

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accumulated PM mass and the ultra-fine size fraction (0.1-2.5 μm) recorded the smallest proportion. Expressed as a percentage, 27.3%, 49.5%, and 23.2% for coarse, the fine and the ultra-fine size fractions, respectively.

In the light of fact that green walls (GWs) (vertical greening) climbing on the surface of buildings and along barriers of urban streets and roads that increasingly exposed to progressive PM pollution rates, climbers can act as an attractive alternative to trees and shrubs as it can overcome most of the limitations related to large leaf area such as space accessibility, sub-surface infrastructure, sunlight, and suitable soil conditions, through minimizing space-take and providing further advantages including thermal insulation, noise lessening, conservation of urban biodiversity and air purification [37–41]. Furthermore, climbers through interacting with trees and shrubs, can capture more quantities of particulate matter than other species. As they can produce an enormous leaf area (in relation to the land area occupied) comparing with low height strata which enhance the capability of accumulating PM and other pollutants [33]. Dioscorea Japonica has recorded total of 76.8 $\mu g.cm^{-2}$, which is the second-highest particulate matter deposition among the analyzed species. Coarse size fraction (10 -100 μm) represents the highest portion with 52.1%, while fine size fraction (2.5-10 μm) recorded 28.05% and finally 19.8% for ultra-fine size fraction (0.1-2.5 μm) of all deposited PM on the leaves of the analyzed climber (Dioscorea japonica).

Weber et al., [42] confirmed the significant PM removal capacity of 16 species of herbaceous plants, especially, because herbaceous plants can capture the fallen particulate matter from trees, shrubs, and climbers, moreover, can tolerate urban conditions effectively and adapt to air pollution chemical and physical alternations, whereas some species can resist these changes completely [43,44]. However, in this study, Cirsium vulgare (herbaceous species) captured lower particulate matter (18.6 $\mu g.cm^{-2}$, 10.4%) than the other tested species due to smaller surface area, higher influence of weather conditions. Fine size fraction (2.5 -10 μm) dominated the largest portion for with 7.7 $\mu g.cm^{-2}$(4.3%), followed by coarse size fraction (10 -100 μm) with 6.9 $\mu g.cm^{-2}$ (3.9%), whereas ultra-fine size fraction (0.1 -2.5 μm) recorded the lowest deposition with 3.9 $\mu g.cm^{-2}$ (2.2%). The particulate matter accumulation was similar in both samples of Cirsium vulgare (Figure 4), in contrast of other species which displayed deterioration in the second sample, because of weather conditions such as air turbulence and precipitation that proved to wash off 20% of the deposited PM on plants leaves according to Kaupp et al., [45].

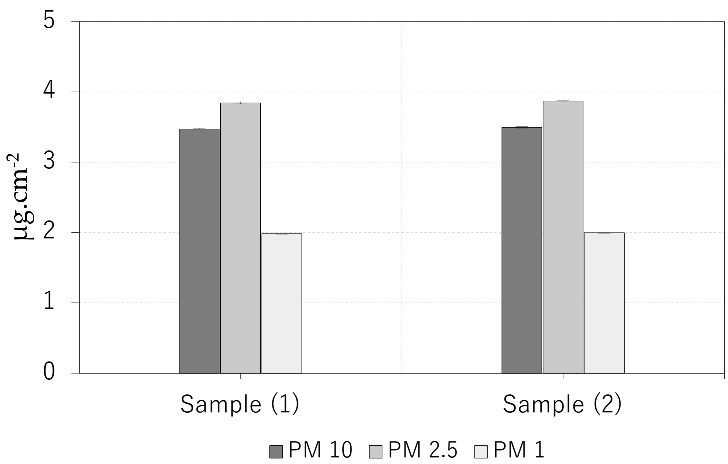

Figure 4. Accumulation of particulate matter (means± SE) ($\mu g.cm^{-2}$) within three different size fractions on leaves of Cirsium vulgare.

Trees, shrubs, climbers, and herbs can filtrate the ambient air through natural based phenomenon which called phytoremediation, through transferring, removing, destroying, or stabilizing and accumulating of aerosol particles and other pollutants via leaf stomata or roots [9,10], [46].

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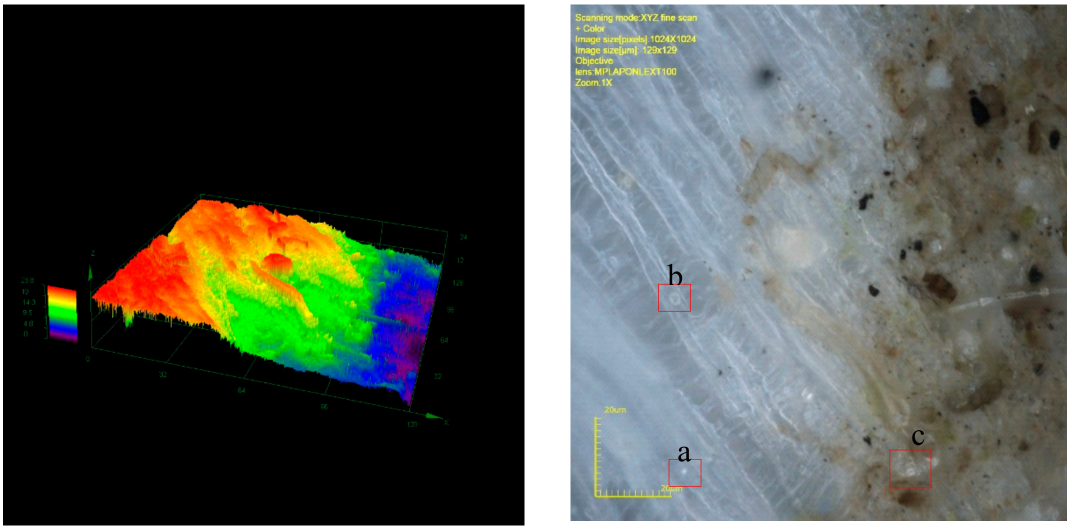
Nevertheless, these species differed in their capabilities to accumulate particulate matter and other pollutants which affected with several factors. A strong association between particulate matter adsorption capabilities and leaf morphological traits, such as leaf shape, leaf size, hair presence, rough leaf surface (grooves). etc. was demonstrated in the previous studies such as Freer-Smith et al., [26] and Leonard et al., [47].

To exemplify, Elaeagnus pungens, which has a leathery rough compositional surface, as it is clear in the 3D microscopic image (shown in Figure 5 (a) (3D Laser Measuring Microscope), captured the highest total PM among the analyzed species with 83.6 μg.cm^{-2} included three size fractions (coarse, fine and ultra-fine) as it shown in Figure 5 (b). Another factor affects the deposition in the analyzed shrub, its location which was close to two of air polluted resources (the main road and car parking) this may be the reason of higher deposition on its foliage comparing with the other analyzed species.



(A) (B)

Figure 5. (A) Topographical image shows the roughness of the leathery surface of E. pungens leaf. (B) 3D Microscope image of leaf micromorphology on the adaxial surface of E. pungens, (a, b and c) represent different sizes of particulate matter (x100).

Furthermore, leaves with ridges and grooves such as Dioscorea japonica allowed to capture significant quantity of large size particles (61.6 μg.cm^{-2}) as their size can perfectly adhere to surface grooves, meanwhile, ultra-fine size fraction (0.2-2.5μm) which recorded 15.2 μg.cm^{-2} for both samples, can partially adhere to the same microfeatures with high possibility to be removed with air turbulence and precipitation which causes flowing down of particulate matter to the soil, where it can be absorbed by the plant's rhizosphere and the roots [48]. (Figure 6).

Considering Dzierżanowski et al. [21], Sæbø et al. [23], PAULL et al. [49] and Fernàndez et al. [50] findings, it can be concluded that the presence of the leaf hair (trichomes) has been associated with positive PM accumulation in diverse analyzed plants as some leaf hairs presenting hydrophobicity, attracting charged particles, for instance, heavy metals. In this work, we found also that hair presence can enhance the PM accumulation on the leaves. The hairy surface of Cirsium vulgare allowed to accumulate considerable amount of course, fine and ultra-fine size fractions of PM with 15.6 μg.cm^{-2}, 20.9 μg.cm^{-2} and 12.02 μg.cm^{-2}, respectively.

That proves that leaf structural traits such as roughness, grooves, trichomes (leaf hair) and leaf morphology may influence the transition from a laminar to a turbulent airflow, therefore the accumulation speed on a rough surface will be higher than a smooth surface. [51,52]. Furthermore, rough surface can increase the contact time with rain droplets as it can create dissimilar contact angles (θ) (representative of leaf wettability) between the water droplet and the leaf surface, which enriches larger sized particles adherence to the leaf surface for a long time and delays their washing off.

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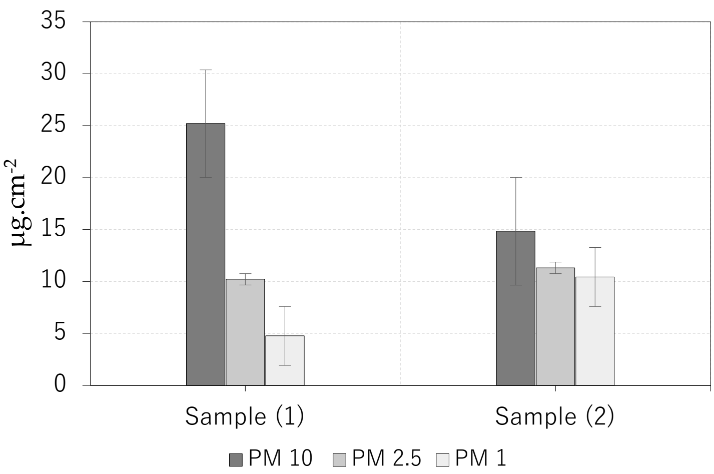

Figure 6. Accumulation of particulate matter (means± SE) (µg.cm^{-2}) within three different size fractions on leaves for Dioscorea japonica.

According to Mitchell et al. [53], PM deposited mainly on leaf margins or along the main leaf nerves, therefore, leaf margin can also enhance particulate matter accumulation as Elaeagnus pungens, undulate margin, with wavy shape that curves up and down, for instance, can prevent the particulate matter from falling from the leaf surface, while the lobed margin which belong to Dioscorea japonica and Cirsium vulgare can partially impede the falling of particulate matter from the leaf surface with the support of additional traits such as grooves and trichomes in each species.

On contrary, leaf surface area (LA) was not significant trait to collect more particles in the analyzed species. Although, Cirsium vulgare which has the largest LA that can reach 34.39 cm^2, it did not record the highest deposition of PM, as a comparison between the three species, while the lowest surface area (average per leaf) represented by Dioscorea japonica with a 1.6 cm^2 recorded high deposition in the three size fractions (coarse, fine and ultra-fine). On the contrary, Weerakkody et al. [53] noted that the small-leaved species showed comparatively low PM deposition, suggesting that this was due to their lower roughness and the low capacity to withstand PM contaminated air flow, thus lowering the turbulence surrounding the leaf boundary. Nonetheless, the study concluded that small-leaved species with enough complex morphology were the most efficient species for capturing PM, proving that the leaf size can provide only marginal assumptions about PM accumulation capabilities of the studied plants.

Consequently, the morphology of the leaves should be considered more carefully when prioritizing the plant species for improving the air quality, as the interaction of other macro-features, micromorphological traits and environmental factors can outweigh the influence of capturing ability [44,55].

It must be mentioned that the quantities that were captured with these species do not represent the actual PM quantity number as there are two factors (precipitation and air turbulence) that can distract the accumulation process, and in order to determine the total real PM accumulation, the dynamics of this process, precipitation and air turbulence should also be evaluated according to Van et al. [56].

5. Conclusions

Shrubs can capture more particles than the other analyzed species, as the lower height can be an advantage for collecting PM from different emission sources such as air and soil. In this study, the analyzed species captured total of 208.9 µg.cm^{-2} from ambient particulate matter, while most of these particles accumulated on the plant's foliage belonged to the fine fraction size (2.5-10 µm) followed by the coarse fraction (10 -100 µm), and finally the ultra-fine fraction size (0.1 -2.5 µm). Elaeagnus pungens captured the highest deposition of particulate matter among the analyzed species with 83.6 µg.cm^{-2}. Vegetation characteristics such as leaf margin and surface compositional morphology traits as a taxonomic function have been addressed as significant factors associated with different PM

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deposition. This study clearly showed that, roughness, grooves and trichomes (morphological characteristics in shrubs, climbers, and herbs) were substantial factors that helped in capturing particulate matter. In addition to the other structural traits, leaf margin can also affect the accumulation process. Undulate margin in Elaeagnus pungens is a key factor of preventing the particulate matter from falling from the leaf surface through the weather conditions such as air turbulence and precipitation which can interrupt the accumulation process. On other hand, total surface area of the leaves had no direct correlation to PM deposition. As a larger leaf area may contribute to a larger boundary layer (refers to the still air surrounding the leaf) which in theory prevents the particles from settling on the leaf, it may also increase its resistance to the deposition of particles around leaf surfaces which lead to a negative correlation of the PM deposition, on the other hand, particulate matter capturing is effected by the accessible area to the particles due to the leaf surface structures (i.e., surface grooves).

Shrubs, climbers, and herbs, the plants selected for this study, recorded a full deposition for coarse and fine particulate matter as these large particles may be decomposed into fine ones in a process related to attraction and reactivity to deposit on the leaf surfaces via sedimentation. At the same time, the ultra-fine particles can be partially deposited and moved through Brownian motion that prevent particles from settling down under the different weather conditions. In conclusion, climbers' type has the best deposition of coarse particulate matter fraction (10-100 μm), and shrubs type has the best fine (2.5–10 μm) and ultra-fine (0.1 -2.5 μm) fractions deposition, on other hand, herbs type has the lowest portions in all particulate matter quantity in three fractions (coarse, fine and ultra-fine) that had been captured.

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