PARTICULATE MATTER PHYTOREMEDIATION CAPACITY OF JAPANESE ROADSIDE GREEN BIOFILTERS

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PARTICULATE MATTER PHYTOREMEDIATION CAPACITY OF JAPANESE ROADSIDE GREEN BIOFILTERS

日本における沿道のグリーンバイオフィルターの粒子状物質浄化能力

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Interdisciplinary Graduate School of Engineering Sciences (IGSES)
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DEDICATION

To

Father

Mother

Sister and brothers

Friends in Palestine

in recognition of their worth

SUMMARY

The effects of air pollution on public health and environment are ubiquitous. Small sized particulate matter is classified as the most harmful primary pollutant associated with causing convoluted neurological and biological chronic changes in functions of main organs of human body such as, liver, spleen, kidney, the brain, and lower respiratory system through penetrating olfactory nerves and blood stream, moreover, initiate systemic inflammatory vicissitudes which can lead to serious impairments in blood coagulability and exaggerate fatal diseases.

Applied chemical and mechanical remediation techniques are complicated, short term life cycle and insufficient in remote areas. Phytoremediation was already indicated to be an alternative effective plant-based, environmentally friendly biotechnology, involves transferring, removing, detoxifying, degrading, or sequestrating, partially metabolizing, and accumulating air and soil contaminants include particulate matter, through adherence to leaves foliage or absorbing inside roots (plant's rhizosphere). Nevertheless, prioritizing that particulate matter deposition on vegetation foliage differs according to various factors, such as environmental conditions (precipitation and air turbulence), emission source (type, location) and plant characteristics (type, structural characteristics, morphological traits…etc).

Therefore, this dissertation investigates remediation capability of different sizes of ambient particulate matter in polluted areas plants while considering the differences among plant types. The first chapter analyzes particulate matter removal capacity of trees. We focused on particulate matter leaves quantification while investigating the role of leaf traits in enhancing the accumulation process of three size fractions of particulate matter (large, coarse, fine) using electronic and microscopic scanning. The findings confirmed the effective capability of trees due to large leaf surface area (LA) and crowns resulting from complex structure of foliage leaves, thus promotes vertical transport and increases turbulent air movement, as a result enhances capturing substantial quantities of particulate matter. Furthermore, there is a positive correlation among particulate matter accretion and trees with rough surface and hairy small sized leaves with high edged margins than trees with smooth surface and large leaves. On other hand, no considerable influence was observed for other structural traits (leaf size).

Large leaf areas plants may intensify local air pollution values as a result of occupying large spaces, plummeting near-surface air exchange and pollutant dispersal, for that reason, the second chapter incorporated statistical data (meteorological variables) from data sheet sensor with other analytical data such as particulate matter daily concentrations, humidity, temperature, and pressure from May until October 2022 to examine particulate matter capturing ability in diverse types of vegetation in different seasons(spring, summer and autumn). The efficiency of particulate matter accumulation in the selected species enlarges in autumn and summer seasons due to pollution (type, rate) and leaf size. While spring season showed lower deposition on leaves of tested species. In addition to trees, other types of plants, especially low strata of plantings

remediation capacities were inspected in this chapter depending on hypothesis of low height plants may exposed to higher rates of air and soil pollution compared with other species. We found that shrubs are more capable to accrue significant quantities of particulate matter comparing with other types of vegetation species include, trees, climbers and herbaceous plants. Also, climbers, through interacting with other species, may accumulate approximate quantities of particulate matter to deposited particles on leaves of shrubs and trees. Vegetation features such as, leaf shape, type, arrangement, margin and surface compositional morphology traits (roughness, grooves, and trichomes) as taxonomic functions have been referred as key factors correlated with diverse particulate matter depositions on leaves of the analyzed species. To illustrate, lanceolate shape, serrate margin, compound type and opposite arrangement were proved to enhance particulate accumulation process in tested species. Large sized particles (2.5-100μm) are demonstrated to occupy the highest portion of deposited particulate matter in vegetation foliage.

Weather conditions (air turbulence and precipitation may influence deposition of small sized particulate matter via completed resuspension or washing off inorganic components of particulate matter from the surface of the leaves to surrounding soil and degrading organic components, hence, we noticed low deposition of fine particles on leaves of analyzed species.

Location of emission source may affect chemical composition, quantity, and size of the deposited particulate matter, the study found that high quantities of large particles were accumulated on species leaves near natural sources and anthropogenic activities, whereas coarse and fine particles mainly were deposited near transportation emission sources (cars, parking…etc).

Last chapter scrutinized the acute effects of particulate matter on marginalized herbaceous plants in urban areas. Visual assessment was conducted in addition to chemical analysis of PM accumulation on leaves of the selected species. Considerable amount of quantified particulate matter from sampled leaves was observed as simulated potential for air pollutants mitigation closes to some trees. Polluted particles may cause and trigger miscellaneous structural, chemical and biological alterations in stressed plants, despite the ability to apprehend large amount of particulate matter. Although herbaceous plants showed higher tolerance to urban stress through reducing photoinhibition in pervious analytical studies. Results of this study show clear symptoms include necrotic spots, chlorotic patches, loss of chlorophyll pigmentation, surface injury and abscission on the selected leaves of herbaceous species. Moreover, shrinkage in selected leaves surface area was detected comparing with healthy leaves.

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TABLE OF CONTENTS

TABLE OF FIGURES

CHAPTER I: INTRODUCTION

CHAPTER II: PARTICULATE MATTER PHYTOREMEDIATION CAPACITY OF FOUR JAPANESE ROADSIDE GREEN BIOFILTERS

CHAPTER III: PARTICULATE MATTER PHYTOREMEDIATION CAPACITY OF JAPANESE ROADSIDE GREEN BIOFILTERS IN DIFFERENT SEASONS

PART I: PARTICULATE MATTER PHYTOREMEDIATION EFFECTIVENESS OF JAPANESE PRUNUS X YEDOENSIS TREE THROUGH SPRING AND SUMMER SEASON

PART II: PHYTOREMEDIATION EFFECTIVENESS OF MARGINALIZED VEGETATION AS GREEN BIO-FILTER OF PARTICULATE MATTERS IN THE SOUTHWESTERN OF JAPAN (FUKUOKA)

PART III: EFFICIENCY OF EVERGREEN E. PUNGENS LEAVES AS BIOFILTER FOR AMBIENT PARTICULATE MATTER POLLUTION IN SUMMER SEASON

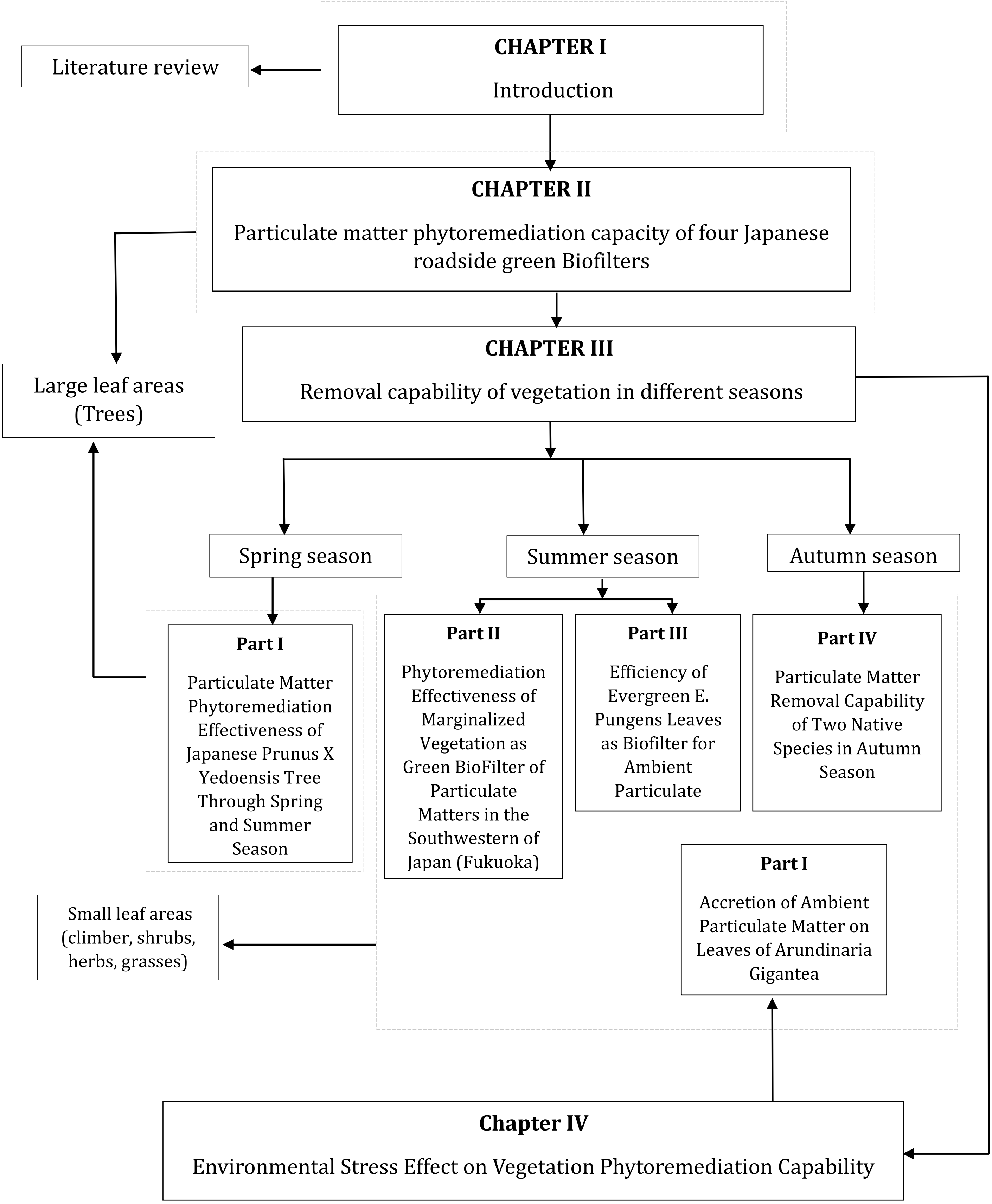
PART IV: PARTICULATE MATTER REMOVAL CAPABILITY OF TWO NATIVE SPECIES IN AUTUMN SEASON

CHAPTER IV: ENVIRONMENTAL STRESS EFFECT ON VEGETATION PHYTOREMEDIATION CAPABILITY

APPENDIX A: ENVIRONMENTAL PARAMETERS IN SAMPLING SITES

LIST OF TABLES

STRUCTURE OF SECTIONS



RESEARCH OBJECTIVES

The role of different types of vegetation in PM phytoremediation has not been widely reported, especially in different seasons. Effects of environmental urban stress on plants remediation capability were not included in similar studies.

Therefore, this thesis aims to

1- Inspect PM phytoremediation capacity in large leaf area species (trees) and smaller leaf areas species (shrubs, climbers, herbaceous and grasses) using PM quantitation dry gravimetric analysis.
2- Investigate the influence of different seasons (Spring, Summer, Autumn) on PM accumulation capability in plants.
3- Use the key environmental parameters that identify surrounding factors ate sampling zones to determine the role of vegetation in degrading ambient PM.
4- Determine the relationship among structural, morphological (leaf traits) and weather factors and enhancing PM phytoremediation in plants.
5- Assess effects of environmental urban stress (PM absorption) on structure and plants PM remediation capacity.

CHAPTER I: INTRODUCTION

INTRODUCTION

Achieving sustainable development goals (SDGs), especially, affordable, and clean energy are considered as essential remarks for green cities which on the opposite side of urbanized cities with ambient air, polluted water, and soil. Intensive using of fossil fuel sources threats affording energy to remote areas, moreover, increasing green house gases rates, thus affecting living parameters, biotopes components (air, soil, water) and the whole system of future cities **[1].**

Long term exposure to air pollutants originated from fossil fuel sources including particulate matter (PM), formaldehyde, volatile organic compounds (VOCs) (benzene, toluene, ethylbenzene, xylene (BTEX), and inorganic pollutants (NOx, SO_2, CO_2, O_3) could aggregates short, medium, and long-term health issues though penetrating human functional systems and disorder the bio processes in different organs, hence affects life quality of 80% of urban population and kills 7 million premature deaths per year globally **[2-9].**

Among addressed air pollutants, Particulate Matter (PM) which consists of liquid and solid aerosol composed of different organic and inorganic compounds such as polyaromatic hydrocarbons (PAHs), sulphides, environmentally persistent free radicals (EPFRs), volatile organic compounds (VOCs), and other substances suspend in the atmosphere, transfer in the ambient air for long time and long distances with an aerodynamic diameter in the range of 0.001–100 μm, is considered as the most inhaled detrimental toxic component to human habitants **[10-12].** PM includes four sizes: Total Suspended Particles (TSP) with a diameter ≤100 μm, large particulate matter (PM_{10}) which is the airborne particulate matter with an aerodynamic diameter ≤10 microns, coarse particulate matter ($PM_{2.5}$) with an aerodynamic diameter equal to or less than 2.5 microns and fine particles (PM_1) with a diameter of ≤1.0μm **[13,14].** The main sources of PM are anthropogenic and natural resources such as fossil fuels, industrial activities, soil erosion, volcanic eruptions and sandstorms with different shapes and chemical compositions **[15-20].** Dramatically, particulate matter concentrations around the world have exceeded WHO standards **(Table.1) [21,22].**

Table I.WHO guidelines for standard particulate matter concentrations

Region	Pollutant	Averaging Period Max	WHO Guidelines
International	PM10	1 day	50 μg/m³
		1 year	20 μg/m³
	PM2.5	1 day	25 μg/m³
		1 year	10 μg/m³
Japan	PM	1day /hour	0.10 mg/m³, 0.20 mg/m³

Toxicity of PM is directly correlated with different types of potentially fatal diseases affects all segments of the society, especially, children and pregnant women which could suffer from post-neonatal infant mortality, sudden infant death syndrome (SIDS) **[23-25].** Furthermore, larger size of PM is associated with noncommunicable diseases (NCDs), cardiovascular and respiratory diseases that may contributed to the so-called 'sick building syndrome' (SBS) and 'building-related illnesses'(BRI). On other hand, fine size particles (PM_1), can reach major organs of human body such as, liver, spleen, kidney, the brain, and lower respiratory system through the olfactory nerves and alter alveolar macrophage functions, moreover, cause systemic inflammatory changes which can lead to serious impediments in blood coagulability **[26-29].**

Although, the concentration of PM air pollution from the Asian continent during the winter and spring, particularly in regions of western Japan is gradually declining, because of strict daily policies and measurements of PM in disrupted air pollution stations along Japan regions (Figure I) **[30-35].** Concentrations of PM $_{2.5}$ are recording high values exceed environmental quality Japanese standards for 24 h of 35 μg.m^{-3} **[36].**

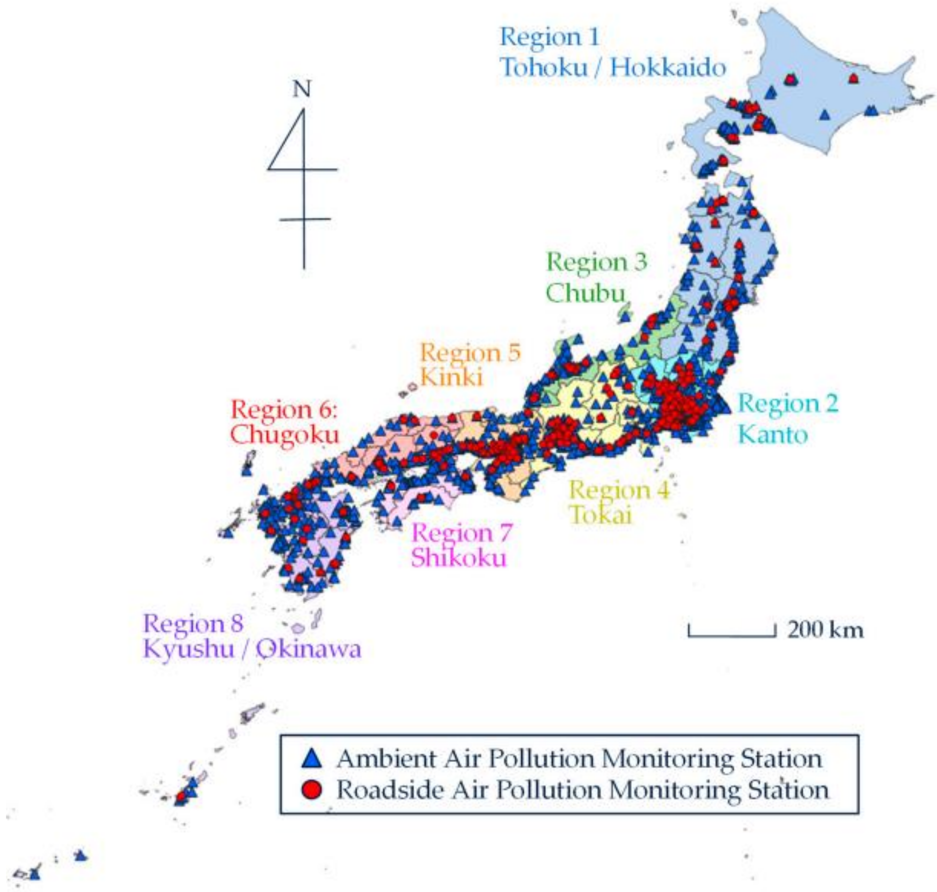


Figure I. Distribution of ambient air quality monitoring stations (AAQMS) and roadside air quality monitoring stations (RsAQMS) in Japan **[37]**

Long term exposure to PM components such as toxic components include heavy metals such as Se, Mo, Pb, As, Zn, W, Sb, Cu, V, Cr, Ni, and Cs found to be correlated with nasal, ocular, skin symptoms and severe non-carcinogenic risk to moderate carcinogenic risks for healthy school children in Fukuoka **[38].** Moreover, **Takeda et al. [39]** confirmed that sulfate is associated with diminishing placental weight in the third trimester with a difference of -6.7g from the normal birth weight. While short term exposure to large and coarse PM was related to non-accidental mortality with 1.3% (95% confidence interval (CI),0.9-1.6%), in total, on other hand, an increase in cause specific mortality with 1.4% (95% CI, 0.2-2.6%) with 10 μg.m^{-3} increase in coarse PM **[40].** In addition to health effects, acute environmental issues can be induced by PM, as the ambient aerosol can absorb and scatter the solar radiation, influences the cloud condensation, nuclei activity, and engenders oxidative stress **[41, 42].**

Consequently, various standards, policies, chemical and conventional mechanical techniques were authorised and applied to eliminate air pollutants. Removal of particle matter is applied to two forms of deposition, dry and occult (wet) deposition **[43].** Dry deposition occurs according to particles size, for instance, large size particles can be deposited through gravitational settling, on other hand, smaller size particles (coarse and fine) may apply to impaction, interception, or diffusion. Whereas wet deposition is the removal of air pollutants using natural mechanisms such as precipitation **[44].**

Nevertheless, complexity and costly continuous maintenance have addressed these techniques as short term and insufficient. Hence, there is a growing prerequisite for obtaining more cost-effective, simple, and environmentally friendly mechanisms of particulate matter pollution monitoring and remediation such as phytoremediation which has been shown to be effective plant-based and cost-efficient biotechnology in purifying ambient air via degrading, detoxifying, or sequestrating and partially metabolizing the pollutants inside aboveground and underground plant parts, leaves and roots **[45-47].**

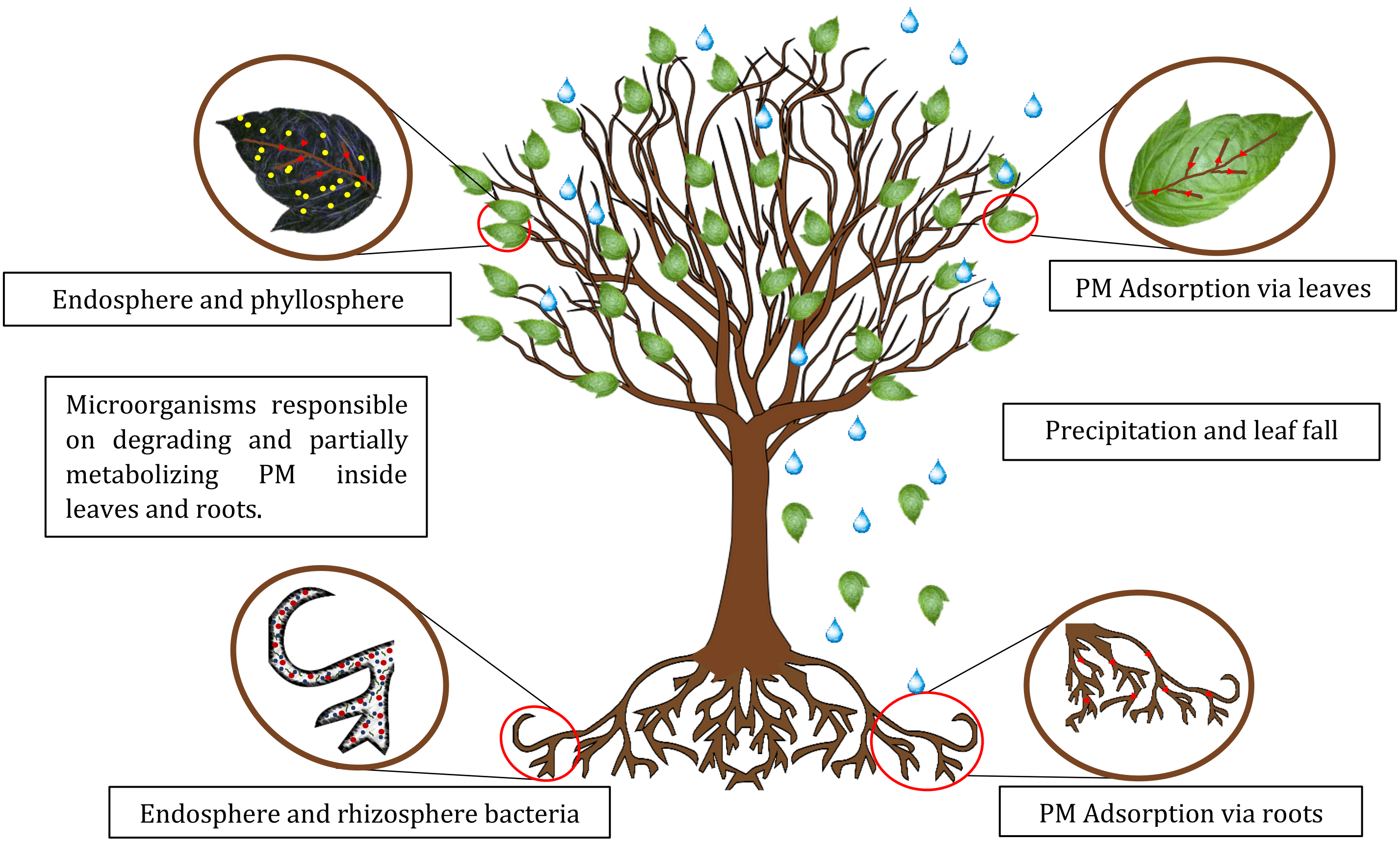


Figure II. Systematic mechanism of phytoremediation in plants **[47]**

The mechanism of phytoremediation happens when leaves surfaces and stems adsorb significant amounts of pollutants. Part of the adsorbed pollution penetrate to leaves tissues to come cross with phyllospheric and endophytic bacteria that can detoxify part of the pollutants by means of degradation, transformation or sequestration. Further, rainfall causes flowing down of the pollutants to the soil right below the plant, where the pollutants come into contact with the soil, the plant's rhizosphere and the roots (Figure II) **[47].**

Generally, plants tend to enhance the quality of microclimate, biodiversity and achieve climate adaptation through eradicating negative effects of global warming in miscellaneous ways through acting as a broader in front of sun radiation during summer, and releasing humidity to the air via evapotranspiration, thus decreasing high temperature inside/outside buildings (heat island effect**) [48],** furthermore, enhancing building energy reserves **[49-51],** controlling water run-off **[52, 53],** reducing air turbulence speed to compensate heat loss in winter and improving urban air quality **[47,54].**

Empirical studies have demonstrated removal capacity of trees, shrubs, herbaceous plants, climbers, and grasses such as **Sgrigna et al. [5], Ottelé et al. [55], Sæbø et al. [56], Dzierzanowski et al.[57], Przybysz et al.[58]. Dzierzanowski et al. [57]** proved purifying ability of 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.). **Sæbø et al. [56],** observed differences among species due to leaf traits in Particulate matter (PM) accretion on leaves of 22 trees and 25 shrubs in test fields in Norway and Poland. While **Przybysz et al. [58]** analyzed capacity of evergreen species (Taxus baccata L., Hedera helix L. and Pinus sylvestris L.) to accumulate particulate matter and trace elements from ambient air at three sites with different pollution levels and exposure to precipitation. Five species (Pitosporum tobira, Lavandula angustifolia, Lampranthus spectabillis, Sedum album, and Sedum reflexum) for GRs and four species (Aptenia cordiflora, Erigeron karvinskianus, Sedum palmeri, and Sedum spurium p.) for LWs were examined in **Viecco et al., [59]** study to quantify the dry deposition of $PM_{2.5}$ and PM_{10} on vegetation leaves as well as PM captured by the leaf wax. On other hand, **Ottelé et al., [55]** quantified the deposition of particulate matter on the underside and upper side of the leaves of climber vegetation. Similar results were reported by **Sgrigna et al. [5] and Weerakkody et al., [60]** who studied other species. The majority of the analyzed plants species in the empirical studies **(i.e. Sæbø et al. [56] and Dzierżanowski et al. [57])** can easily capture larger PM particles, in contrast, **Freer-Smith et al. [61]** demonstrated higher accumulation of fine PM compared with fine and coarse PM of the analyzed species.

There are significant differences among plant types in PM removal capacity. Evergreen species which keep their leaves throughout the whole year, resulting with capturing more particles than deciduous species **[58, 62].** Furthermore, trees were demonstrated to be the most effective plant group in PM scavenging due to their large surface areas and crowns that endorse vertical transport of air by enhancing turbulence and increase particles gravitational sedimentation, impaction, direct interception, and Brownian motion **[44,56,63]** However, there is hypothesis that very large and too dense roadside trees in urban areas can amplify local air pollution rates as a result of reducing near-surface air exchange and pollutant dispersal **[64-67].**

Due to limited space in urban areas and roadsides, low strata planting which can accumulate pollutants and diminish the recirculation and residence times of pollutants could be placed as sufficient alternative to highly dense trees **[66-68].** Also many studies **(i.e. Stapleton and Ruiz-Rudolph,[69], Perini et al., [70])** proved the efficient removal capacity of shrubs and climbers which collected more PM quantities than trees in **Dzierzanowski et al. [57]** study, the study concluded that these types of plants are more exposed to different types of pollutants due to their lower height (low strata planting) that allows to interact with soil and capture soil splash besides air pollutants**.** Additionally, shrubs may tend to display lower wash-off rates compared with trees owing to their waxy hydrophobic leaves during natural precipitation events **[71].**

While climbers, which can reduce building temperature up to 20 ℃ through absorbing 60% of solar radiation and improve weather conditions and decrease air temperature by 6.1 ℃ in summer and by 4 ℃ in winter and thereby reducing energy for air conditioning between 25% and 80% **[72,73].** Through expanding the particulate capturing area, as a result of large surface area, green walls (GW) reduced $PM_{2.5}$ rates by 74.1% rendering to **Viecco et al., [59],** and PM_{10} to 23% **[74].** Furthermore, climbers were demonstrated to be more effective at accumulating smaller size of PM, especially fine size particles which are considered as the most harmful size among PM components **[58, 60].**

Urban herbaceous species have considerable ecological effects, include carbon confiscation, pollinator support, noise diminishing, flood and erosion risk management, local climate regulation and air quality improvement **[75-78].** Despite their low leaf surface area, some herbaceous species also showed significant removal capacity compared with trees attributable to their adjacent location to emission source and their ability to accumulate PM during the growing season, despite ongoing drought **[79,80]. Weber et al. [81]** who verified PM removal capability of 16 species of herbaceous plants (grasses and forbs) according to differences of site-specific traffic densities, plant species and morphological types, mentioned that the analyzed species showed substantial ability of accumulating PM. Unexpectedly, Festuca rubra (herbaceous species) showed a capacity for air pollution remediation closes to trees in **Speak et al., [82]** study**.** Nonetheless, **Currie and Bass [83]** demonstrated grass species as the least efficient group among plant species compared to trees and shrubs, moreover, **Dochinger [84]** found that PM deposition of deciduous species without leaves (as in late autumn and winter) are equal to the accumulated PM on leaves of grasslands representing their poor removal capacity. Also, variation in PM deposition across size fractions was found in various studies **(i.e. Slin [85]; Weerakkody et al. [86]).** The reason may related to different deposition velocities and processes i.e. interception, impaction and sedimentation under gravity as consequence of different aerodynamic diameter of the deposited particles **[60].** Comparing among herbaceous plants and grasses, **Speak et al. [82]** observed that grasses are more effective than herbaceous, whereas, **Weber et al. [81]** did not find any substantial variances between these two groups of plants.

Despite uncertainty surrounding the effect of individual leaf traits on PM capturing due to different conclusions from previous studies **[86].** Various studies consider leaf functional traits as reliable markers of environmental stress that can also influence PM accumulation in plants species. **Sgrigna et al. [87]** addressed 9 factors includes physiological and morphological characteristics of species leaf (roughness, pore density, hair, leaf shape, time of leaf growth and leaf size) that are combined and interacted with each other. While **Kováts et al., [88]** addressed additional sensitive traits such as non-enzymatic and enzymatic antioxidants, leaf water level, leaf ultra-structure, photosynthetic pigments and nutritional rate. Furthermore, **Nowak et al. [46], Beckett et al. [89] and Leonard et al. [90]** included air pollution concentration, environmental conditions (air turbulence and precipitation), species structure as additional factors affecting PM capturing process. **Dzierzanowski et al. [57] and Beckett et al. [89]** observed higher capacity of evergreen coniferous species (most of these species keep their leaves for several years), in comparison to broad leaved species for PM accumulation as small, rich epicuticular wax, narrow stems needles with complex shoot structure which have higher tendency to bend effortlessly with wind flow due to their narrow bases or petioles are more easily exposed to particles in the air than large and flat leaves, which have thicker and wider boundary layers **[56, 61,71, 86, 91].** Moreover, waxy leaves surfaces of needle shaped species are advantage to prevent PM washing off through precipitation events, as a result, there is higher possibility for broad-leaved species to be more exposed to PM washing off, especially smaller size particles **[71].** However, some studies, recommended that planting needles shaped leaves are not useful comparing with deciduous leafy species due to reduced tolerance of conifers to high traffic-related pollution which accumulate in their leaves for several years and thus cause physiological damage to the biological mechanisms inside plant that can lead to their collapsing due to sensitivity to HM resulted from greater quantity of PM according to **Dzierżanowski et al. [57], Godzik et al., [92], Lamppu and Huttunen [93] and Gawroński et al. [94]**. In term of leaf size, higher PM deposition rates were observed for smaller sized leaves with larger edges due to elevated resistance of smaller leaves to move with air turbulence, and thus resuspend accumulated PM compared with larger size leaves that have larger boundary (surrounding air) layer which influence accumulating process negatively as larger boundary layer is correlated with increasing the resistance to deposit PM**[60,61,86,90].** On contrary, **Vogel, [95]** found that leaves with larger leaf surface areas and increased petiole length have bigger chance to accumulate more particles. Other leaf characteristics such as leaf shape, margin and arrangement, previous studies have indicated that lanceolate leaves and whorled leaf arrangement demonstrated more effective PM accumulation than other leaf characteristics **[89].** On contrary, **Weerakkody et al. [60]** suggested that species that have simple leaf arrangement with larger gaps between their leaves may accumulate lower PM deposition due to lower turbulence surrounding the leaf.

Generally, leaf margin and main nerves were recognized as key markers of enhancing PM accumulation in plants species according to **Mitchell et al. [96]**, furthermore, various types of leaf margin could affect PM capturing process differently, for instance, toothed edges(serrated margin) can prevent PM washing off or resuspention, while lobed and entire margins could be considered as limitations of accumulating high quantity of PM. **Leonard et al., [90]** demonstrated that position of leaf margin comparing with main vein can affect PM accumulation process, if leaf margins were closer to the main vein, this will cause fluttering of the leaves, thus reducing the opportunity to capture PM.

Morphological characteristics, such as surface roughness and presence of trichomes and hair, have also been associated with positive PM accumulation on the foliage of the analyzed species **[89, 90,97,98].** Leaf roughness can change the transition from laminar to turbulent airflow which upsurges the deposition velocity of PM comparing to smooth leaf surface. Leaf hair can prevent the resuspension of accumulated PM, through increasing the leaf surface area for the adherence of particles, moreover, the structure of leaf hairs can also change PM chemical deposition, as some leaf hairs displaying hydrophobicity which can attract charged particles and toxic components such as heavy metals **[99, 100].** In contrast, **Perini et al. [70]** found a negative correlation among PM capturing process and leaf hairs presence.

Research on association among emission source location and plant removal capacity is limited to a few studies such as **Ottelé et al., [55] and Muñoz et al. [101]** who noticed that vegetation can be recognized as particulate matter scavenger if it is located closely by an emission source and traffic flow as the quantity of deposited PM on plants leaves decreases dramatically with increasing distance from the emission source **[102, 103]**. If the selected species is too distant (for example, 60 m) from the emission source, the efficiency of PM_{10} (larger size PM) removal become undetectable **[80].** Other researchers considered that distance more than 20 m cannot be involved in investigating studies of suitable plant types of PM removal. According to **Karner et al. [104]** the PM rates in the ambient air rapidly decreases within a short distance from the street. Type of emission sources can also affect size and chemical deposition of PM, as natural sources and anthropogenic activities can produce course particles. While fine particles can be originated from vehicle emissions (gasoline and diesel), combustion, and industrial processes. The most harmful size particles (fine particles) originate mainly from transport and photochemical reactions in the atmosphere **[105].**

The dynamics of PM deposition are greatly influenced by weather and climate factors (air turbulence and precipitation) **[89, 97, 106, 107].** After deposition, precipitation may remove PM from the surface of the leaves and decompose the organic components of PM, whereas the inorganic components are washed off to the soil surroundings the subjected plants **[57]. Przybysz et al. [58]** found that precipitation may remove a considerable proportion of particles accumulated on foliage. The removed quantity of PM can reach 20% of total PM according to **Kaupp et al. [108].** In particular, coarse and fine particles which are the most affected fractions by weather conditions. Also, it can affect the chemical composition of the washed off PM. Some toxic components such as Heavy metals (HM) and polycyclic aromatic hydrocarbons (PAH) were found in the analyzed samples of fine PM removed from leaves after precipitation according to **Jouraeva et al. [97].** On other hand, **Freer-Smith et al. [61] and Kwak et al., [71]** noticed that precipitation has no considerable effect on the accumulated PM on the foliage of plants species. Air turbulence has also significant effect, as wind is associated with resuspension of large portion of the deposited PM. According to **Slinn [85] and Petroff et al. [109]**, the amplified turbulence in leaf boundary layer surrounding the deposition surface may cause turbulent transfer and resuspension of smaller PM size fractions. This can explain the insufficient removal capacity of fine particles in most of the tested plants species in the pervious studies. However, **Ould-Dada and Baghini [110]** indicated that re-suspension of total PM deposition can happen with speed more than 5 m s^{-1}. In coastal areas with stronger wind, substantial re-suspension may occur to the deposited PM **[58].** Nevertheless, some leaf traits may act as protection from weather conditions, for instance, leaf surface roughness and hair presence can create different contact angles (θ) between the rain droplet and leaf surface, which increase the contact time and delay PM washing off and resuspension **Przybysz et al., [58], Fowler et al. [111], Vadlamani et**

al.[112] and Pikridas et al., [113] found that rainy months in winter recorded higher deposition than other seasons. In winter, pollutants concentration increases intensely due to high using of fossil fuels for heating and warming purposes and when air exchange is limited. Opposed to **Nakata, [114]** that showed higher PM concentrations during spring and summer, because of dust events in spring and photochemical reactions in summer compared with winter season.

Although plants can absorb particulate matter effectively, environmental stress induced by particulate matter has severe effects on roadside plants. In general, urban plants are constantly imperiled to anthropo-pressure, soil compaction, inadequate sun energy, limited spaces, augmented temperature, resulted from pollution and global warming **[115].** The harmful effects of PM on vegetation have already been observed by numerous research **[116-122].** Stressed plants show visual, morphological, and biological symptoms include degradation of structure and composition of the surface wax layer following exhaust gas exposure due to the effect of lipophilic aromatic hydrocarbons **[123].** Different sizes of particles could decrease specific leaf area (SLA) **[121]**, increase leaf senescence **[124]**, decline the efficiency of photosynthesis through blocking leaves stomata gaps thus affecting their optical functions by absorption/reflection of PAR (photosynthetically active radiation). According to **Przybysz et al. [58]**, a negative association was found amongst photosynthesis rate and the deposited PM. Furthermore, disrupt gas exchange among tissues **[125]**, upsurge the leaf temperatures **[126],** generate less reducing power and causing leaf senescence and abscission which leads to decrease chlorophyll content (TChl) in plants system and cause chlorosis (yellow and brown patches), affect the specific activity of catalase or peroxidase such as glutamate dehydrogenase, glucose-6-phosphate dehydrogenase , isocitrate dehydrogenase, alanine aminotransferase, asparagine aminotransferase and peroxide dismutase, moreover decrease protein and sugar content which are major factors in plant life cycle **[58,103, 127-130].** Decreased levels of chlorophyll a and b was already stated by **Rabe and Kreeb [127].** Furthermore, toxic components such as nitrate, sulfate, organic pollutants, and heavy metals may cause physical and chemical alterations in plants tissues which is called necrotic spots (colorless spots) **[131].** PAHs and trace elements may be involved in increasing reactive oxygen species and trigger intracellular oxidative stress **[88].** Some particles with pH values of ≥ 9 can change the soil PH which may trigger direct injury to leaf tissues **[132].** Shrinking in plant leaf size and stomatal density enlargement could be occurred in high PM rates areas **[98,133].** While plants with higher SLA could be acclimatized to the urban environmental stress **[87].** According to **Pal et al. [134]** two-fold escalation in stomatal frequency and trichome length in the analyzed samples from high-traffic sites compared with samples from low-traffic sites. PM pollutants triggers morphoanatomical changes in plants, including, leaf stomatal thickening, biomass dilapidation and fluctuation in chemical composition of waxes, as a result of its effect on leaf extract (pH), plant relative water content (RWC), microbiological parameters **[10, 58, 97, 131, 132,135-140].** Moreover, acute symptoms might be advanced due to synergistic effects or exacerbated to secondary stresses including pathogen stress and drought stress and thereby cause degradation of plant species **[141, 142].** Especially, low height and small plants that may exposed to higher values of PM deposition compared with other species resulting with species-specific changes in growth and phenology, with a dramatic rate of accelerated senescence, reduction in fruit production and delayed flowering according to **Bell et al. [11], Honour et al. [143] and Rai et**

al. [144]. Besides, toxic effect of PM on plants, living parameters can also be influenced. PM may diminution the endurance rate of insects **[145],** terrestrial biomass evolution and eradicates oxidative stress, leaf and root bacteria **[146].** Nanoparticles of particulate matter may affect human through consuming stressed plants leaves which is more affected part of plants system **[147].** Nonetheless, most of plants in the previous studies (i.e. **Armbrust et al., [106], Heerden van et al., [107], Vardaka et al., [132])** showed PM-induced oxidative stress symptoms, some plant species such as Ilex rotunda trees **[128]** and Sorbaria sorbifolia **[58],** showed higher photosynthetic rate despite their presence in more polluted urban areas. The reason may related to protective role of PM displaying by reducing photoinhibition. It worth mentioning that though many changes in stressed plants are generally related to ambient air pollutants deposition, concentration-effect associations are still inadequately known **[148].**

CHAPTER II

Particulate Matter Phytoremediation Capacity of Four Japanese Roadside Green Biofilters

This chapter has been originally published in Environment and Sustainable Development, 2022

ABSTRACT

Particulate matter (PM) accumulation on leaves of two broad leaves trees and two shrub trees were examined for 14 days to study the ability of broad leaves to capture particulate matter in Japan.

Two healthy mature leaf samples of each specimen were carefully collected and analyzed through the gravimetric analysis method (four filtration steps). PM in different divisions can be captured and deposited inside the leaf foliage. Fine particles were the highest portion of the PM content captured by the analyzed trees.

Leaf features such as hair and wax have been associated with high PM accumulation as presented in Toxicodendron succedaneum (waxy glossy leaves) which was the best shrub among the four species for all PM fraction divisions (PM_{10}, $PM_{2.5}$, and PM_1), and Prunus × yedoensis (hairy leaves) that captured the largest portion of fine PM. The surface area has no effect on the accumulation of particulate matter since the Ficus erecta shrub has the largest surface area, but the lowest portions of PM among of investigated species.

Keywords: Remediation, leaves, Particulate matter

INTRODUCTION

Air pollution threatens the health of people in numerous cities of the world. WHO revealed that 9 out of 10 people breathe air containing high levels of pollutants which can kill 7 million deaths globally per year with noncommunicable diseases (NCDs), causing approximately one-quarter (24%) of all adult deaths from heart disease, (25%) from stroke, (43%) from chronic obstructive pulmonary disease, and (29%) from lung cancer **[1], [2].**

The primary sources of air pollution are mobile sources, stationary sources, and natural sources, which contain several toxic compounds, including intense organic compounds, toxic gases, and trace elements (TE) which are considered the most dangerous component, particulate matter (PM) **[3].**

Particulate matter (PM) is the heterogeneous solid or liquid atmospheric aerosol suspended in the air for long periods and transported over long distances. PM includes four types: the total suspended particles (TSP) with a diameter ≤100 μm, large particulate matter (PM_{10}) which is the airborne particulate matter with an aerodynamic diameter ≤10 microns, coarse particulate matter ($PM_{2.5}$) which is the inhalable particulate matter with an aerodynamic diameter equal to or less than 2.5 microns and fine particulate matter (PM_1) with a diameter of ≤1.0μm which also recognized as the most harmful fraction **[4-8].**

Different sizes of PM have variable effects on human health **[9-14].** For instance, in Japan, $PM_{2.5}$ which has a high concentration exceeds the environmental quality Japanese standards for 24 h of 35 $\mu g.m^{-3}$ **[2].**

Moreover containing 29 heavy metals such as Se, Mo, Pb, As, Zn, W, Sb, Cu, V, Cr, Ni, and Cs can be correlated with nasal, ocular, skin symptoms and severe non-carcinogenic risk to moderate carcinogenic risks for healthy school children **[15], [16].**

Many protection policies and actions are focused on the reduction of particulate matter, especially near busy roads in urban areas, but the removal of particulate matter and air pollutants, in general, is very complicated. Vegetation can be used as green biofilters, and bio-monitors of anthropogenic pollutants, such as heavy metals (HM) in the urban environment **[17].**

Many studies investigated the efficiency of PM capturing in plants, for example, Sæbø et al. **[18]** proved the ability of 22 trees and 25 shrubs stomata to accumulate particulate matter inside their foliage **.**

This work aims to study the accumulation ability of PM in 4 types of Japanese trees that are un fully studied in previous studies and investigate the role of the leaf features and leaf surface area in enhancing PM accumulation.

MATERIALS & METHODS

Plant types and experimental sites

Four broad leaf types of trees and shrubs were included in this study, camphor tree (Cinnamomum camphora), Yoshino cherry (Prunus × yedoensis), Japanese fig (Ficus erecta Thunb), and Japanese Hazenoki tree (Toxicodendron succedaneum) (more information shown in table I) which were located in two nearby sites near parking and busy road (33°31'24" N 130°28'40"E, 33°31'25"N 130°28'43'E and 33°31'21"N 130°28'44'E) in Kyushu university campus (Fukuoka, Japan) (shown in Figure I) with a distance between road and trees ranges from 2 to 3 m.

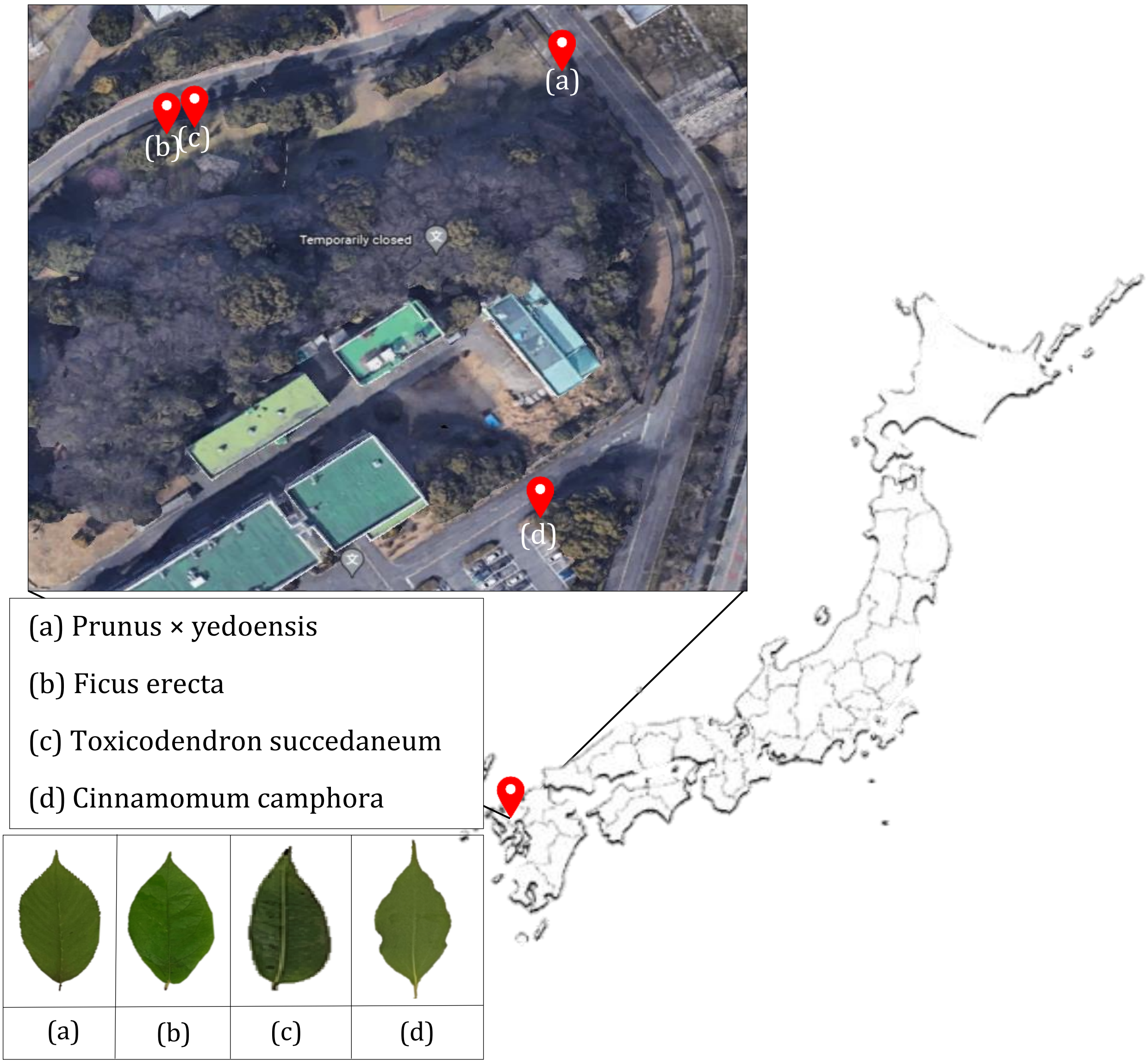


Figure I. Distribution of sampling sites of the analyzed species [19]

The weather conditions during those two weeks were standard for the spring season in Fukuoka: warm, and sunny with low rainfall, and the monthly temperature ranged from 16.8-24.9 °C. Fukuoka WMO on-site station (ID:47807) recorded monthly total precipitation in May 2022 with 45 mm in this location, according to Japan Meteorological Agency **[20].**

Rates of PM pollution in the sampling site was high (see Appendix A). Therefore, two young mature healthy leaf samples were collected from every specimen with a time gap of two weeks, when there was no previous rainfall for more than five days.

Sampling height varied from 1.5 to 1.7 m above ground level depending on plant structure. All leaf samples were placed in plastic bags, sealed, labeled with numbers, and stored at 4 °C in a clean laboratory refrigerator prior to analysis.

Table I. Plant species were used in this experiment to assess the capturing efficiency, (North Carolina Extension Gardener Plant Toolbox)**[21]**

No.	Botanical name	Common name	Plant type	Leaf characteristics	Image
(1)	Cinnamomum camphora	Camphor tree	Evergreen	Leaf type: simple Leaf shape: ovate, obovate Leaf margin: entire Leaf surface: glossy and leathery Leaf arrangement: alternate Leaf length: 3-6 inches Leaf width:1-3 inches	
(2)	Prunus × yedoensis	Yoshino cherry	Deciduous	Leaf type: simple Leaf shape: oval, veined Leaf margin: serrated Leaf surface: leathery hairy Leaf arrangement: Alternate Leaf length: 3-6 inches Leaf width: 1-3 inches	
(3)	Ficus erecta	Japanese fig	Deciduous	Leaf type: simple Leaf shape: broadly ovate Leaf margin: entire Leaf surface: smooth hairless Leaf arrangement: alternate Leaf length:3-7 inches Leaf width: 1-3 inches	
(4)	Toxicodendron succedaneum	Japanese Hazenoki tree	Deciduous	Leaf type: Compound Leaf shape: elliptical Leaf margin: entire Leaf surface: rough, hairless Leaf arrangement: alternate Length: 1.5-4 inches Width:0.5-1 inches	

Quantitative Gravimetric analysis of PM

The quantity of PM was analyzed using Dzierżanowski et al. [22] method (Table II) which divided PM analysis into two categories: (i) PM content from water washing and (ii) PM content from chloroform washing .

In this study, Sodium dodecyl sulfate (SDS)($CH_3(CH_2)11OSO_3Na$) (Merck, Germany) with a 5% concentration (more than 5% concentration can produce more bubbles and impedes the filtration process as the bubbles can occupy the filter holes instead of particles) was used instead of chloroform to achieve the best extraction of PM and deep washing of plants stomata as it is a strong anionic surfactant.

Thus, the leaves of plants were washed with 250 mL of distilled water for 60 seconds and after with 150 ml of SDS for 60 seconds, thereafter the washing solutions from both steps were applied to 4 sieving and filtration tools through (i) polypropylene sieve (retention 106 µm, Itoh seisakusho Manufacturing, Japan) then (ii) through a 10 µm Omni pore membrane filter (Merck millipore Ltd, Ireland) (iii) through a 2.7 µm glass microfiber filter (Whatman filter, Cytiva, Japan) then using (iv) a 0.2 µm PTFE membrane filter (Merck Millipore Ltd, Ireland), the filtration was conducted using a 25 mm vacuum filter holder with stopper support assembly connected to a vacuum pump to have three fractions of PM: (i) 10–100 (large), (ii) 2.5–10 (coarse) and (iii) 0.2–2.5µm (fine).

The filters were dried before and after filtration for 30 min at 60 °C, stabilized in the weighing room for 30min, and weighed using (electronic balance HT124R, ViBRA, Japan). The total leaf area of plant leaves was calibrated and measured using ImageJ software (Rasband, W.S., ImageJ, U. S. National Institutes of Health, Bethesda, Maryland, USA, http://imagej.nih.gov/ij/, 997–2012).

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) and a scanning electron microscope (SEM) (JSM-7900F, Japan Electronics Co., Ltd., Japan) to test the relationships between leaf traits in each selected species and PM accumulation.

Table II. Condition table of quantitative gravimetric analysis of PM

Parameter	Condition /type
Water washing phase	250mL, DW, 60 seconds
SDS washing phase	150mL, SDS, 60 seconds
Filtration phase	100µm sieve, 10µm filter, 2.5µm filter, 0.1µm filter, vacuum filter holder, pump
Flow rate	4 ml/min
Run time	90 min/sample
Weighting room temperature	Conditioned room/ 23°C
Oven temperature	60°C
Weighting unit	mg

CALCULATIONS

The particulate matter quantity was measured through this equation **[7],**

$$PM\ Deposition\ (\mu g.\ cm^{-2}) = Wf - Wi / A \qquad (1)$$

W_f: Weight of filters after filtration in µg.

W_i: Weight of filters before filtration in µg.

A: Total surface area of leaf in cm^2.

RESULTS

Total PM accumulation differed considerably between species and between periods of time, but the four species showed a good ability to capture PM. Most species presented an increase in PM deposition throughout the experiment period (14 days). In total, coarse PM fraction (2.5 -10 µm) recorded the highest portion, whereas fine fraction (0.1-2.5 µm) is the lowest (Figure II).

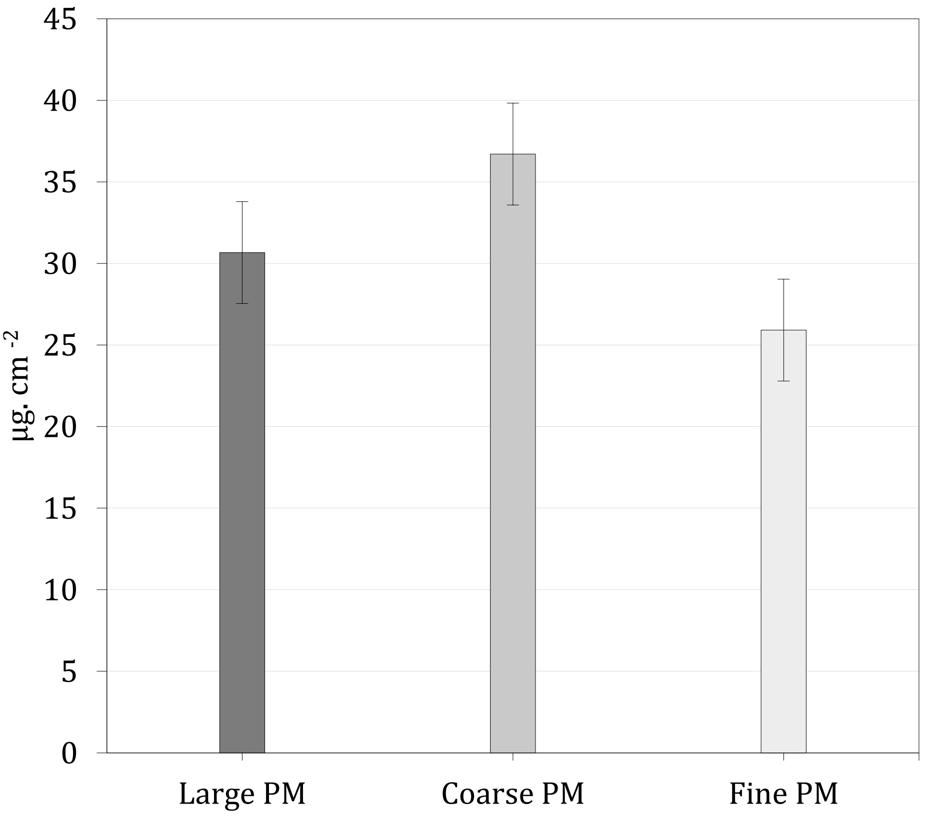


Figure II. Total PM deposition of the analyzed species, mean±SE

Among the species analyzed, Toxicodendron succedaneum (wax tree) had the highest PM deposition between 15 $\mu g.cm^{-2}$ to 26.4 $\mu g.cm^{-2}$, respectively, in terms of the different sampling times (Figure III). While the Ficus erecta which has the largest surface area (large boundary layer) among the four species (37-98 cm^2) showed the lowest PM deposition (6.68 $\mu g.cm^{-2}$) for the first sample (blank sample) and (5.9 $\mu g.cm^{-2}$) for the second sample (after 14 days sample).

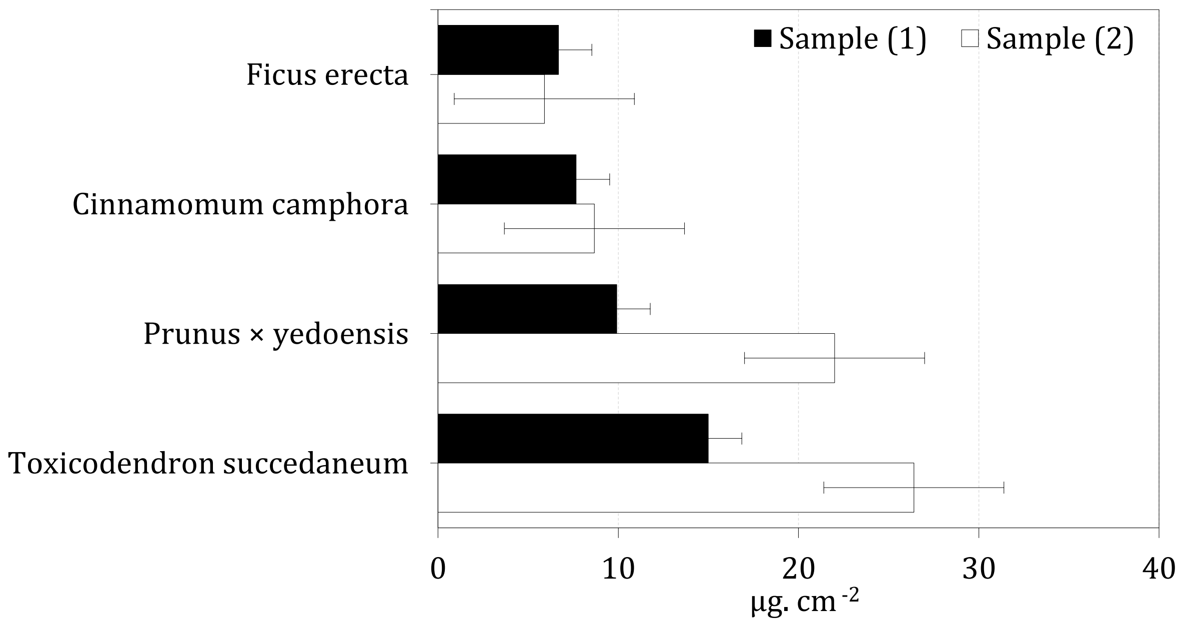


Figure III. Total PM samples deposition of the analyzed species, mean ±SE

In Toxicodendron succedaneum species, large fraction (10 µm -100 µm) made up the greatest proportion of accumulated PM mass and fine fraction (0.1 µm-2.5 µm) made up the smallest proportion, it is worth mentioning that the coarse fraction (2.5 µm-10 µm) the showed a higher portion more than fine fraction. Expressed as a percentage, the PM fractions as 40.3%, 33.3%, and 26.3% for large, coarse and fine, respectively (Figure IV).

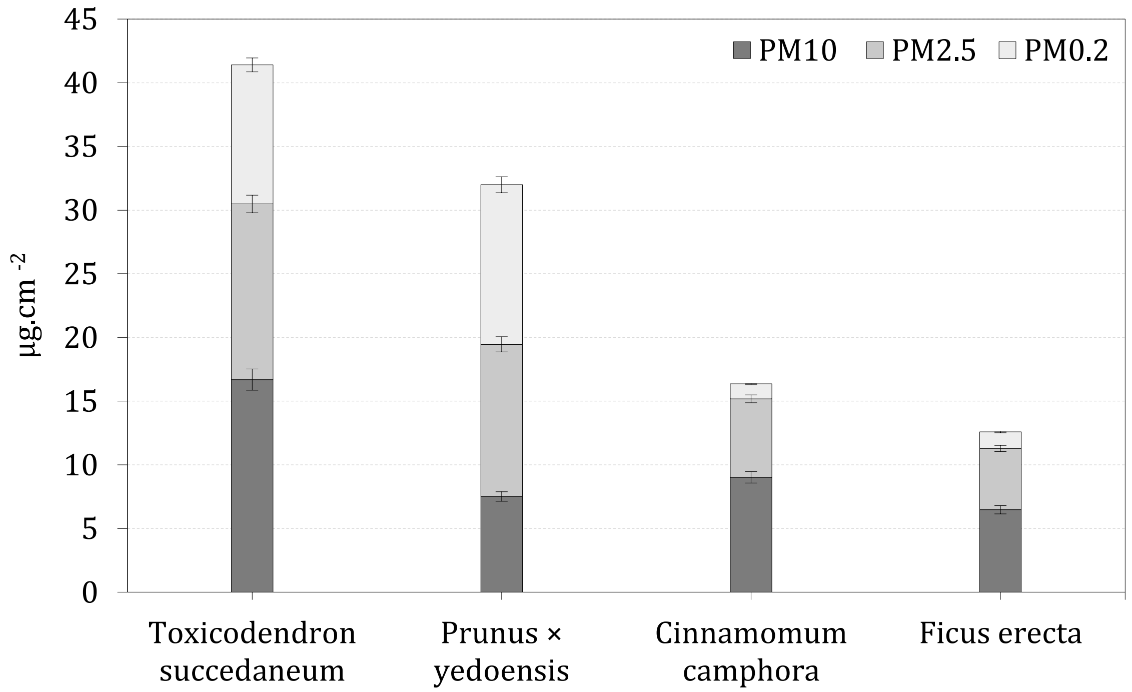


Figure IV. Difference in PM deposition in three fractions among the analyzed species, mean ±SE

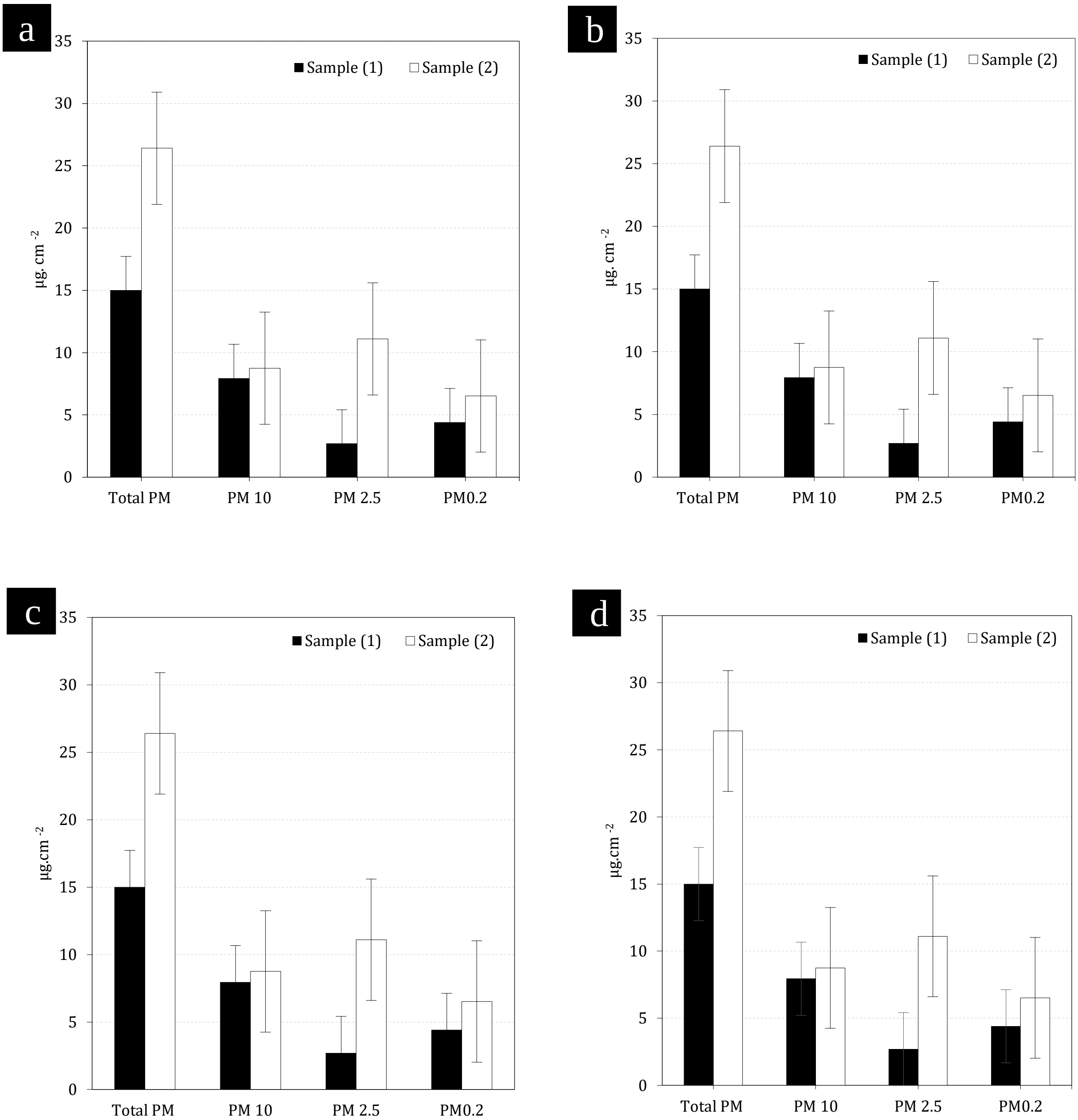


Figure V. PM deposition (samples, fractions) in the four species, (a) Toxicodendron succedaneum (b) Prunus × yedoensis, (c)Cinnamomum camphora , (d) Ficus erecta

Prunus × yedoensis had the highest fine fraction (0.1 µm-2.5 µm) with 48.3% among the four species, then Toxicodendron succedaneum species with 42.1%, followed by Ficus erecta with 5 % and finally Cinnamomum camphora with 4.5%. For coarse fraction (2.5 µm-10 µm), the highest portion is for Toxicodendron succedaneum with 13.79 µg.cm^{-2}, followed by Prunus × yedoensis which presented clusters of fine particles in a cross-section of 2.5 µm filter membrane as a high content with 11.95 µg.cm^{-2}, then Cinnamomum camphora with 6.16 µg.cm^{-2}, and the last for Ficus erecta with 4.81 µg.cm^{-2}(Figure V and VI).

Toxicodendron succedaneum is the smallest leaf among the four species with a surface area of (2-23) cm^2 had the largest portion for large fraction (10 µm-100 µm) followed by Cinnamomum camphora which has the second small surface area with 8-21 cm^2, then Cinnamomum camphora and Ficus erecta, respectively.

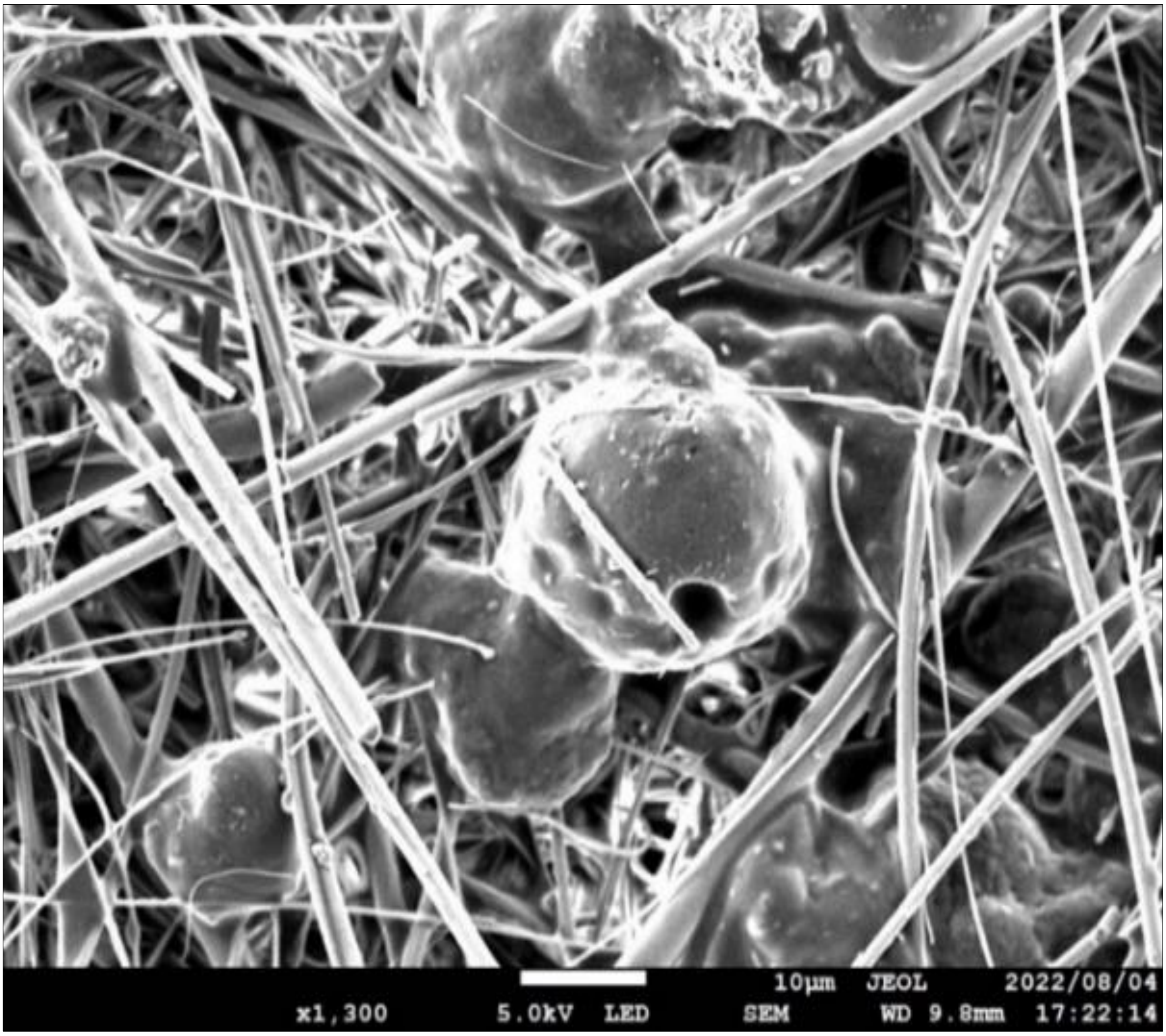


Figure VI. SEM photomicrographs of Fine PM fractions (2.5µm-10 µm) in Prunus × yedoensis 2.5µm membrane filter

Trees and shrubs can reduce human exposure to air pollution through the interception of aerosol particles or through the uptake of air pollution toxic gases via leaf stomata on the plant surface **[23-25].**

Significantly, trees and shrubs can capture large-size particles (large and coarse particles), but not fine particles as it is more sensitive to air turbulence and rain which distracts the continuous stable deposition as a result of their small size.

This is confirmed by **Dzierżanowski et al. [21] and Sæbø et al. [26]** which approved that the large and coarse fractions covered the largest proportion of all accumulated PM. On the contrary, **Freer-Smith et al. [27]** study recorded a greater accumulation of fine PM compared with large-size PM.

The amount of natural PM on the leaf surface of Ficus erecta which has a smooth inner structural surface, and a large surface area was lower than the amount of PM in other specimens, Ficus erecta can capture an unpretentious amount of large, coarse and fine PM fractions.

It is related that a larger leaf area (LA) may contribute to a larger boundary layer (refers to the still air surrounding the leaf) which increases its resistance to the deposition of particles around leaf surfaces, which may lead to a negative correlation to PM deposition **[28].**

While Toxicodendron succedaneum (wax tree) that has a rough dense inner structural surface (even though the leaves' external surface is smooth and glossy), compound leaves type and the smallest leaf area among the investigated types can capture large, coarse and fine PM effectively, with the highest portion for fine particles. This supports that the leaf size or area does not influence PM capturing capacity.

The same conditions are applied for Prunus × yedoensis which specifically has a serrate margin and rough surface(hairs), this structure can be the reason to capture coarse and fine PM as the same for large PM with a large portion for fine PM, as the presence of the leaf hair has been associated with high PM accumulation in many studies **[21], [29], [30].**

Cinnamomum camphora has smooth and glossy surface leaves, but the microscopic image shows a circular hollow surface, which allows capturing the coarse and fine PM in a middle range, it is not the same for fine particles which can be washed with rain very quickly (Figure VII).

Leaf microstructural features such as rough surface and hair in Toxicodendron succedaneum and Prunus × yedoensis can create different contact angles (θ) between a water droplet and the leaf surface, as a result, rough leaf surfaces can increase the contact time between rainwater and the leaf surface to remove PM from leaves, this helps leaves to keep particles more than the smooth surface leaves like Ficus erecta and Cinnamomum camphora which in same weather conditions, raindrops with low-intensity influence can quickly cross the water-repellent leaf surface and take away particulates **[31].**

Remarkably, the surface roughness can influence the transition from a laminar to a turbulent airflow **[32], [33].** Thus, the deposition velocity can be larger across a rough surface compared to a smooth surface **[34].**

Tree or shrub location does not have any significant influence on the particulate matter deposition capacity, two types of locations (busy roads and parking) were chosen in this study. Toxicodendron succedaneum and Ficus erecta trees are on the same busy road, but they recorded a high difference in PM deposition, while Prunus × yedoensis and Cinnamomum camphora are near car parking and busy road, Prunus × yedoensis recorded PM deposition more than Cinnamomum camphora, which is supposed to have a significant PM deposition percentage as it is very near to two PM main resources.

It is worth mentioning, that SDS was a sufficient remover, the particles removed using SDS is more than the DW, it is related that some particulate matter organic compounds are Hydrophobic such as polycyclic aromatic hydrocarbons (PAHs) **[35].**

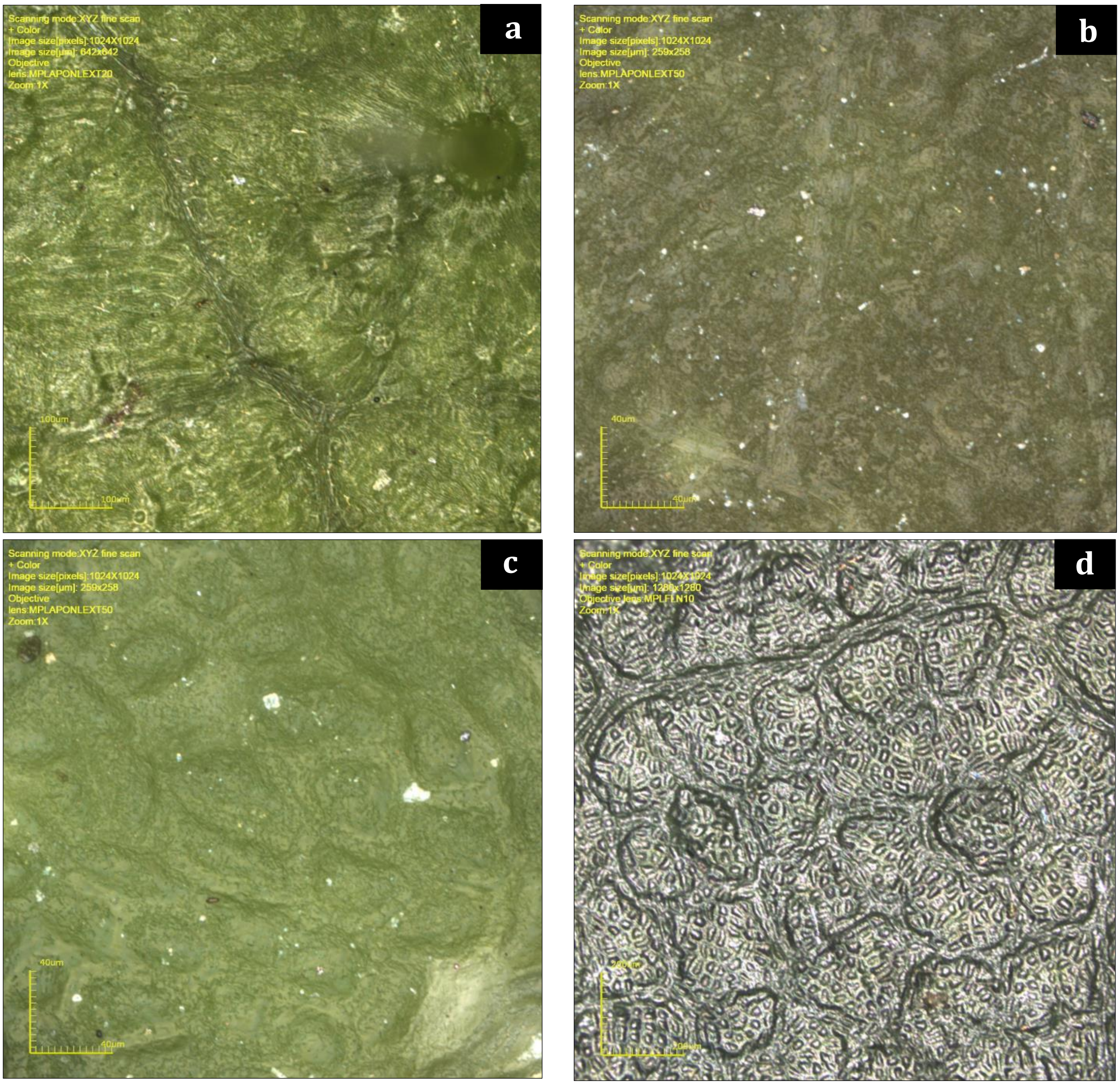


Figure VII. 3D Laser Measuring Microscope image for the four species leaves, (a) Ficus erecta, (b) Toxicodendron succedaneum, (c)Prunus × yedoensis, (d) Cinnamomum camphora

CONCLUSIONS

Investigating new plant species for phytoremediation is an essential part of research to enhance understanding of effects of air pollution. Whereas analyses that help to quantify the contributing effects of leaf traits can improve the understanding of the mechanisms of PM deposition.

In this study, the analyzed species captured 93.299 μg.cm^{-2} from ambient particulate matter, most of these particles accumulated in the plant's foliage belonged to coarse fraction size (2.5–10 μm) followed by large fraction, and finally fine fraction size.

The leaf surface area or leaf size has no effect on PM accumulation, as the highest amount of PM (41.4 μg.cm^{-2}) was captured by Toxicodendron succedaneum, which has the smallest surface area among the analyzed species while Ficus erecta which has the largest surface area, recorded the lowest portion of PM.

Leaf features such as hair and rough surfaces were traits with a positive correlation with PM accumulation which is similar to the same results obtained from previous studies **[9], [22], [24],[29], [31].** For instance, Prunus × yedoensis which has hairy rough surface leaves can capture coarse and fine PM as the same for coarse PM with a large portion for fine PM.

The best type of trees to capture PM among the analyzed species are Toxicodendron succedaneum and Prunus × yedoensis. In conclusion, the leaf that has surface traits, such as hair and surface roughness can retain more particulate matter than the smooth hairless leaf.

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CHAPTER III

Removal Capability of Vegetation in Different Seasons

III- I

Particulate Matter Phytoremediation Effectiveness of Japanese Prunus X Yedoensis Tree Through Spring and Summer Season

This chapter has been originally published in 8th International Conference on Industrial, Mechanical, Electrical and Chemical Engineering (ICIMECE 2023) October 30th -31st 2023, Indonesia.

ABSTRACT

Using green biofilters can help eliminating air pollutants and improving urban air quality. In the current study, ability of phytoremediation of Prunus × Yedoensis was investigated as a bio filter of Particulate matter (PM) in spring and summer seasons in Japan. Two samples were analyzed to extract three fractions of PM (large, coarse and fine).

Results showed low deposition of PM in spring season with total of 20.2 $\mu g.cm^{-2}$ and high proportion for coarse fraction (2.5-10μm), comparing with summer season which showed a higher deposition of PM with total of 31.9 $\mu g.cm^{-2}$.

Fine fraction (0.2–2.5μm) recorded the highest values with a percentage of 29.9% of the total accumulated PM deposition. Leaf micromorphological characteristics such as, rough surface and serrated margin can enhance the Prunus × Yedoensis ability of particulate matter accretion.

This study confirms the efficient role played by vegetation in improving the quality of urban air, moreover, deepen the understanding of phytoremediation phenomena.

Keywords—phytoremediation, ambient PM, retention, leaf traits, air pollution.

INTRODUCTION

Particulate matter (PM) which suspended and transported for long time and distances from the pollution sources in the air, is directly can aggravate many incurable childhood diseases including post-neonatal infant mortality and sudden infant death syndrome (SIDS) **[1-3].**

In Japan, the PM polluted air which has been observed commonly during the spring and summer especially in the west of Japan such as the Kyushu Island is similarly associated with severe health problems for healthy school children and pregnant women **[4-6].**

Therefore, applying various techniques and methods can decline the high concentrations of PM, as it stated in 2018 annual report of Japanese Ministry of Environment, which declared that achievement rate of Japan Environmental Standards for coarse particulate matter ($PM_{2.5}$) has recently increased from 30–40% from 2010–2014 to over 80% in 2017, even though, PM concentration still increasing steadily and using such techniques can be costly and complicated.

Surprisingly, plants can adsorb these pollutants simply, naturally and cost effectively through phytoremediation approach, which has been approved in many empirical studies such as **[7]** and **[8].**

In particular, Japanese plants have been investigated in many studies concerning pollution removal and remediation, such as Acer palmatum, Euonymus japonicus Thunb, Spiraea japonica L. and Aesculus turbinata Blume **[9, 10].**

The present study aims to investigate and address the phytoremediation ability of PM in another Japanese tree which was not studied in the previous studies as it is the growing period though the year is very short. Yoshino cherry (Prunus × yedoensis) is a Japanese tree that just blooms in Spring and its summer leaves falls totally in Autumn with considering the substantial connections between PM deposition and leaf size, leaf compositional morphology, leaf structure, and plant type.

METHODS AND MARTIALS

Plant types and experimental sites

Four samples (n = 20 leaves/ sample) have been collected randomly from Yoshino cherry tree (Prunus × yedoensis) at end of two different season, spring season represented by month of May and summer season by month of August, with a time gap between every two samples of every month is 14 days when there was no previous rainfall for more than 3-5 days. (more details shown in table I).

This tree was commonly planted in urban areas, along roads, streets in Kyushu university campus (Fukuoka, Japan) with a distance between the road and the tree ranges from 2 to 3 m.

Sampling height was from 3-5 m above ground level in four directions (north, south, east and west). All leaf samples were individually placed and sealed into pre-labelled sample paper bags to minimize PM loss and stored at ambient temperature (4^0C) in a clean laboratory refrigerator until analysis.

The weather conditions during the sampling period were standard to two seasons, spring season and the summer season (shown in Table II) according to Fukuoka WMO station (ID:47807) on-site, Japan Meteorological Agency **[11].**

Table I. Analyzed plant characteristics according to North Carolina university plant box

Species	characteristics				
Yoshino cherry	Prunus × yedoensis	Rosaceae	Oblong, veined	Serrated	Hairy rough

Table II. Weather characteristics through spring and summer season in sampling location

Season/month	Characteristics		
	Weather condition	Monthly mean daily temperature	Monthly precipitation
Spring/may	Warm, sunny with low rainfall	16.8-24.9 °c	45 mm
Summer/august	Sunny, very hot, humid with a middle level rainfall	26.8-33.8 °c	266.5 Mm

Quantitative Gravimetric analysis of PM

Both adaxial and abaxial surfaces of the leaves were washed (250 mL of distilled water, 150 mL of SDS), analyzed, according to **[7],** method which was applied through four filtration tools (a-106 μm sieve, b- 10 μm membrane filter, c- 2.5 μm glass microfiber filter, d- 0.2 μm PTFE membrane filter) to extract three fractions of particulate matter (large (10-100μm), coarse (2.5-10μm) and fine (0.2-2.5μm).

The total leaf area of plant leaves was measured using ImageJ software. 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 the selected species and PM accumulation.

EQUATIONS

The particulate matter quantity was measured through this equation according to **[7].**

$$PM_{\text{deposition}} = (\mathbf{W}(F + PM) - \mathbf{W}F)/A \qquad (1)$$

$W_{(F+PM)}$: Weight the filter with the particulate matter after filtration in μg.

W_F: Weight of the filter before filtration in μg.

A: Total Surface area of leaf in cm^2

RESULTS

Quantification of PM

Considering the proved finding of **[13],** that showed that PM concentrations were highest during spring and summer because of dust events in spring and photochemical reactions in summer.

A notably high amount of PM was captured on the foliage of Yoshino cherry (c) tree throughout the experiment period with a higher quantity for summer season samples(total of 31.9 $\mu g.cm^{-2}$, 61.2 %) total of than spring season samples (total of 20.2 $\mu g.cm^{-2}$, 38.7%) (shown in Figure I). Lower values were also measured in spring season in different previous studies, for example, **[14].**

Smaller size particles recorded higher deposition than larger size particles in both seasons, in total,coarse particles recorded 21.9 $\mu g.cm^{-2}$ (42.1%), while fine size fraction recorded 15.6 $\mu g.cm^{-2}$ (29.9%), reflecting the findings of [15], the smaller the diameter of particles, the higher the deposition captured (according to Buxus sempervirens with (34.6 mm^{-2} and 12.8 mm^{-2} respectively for PM_1 and $PM_{2.5}$). **Dzierzanowski et al [7]**, on contrary, found comparatively higher deposition of larger size particles on plant foliage leaves compared to smaller size particles.

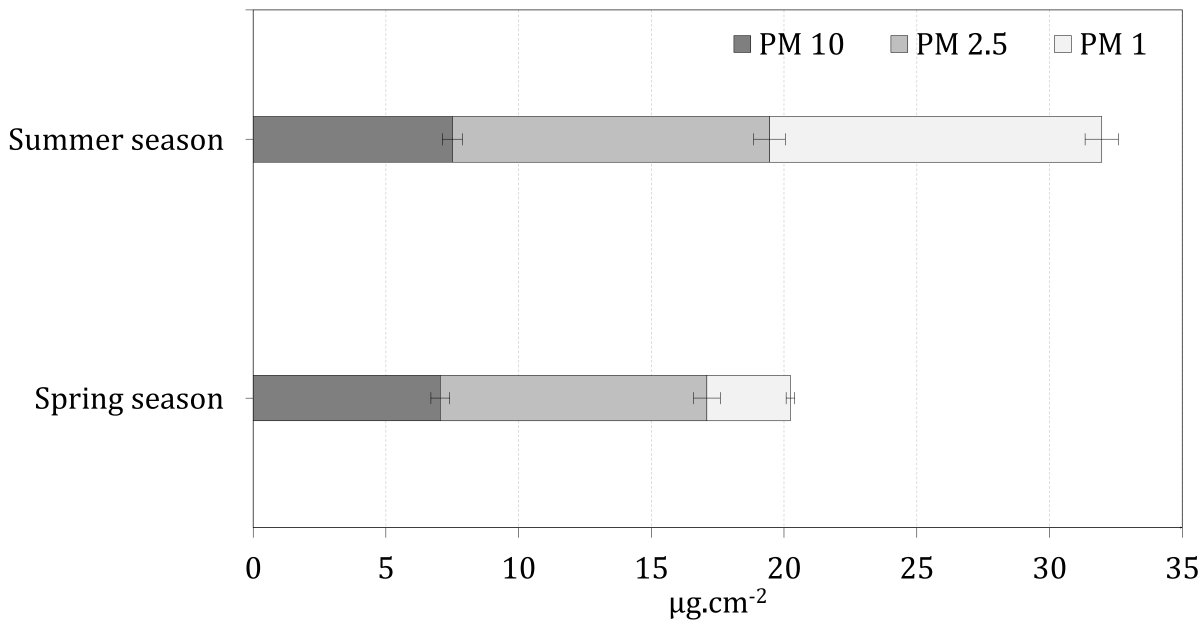

Figure I. Accumulation of particulate matter (means± SD) on leaves within three different size fractions for Prunus × yedoensis in spring and summer season.

In spring season, especially in early May, Yoshino cherry (Prunus × yedoensis) leaves, were young unmatured leaves, as a result it recorded a low deposition of PM. This approved by **[15],** who noted that the unmatured leaved species showed comparatively low PM deposition, suggesting that this was due to the low capacity to withstand PM contaminated air flow, thus lowering the turbulence surrounding the leaf boundary. In second sample (at the end of the spring), the PM deposition was higher than the first sample (at the start of experiment).

The reason may be correlated to accumulative increasing in the surface area of tree leaves as it shown in Figure II. Coarse particles (2.5-10) μm which mainly originate from vehicle emissions, combustion, and industrial processes, recorded the highest proportion of total spring PM deposition with 10.04 μg.cm^{-2} (19.2%), followed with large fraction (10–100) μm with 7.04 μg.cm^{-2} (13.5%), while fine fraction (0.2–2.5) μm was the lowest with 3.14 μg.cm^{-2} (6%) **[16].**

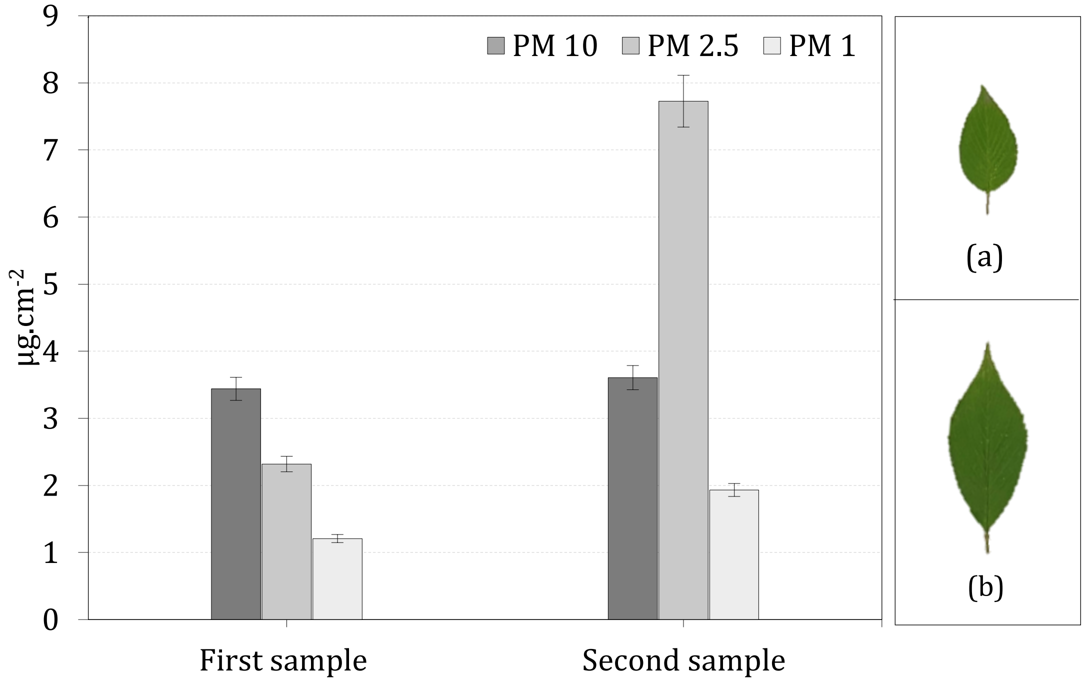

Figure II. Accumulation of particulate matter (means± SD) on leaves within three different size fractions in the spring season for Prunus × yedoensis, (a) and (b) represent different leaf sizes of the analyzed species (at the start and late of Spring season).

Similarly, in the summer season, PM quantity on the leaf's foliage was higher in the second sample, that can be explained, that the leaves became more mature leaves and has a significant larger surface area more than it was in the spring season as it shown in Fig. 3. Additionally, anthropogenic and transportation activities, in summer season are more prevalent than other seasons.

Coarse fraction (2.5-10) μm recorded the highest portion with 12.5 μg.cm^{-2} (23.9%), followed by large fraction (10–100) μm with 11.9 μg.cm^{-2} (22.9%), while fine fraction (0.2–2.5) μm was the lowest with 7.5 μg.cm^{-2} (14.4%), as the main source of smaller size particles (coarse and fine) is combustion of fossil fuels and photochemical reactions **[16].**

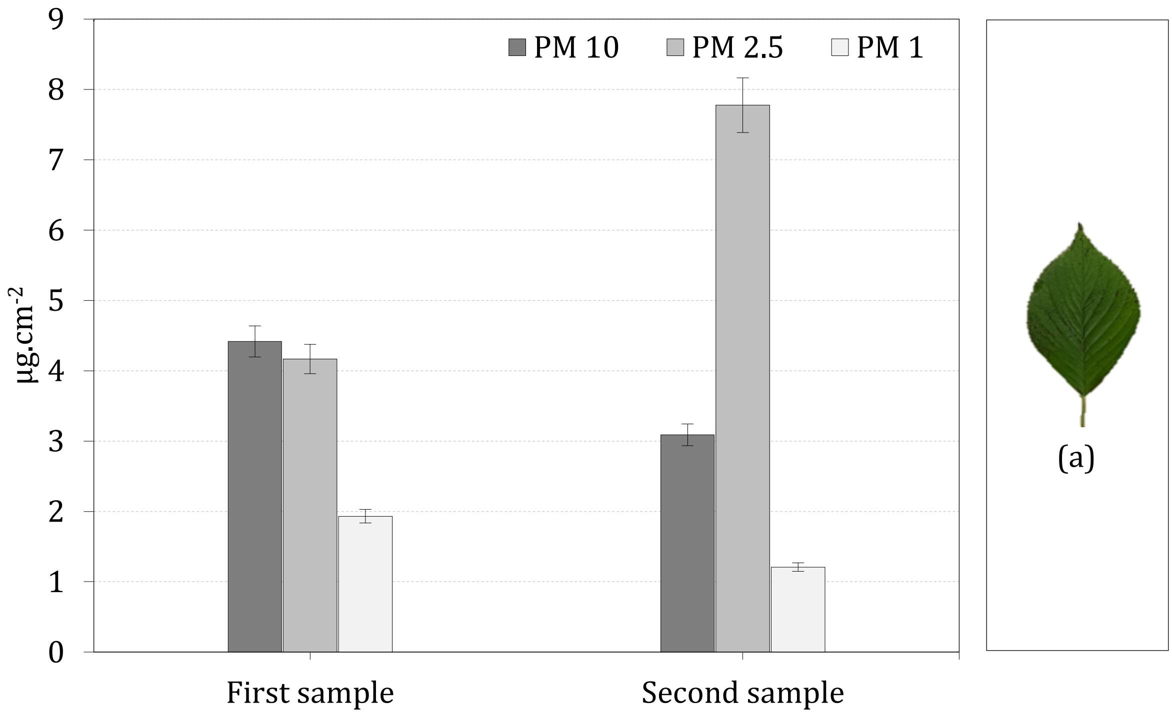

Figure III. Accumulation of particulate matter (means± SD) on leaves within three different size fractions in the summer season for Prunus × yedoensis. (a) represents the size of healthy mature leaf at the late of summer season

Effects of leaf morphological structural traits on PM deposition

Many studies indicate that leaves with complex micromorphological traits, especially, surface roughness owing to three-dimensional leaf structures such as hairs, scales, and veins, has been demonstrated to surge particulate retention **[8, 17 - 20].**

In the same way, present study proved that rough surface,veins and leaf surface hair density (Figure IV) were found to be significant predictors of accumulation of the total sum of PM inside the foliage of Prunus × yedoensis tree. As rough surface and hairs can create different contact angles (θ) between rain droplets and the leaf surface, which will increase the boundary resistance of the leaf and the adhesion period of PM on the leaf surface through weather changes such as air turbulence and precipitation.

Additionally, the presence of leaf hairs can also change the PM deposition as some leaf hairs are hydrophobic which can attract charged particles attached to different compounds and elements such as heavy metals **[21].**

Hairy leaves with a complex micromorphology have been considered as being sufficient in capturing more particulates than smooth leaved species **[8, 18].** High PM_1(fine particles) densities were found on Geranium macrorrhizum due to dense surface hair of their leaves **[15].** Confirming results of Prunus × yedoensis tree in the present work.

On other hand, negative relationship between presence of leaf hairs and PM accumulating according to **[22].** Furthermore, Leaf margin can enhance particulate matter accumulation, Prunus × yedoensis serrated margin with sharp toothed edges can help keeping deposited particulate matter on the leaf surface and decrease repeated resuspension. It was noted that PM deposition is relatively high on the leaf margins on species with convex veins and wavy edges such as F. religiosa **[20, 23, 24].**

This study found that trees are effective species in PM capturing, while it is strongly linked to the characteristics of the species **[8, 25].** Trees with large surface area (LA) and crowns resulting from the complex form of foliage leaves increases turbulent air movement, then can enhance capturing more particles than other types of plant species **[26, 27].**

It worth to note that some of species leaf traits were differed among the selected seasons, especially leaf surface area which can explain the difference in PM deposition among spring and summer season, in spring season, leaf surface area was smaller (15.3 cm^2) than summer season (38.2 cm^2).

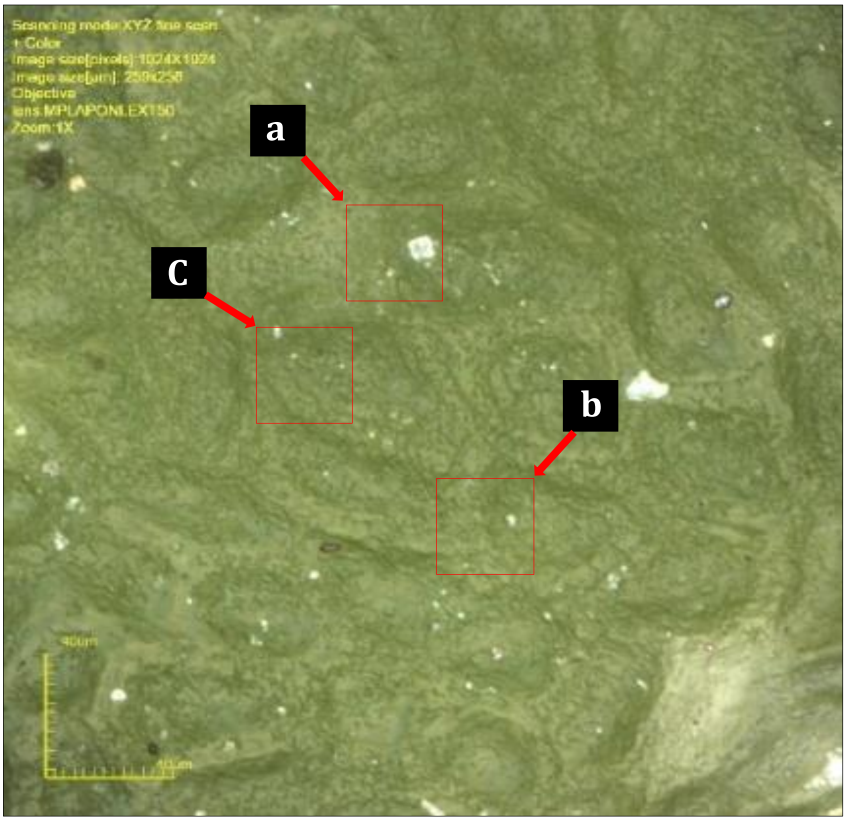

Figure IV. Morphological structure of Prunus × yedoensis leaf surface, (a), (b) and (c)represent different sizes of particulate matter deposited on the analyzed leaves, (3D Laser Measuring Microscope).

Particulate matter accumulation is different through the four seasons, summer season proved to have a high effect on the accumulation of PM, as a result of anthropogenic activities and photochemical reactions.

While spring season had a low effect on the PM accumulation. Leaf morphological traits such as rough surface and leaf hairs enhanced the PM capturing inside the leaves foliage of Prunus × yedoensis through the experiment period.

The PM accumulation on leaves foliage is subjected to repeated wind and rain events which can interrupt the accumulation process and increase PM deposition loss.

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III-II

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

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ABSTRACT

Plant species can act as natural bio-filters and capture, degrade, and metabolize air pollutants in-side their foliage. Immobilization efficiency of Particulate Matter (PM) was investigated on 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. A significant quantity of PM in three size fractions with aerodynamic diameter ranging between (0.1-100 μm) was captured inside the analyzed leaves foliage with a total of 179.02 $\mu g.cm^{-2}$. Coarse particles (2.5 -10 μm) dominated the highest portion of all deposited PM with 70.6 $\mu g.cm^{-2}$(39.5%).

Shrubs were demonstrated as the most efficient plants among the analyzed species. Elaeagnus pungens Thunb. accumulated the largest PM quantity with 83.6 $\mu g.cm^{-2}$, whereas the lowest deposition (18.6 $\mu g.cm^{-2}$) was recorded by Cirsium vulgare. Leaf traits such as grooves, trichomes, roughness, and margin are considered key factors associated with positive PM deposition, whereas the total surface area of the leaves had no direct correlation to PM accumulation process.

Keywords: Pollution; Phytoremediation; Plants; PM mitigation; Deposition; Foliage; Morphological traits; Particulate matter.

INTRODUCTION

Extensive use of fossil fuel is considered 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 is classified according to aerodynamic diameter into four categories, total suspended particles (TSP) with a diameter ≤100 µm, large particulate matter (PM_{10}) with an aerodynamic diameter ≤10 microns, coarse particulate matter ($PM_{2.5}$) with an aerodynamic diameter equal to or less than 2.5 microns and the most harmful component, fine particulate matter (PM_1) with a diameter of ≤1.0 µm **[1,2].**

Unfortunately, short to long-term exposure to high concentrations of PM may initiate 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 be furthering to the so-called 'sick building syndrome'(SBS) and 'building-related illnesses'(BRI) **[3-8].**

As a result, it causes approximately 2.1 million worldwide premature deaths annually [9,10]. Particulate matter pollution in Japan proved to trigger acute health effects, as trace elements, such as Se, Pb, As, Zn, Cu, Cr, Ni, and Cs which distribute homogeneously within the particles were correlated to nasal, ocular, and skin symptoms and severe non-carcinogenic risk to moderate carcinogenic risk for healthy school students **[11].**

Furthermore, **Takeda et al., [12]** found 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. For instance, World Health Organization (WHO) Air Quality Guidelines which are based on systematic reviews and subsequent evaluation methods, serve as a 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, innumerable controlling, monitoring, and remediation methods were used to deteriorate the PM pollution values such as 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 include particulate matter, through adhesion to vegetation leaves foliage or absorbing inside roots (plant's rhizosphere) can be used as an alternative for complicated and costly techniques **[16-18].**

In general, plant–microbe interactions play an important part during phytoremediation. When plants absorb air pollutants, bacteria living on these surfaces, called phyllosphere and endophytic bacteria can detoxify part of the pollutants using degradation or sequestration, moreover, rain may cause flow down of the deposited pollutants to the soil, where it interacts with the soil, the plant's rhizosphere and the roots **[19,20].**

Prioritizing that PM deposition on vegetation foliage depends on different factors, such as environmental conditions (precipitation and air turbulence) and plant characteristics (type, structure, morphological traits…etc) **[21,22].**

Various empirical studies inspected PM removal efficiency in different plant types via deposition onto leaves. PM deposition on leaves of Quercus ilex tree was quantitatively analyzed by **Sgrigna et al., [23]. Viecco et al., [24]** evaluated the impact of biodiverse Green Roofs (GRs) and Green Walls (GWs) on $PM_{2.5}$ capturing, through analyzing seven species using gravimetric dry deposition method. The study concluded that the higher the biodiversity of plants in GRs and GWs, the higher the $PM_{2.5}$ quantity can be captured.

Whereas **Dzierżanowski et al., [25]** 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 the significant quantity of particulate matter on their leaves.

However, plant species differ in their removal capacity of PM. Due to large leaf area and crowns which promote vertical transport by enhancing turbulence **[26].** Trees are believed to be the most efficient plant group in scavenging particulate matter according to **Sæbø et al., [27] and Muñoz et al., [28].**

However, there is an increasing hypothesis that trees can affect air quality negatively, through emitting toxic compounds, such as volatile organic compounds (VOCs), that may contribute to ozone formation **[29].**

Nevertheless, there is still a lack of knowledge about the deposition of PM on other types of plant species, especially marginalized plants that grow and interact with trees while having the possibility to collect the same quantity of PM or larger **[16,30].**

Moreover, native Japanese broad-leaved plants were not studied in the previous studies, though their significant adaption to environmental stress and resistance to air pollution. The focus of previous studies was evergreen linear leaved species as they can accumulate PM inside their foliage for several years **[30,31].**

The present study aims to investigate and address the capturing ability of PM in shrubs, climbers, and herbs while considering substantial associates among PM deposition and leaf size, leaf compositional morphology, structure, and plant type with the hypothesis that the plants with large leaf areas are supposed to capture more particles than smaller plants.

Methods and Materials

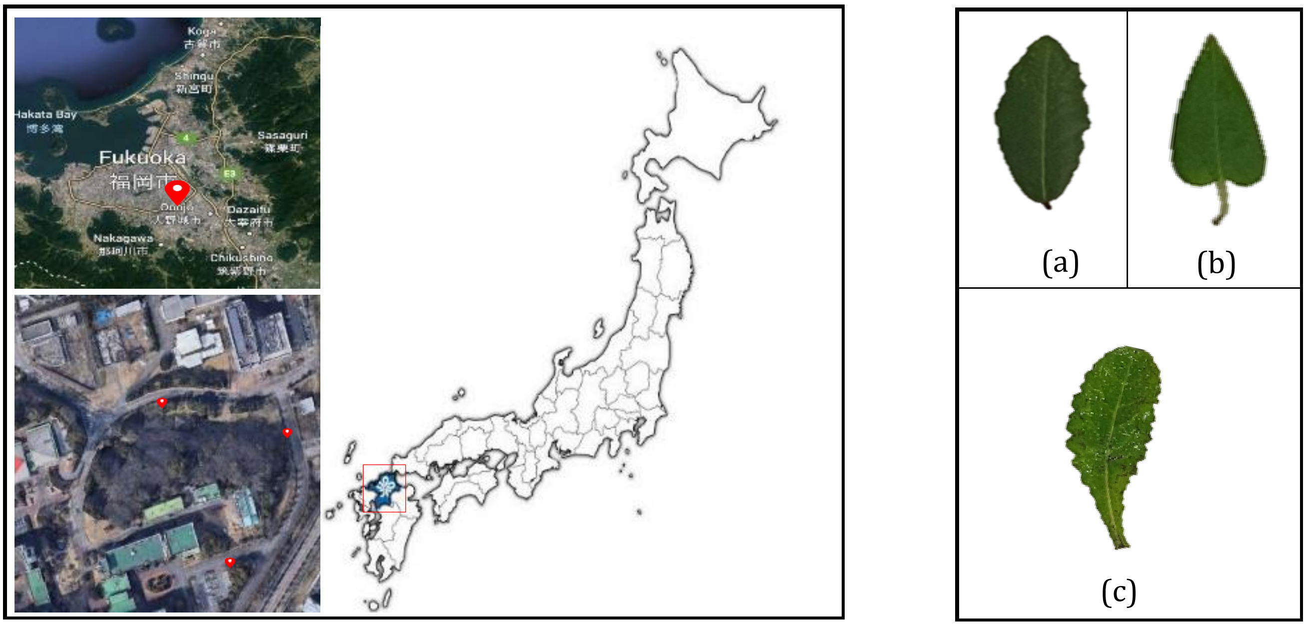
Leaves samples and experimental sites

Three species types of broad-shaped leaves represented by Elaeagnus pungens Thunb. (shrub), Dioscorea japonica (climber), and Cirsium vulgare (herb) were analyzed in the present study (more details shown in Table I).

These were all commonly planted in urban areas, alongside the roads and streets at Kyushu University's campus (Fukuoka, Japan) (shown in Figure I) 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.

Table I. Analyzed species leaf characteristics according to North Carolina university plant box [32].

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



(A) (B)

Figure I. (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 **[33]**

With a gap of 14 days (experimental period), two mature healthy leaf samples (n = 20 leaves/ sample) were collected, when there was no previous rainfall for 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-labeled sample paper bags since PM is susceptible to static electricity and stored at 4°C in a clean laboratory refrigerator before conducting the analysis **[34].**

The weather conditions were standard for the summer season in Fukuoka prefecture, sunny, very hot, and 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 the Japan Meteorological Agency **[35].**

Specific weather information was collected in the sampling zone using Datasheet SPS30 Particulate Matter sensor include concentrations of PM_1, $PM_{2.5}$, PM_4, PM_{10}, humidity, temperature, and pressure through the experimental period (shown in Figure II & Figure III).

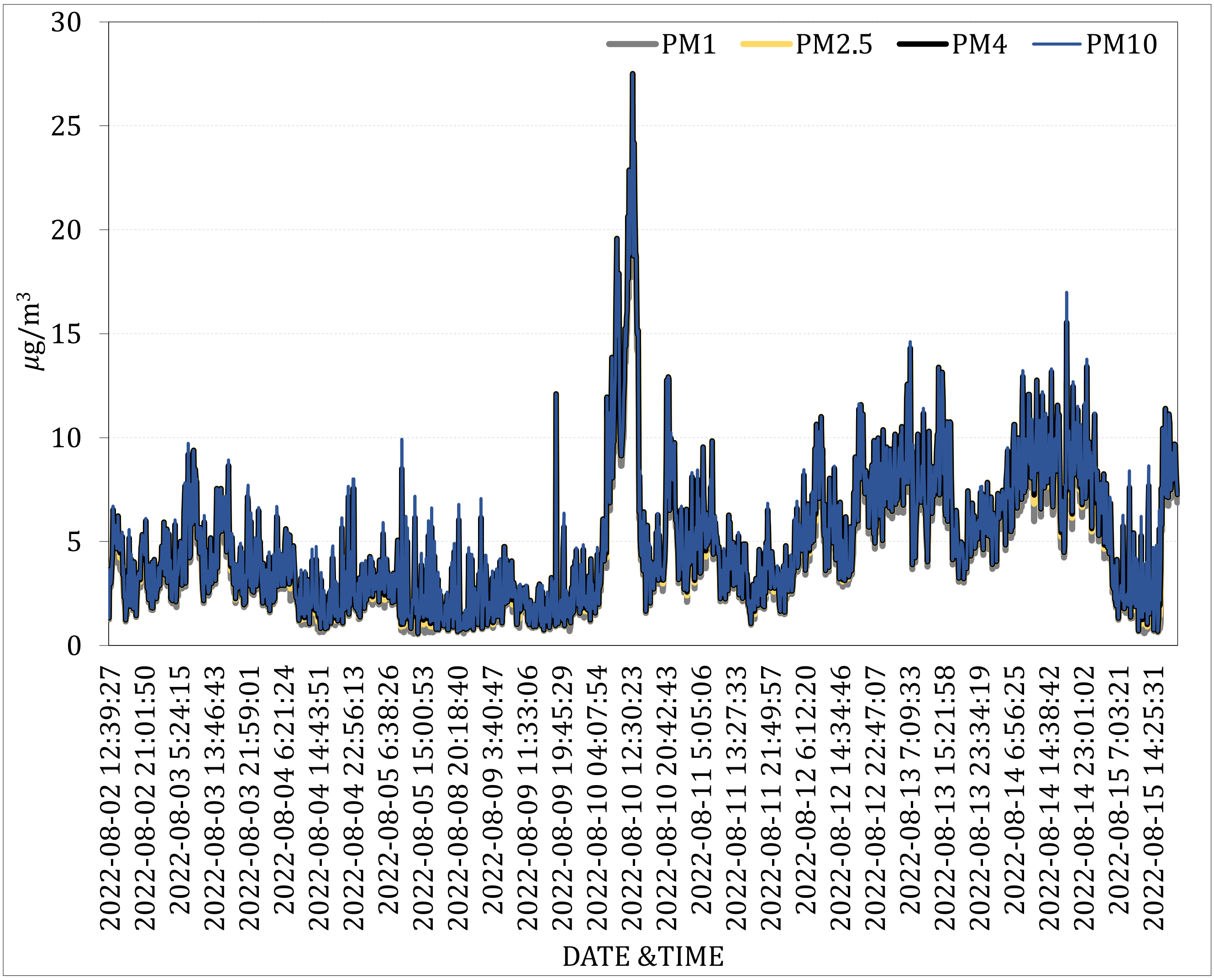


Figure II. Particulate matter concentrations in sampling zone.

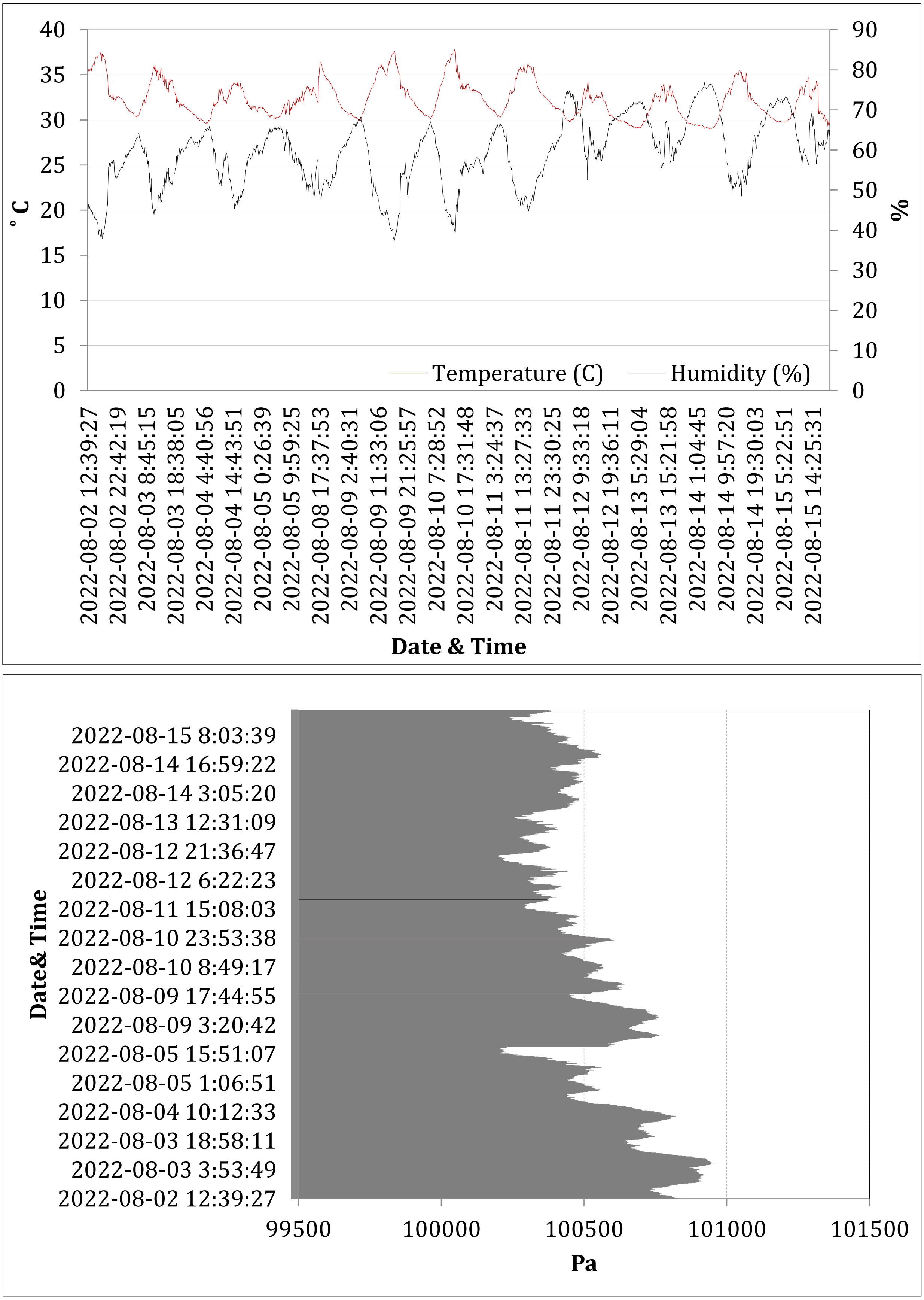


Figure III. Temperature, Humidity and Pressure in sampling zone.

Quantitative Gravimetric analysis of PM

Extracted PM quantity was analyzed according to **Dzierżanowski et al. [25]** dry gravimetric method. Two standard solutions were selected 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)(CH3(CH2)11OSO3Na) (Merck, Germany) with a 5% concentration was used for 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 [36].
A trial experiment with a 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 forced 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, washing solutions were applied to four filtration steps through (i) polypropylene sieve (with 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).

The filtration system setup consisted of a 25 mm vacuum filter holder with a stopper support assembly connected to a vacuum pump. Based on this setup, three fractions of PM: (i) 10–100 µm (large), (ii) 2.5–10 µm (coarse) and (iii) 0.2–2.5µm (fine) were successfully extracted. The filters were dried before and after filtration for 30 minutes at 60 °C, stabilized in the weighing room for 30 min, and weighed using an electronic balance (HT124R, ViBRA, Japan).

The total leaf area of plant leaves was measured using ImageJ software (U. S. National Institutes of Health [37]) depending on the same method as Osunkoya et al. [38]. 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 the selected species and PM accumulation.

CALCULATIONS

The particulate matter deposition in $\mu g/cm^{-2}$ was quantified according to this equation **[25].**

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

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.

RESULTS

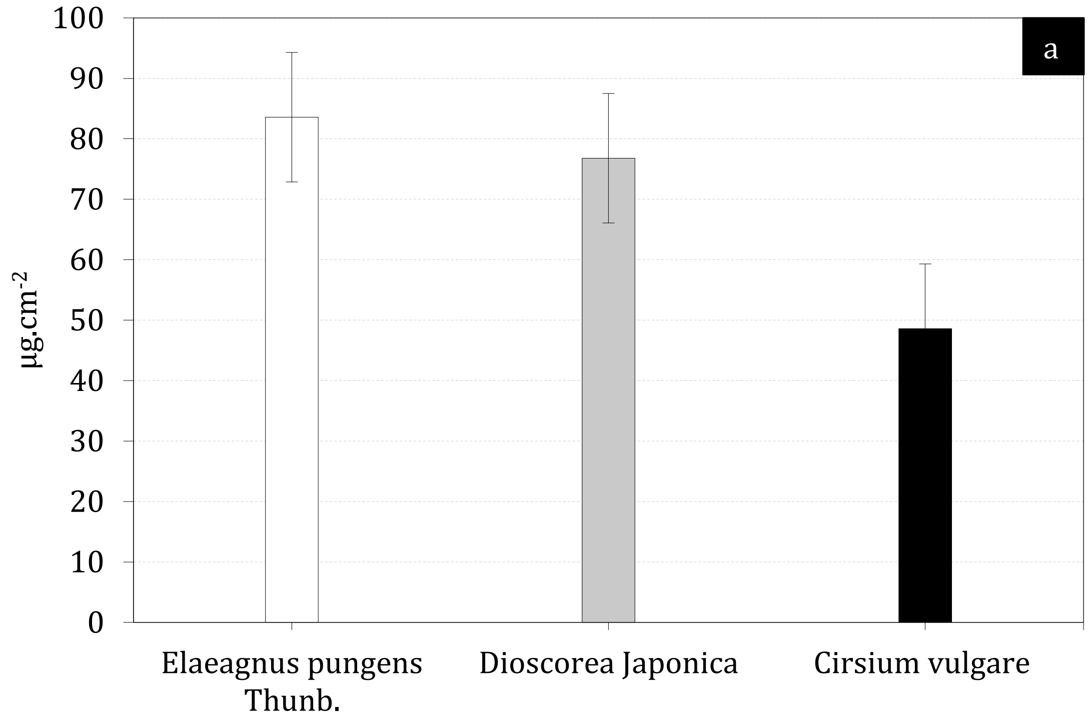
There was a substantial interaction among the analyzed species and ambient PM emerged with a difference in the removal efficiency of all PM fractions in both samples and period. A notable quantity of PM (total of 179.02 $\mu g.cm^{-2}$) was found on the foliage of the analyzed species throughout the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

Considering the findings of **Dzierżanowski et al. [25] and Sæbø et al. [27],** larger size fractions dominated the highest proportion of all deposited PM. Similarly, in the present study, majority of PM deposited on plants` foliage was large-size particles. Coarse size fraction (2.5 -10μm) has recorded total of 70.6 $\mu g.cm^{-2}$ (39.5%), followed by large size fraction (10 -100μm) with 69.8 $\mu g.cm^{-2}$ (39%), whereas fine fraction (0.1 -2.5 μm) was the lowest with 38.5 $\mu g.cm^{-2}$ (21.5%) as it shown in Figure (IV, a). The miniscule size of fine size fraction makes them more susceptible to weather conditions which diminutions the continuous stable deposition and allows them to be washed off by precipitation or resuspended with air turbulence. Shrubs with large leaf areas (crowns) similar to trees were proven to accumulate substantial quantities of particulate matter **[39].** We also found that the analyzed shrub (Elaeagnus pungens Thunb.) showed to be the most efficient plant species in PM accumulation, while Cirsium vulgare (herbaceous species) has recorded the lowest deposition (shown in Figure (IV, b). Similar conclusions were confirmed by **Dzierżanowski et al., [25]** who found largest quantity of PM on shrub leaves (Spiraea). On the other hand, **Ottelé et al. [40]** did not observe any difference in PM accumulation among sampling heights ranged between 0.75–2.0m.

Among the tested plant species, Dioscorea Japonica captured the highest quantity of large size fraction (10-100 μm) with 40.04 $\mu g.cm^{-2}$, while Elaeagnus pungens Thunb. has recorded the greatest values of coarse size fraction (2.5 -10 μm) and fine size fraction (0.1-2.5 μm) with 41.4 $\mu g.cm^{-2}$ and 19.3 $\mu g.cm^{-2}$, respectively (Table II).

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

Species	PM size fractions accumulation($\mu g.cm^{-2}$)					
	Large (10-100μm)	(%)	Coarse (2.5-10 μm)	(%)	Fine (0.2-2.5 μm)	(%)
Elaeagnus pungens Thunb.	22.8± 0.4	12.7	41.4± 2.9	23.1	19.3±2.01	10.8
Dioscorea japonica	40.04±1.6	22.4	21.5±0.2	12.03	15.2±0.9	8.5
Cirsium vulgare	6.9±0.3	3.9	7.7±0.9	4.3	3.9±0.6	2.2



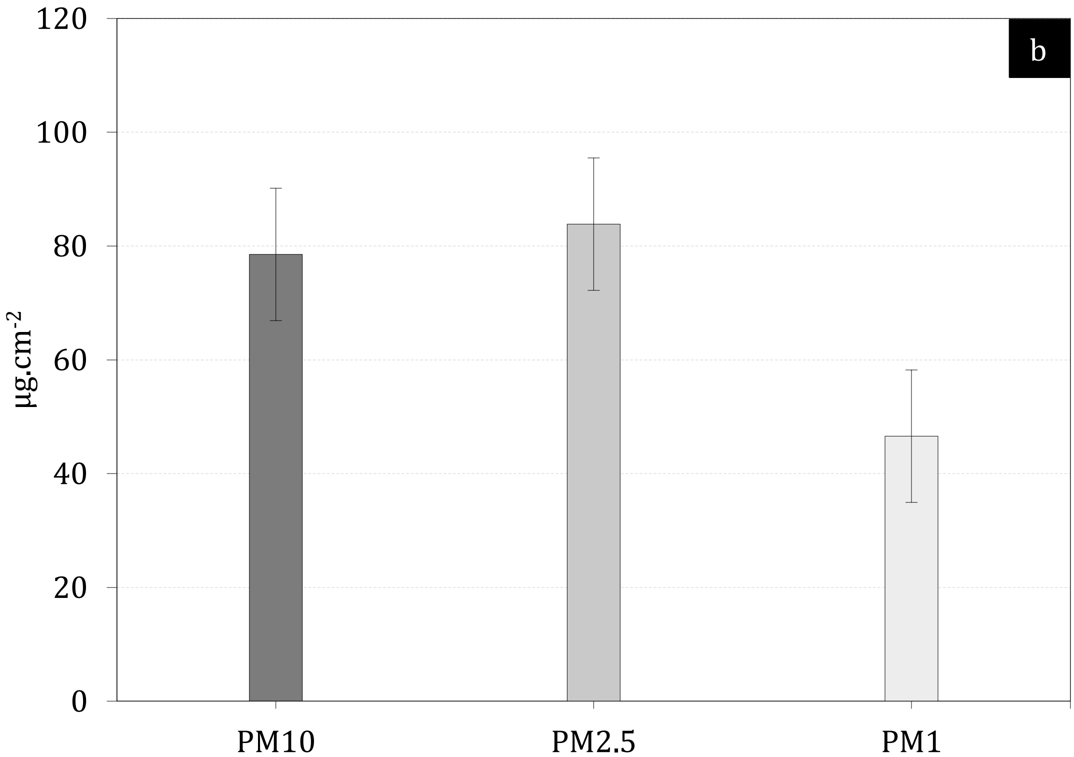


Figure IV. a) Total PM deposition($\mu g.cm^{-2}$) in the analyzed species, b) Total PM fractions($\mu g.m^{-2}$) deposition in the analyzed species

Discussion

Difference in PM accumulation among the analyzed species

Shrubs presented the major percentage of low-height strata being planted on the 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, coarse size particles which considered, besides fine size fraction as the most harmful PM components. According to Stapleton et al. **[41],** shrubs can lessen to 5.9% of $PM_{2.5}$ concentration in small spaces. Moreover, plant species growing near the ground can capture more particles on their leaves, due to their exposure to different types of pollution such as soil splash **[42].**

The analyzed shrub, Elaeagnus pungens Thunb. captured the largest portion of all accumulated PM with total of 83.6 $\mu g.cm^{-2}$. Coarse size fraction (2.5-10 µm) made up the greatest proportion of accumulated PM mass and fine size fraction (0.1-2.5 µm) recorded the smallest proportion. Expressed as percentage, 27.3%, 49.5%, and 23.2% for large, coarse and 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 are 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 areas 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 **[43-47].**

Furthermore, climbers through interacting with trees and shrubs, can capture more quantities of particulate matter than other species. They can produce an enormous leaf area (about the land area occupied) compared with low-height strata which enhances the capability of accumulating PM and other pollutants **[37].** Dioscorea Japonica has recorded a total of 76.8 $\mu g.cm^{-2}$, which is the second-highest particulate matter deposition among the analyzed species. Large size fraction (10 -100 µm) represented the highest portion with 52.1%, while coarse size fraction (2.5-10 µm) recorded at 28.05% and finally 19.8% for fine size fraction (0.1-2.5 µm) of all deposited PM on the leaves of the analyzed climber (Dioscorea japonica).

Weber et al. [48] confirmed removal capacity of 16 species of herbaceous plants. Although, herbaceous plants can capture fallen particles 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 **[49,50].** In this work, Cirsium vulgare (herbaceous species) captured lower particulate matter (18.6 $\mu g.cm^{-2}$, 10.4%) than other tested species due to smaller surface area, and higher influence of weather conditions. Coarse size fraction (2.5 -10 µm) dominated the largest portion with 7.7 $\mu g.cm^{-2}$(4.3%), followed by large size fraction (10 -100 µm) with 6.9 $\mu g.cm^{-2}$ (3.9%), whereas 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 V), in contrast to 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., [51].** In a study to investigate the impact of precipitation(154mm) on concentrations of PM (0.2-50 µm) in diameter, a significant decrease was demonstrated for particles from 10-50 µm in diameter, furthermore, the 10-fold decline in concentrations of particles 10-30 µm in diameter was found under heavy rain event for one hour. The measured particles between 10-50 µm in diameter diminished after rain with a lasting effect of approximately 10-15 h **[52].**

Therefore, different strategies and mechanisms should be applied to enhance phytoremediation under weather circumstances, for example, applying technical solutions (physical barriers, overhead protection..etc), various soil amendments (biochar, organic matters, organic acid exudates, humic substances, phytochelatins), selecting hyperaccumulator plants in urban polluted areas, adding plant growth-promoting bacteria endophytes microbes, using genetically engineered plants **[53,54,55].**

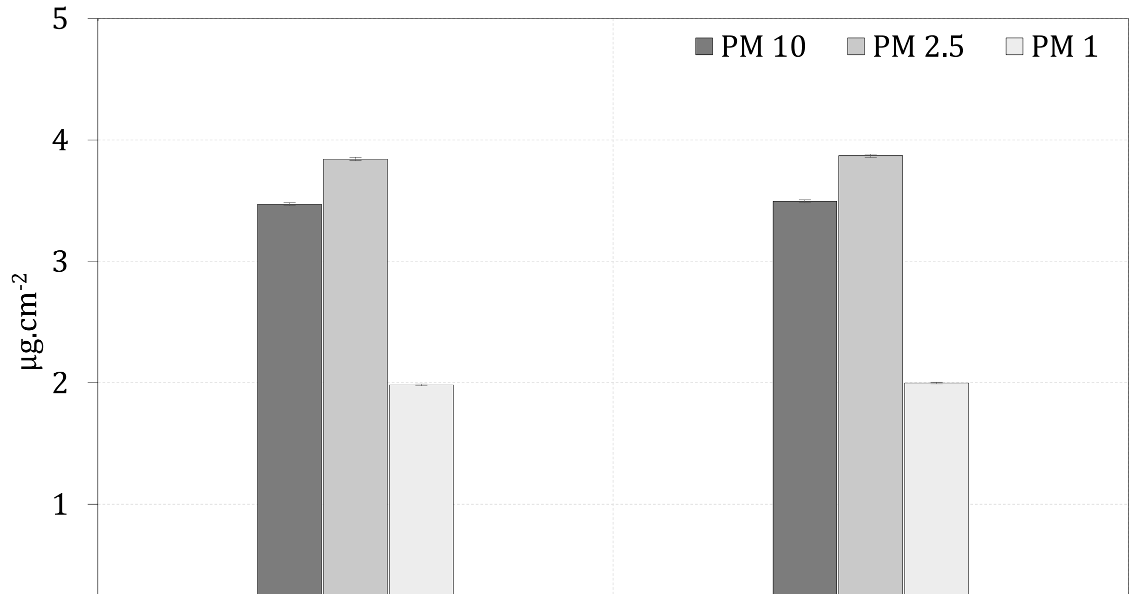


Figure V. Accumulation of particulate matter (means± SE) (µg.cm^{-2}) within three different size fractions on leaves of Cirsium vulgare.

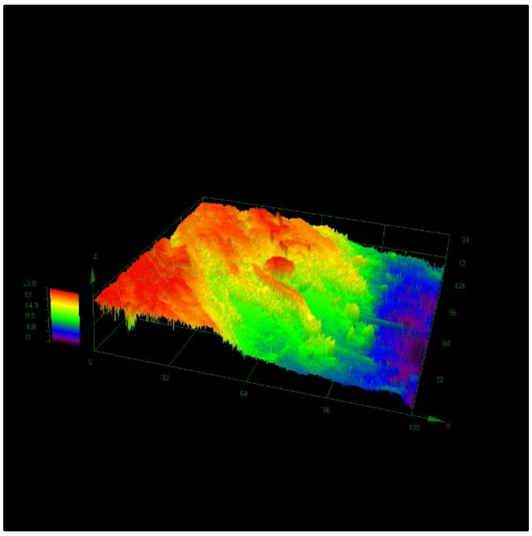
Trees, shrubs, climbers, and herbs can filtrate the ambient air through natural phenomenon called phytoremediation, through transferring, removing, destroying, or stabilizing and accumulating of aerosol particles and other pollutants via leaf stomata or roots **[9,10], [56].**

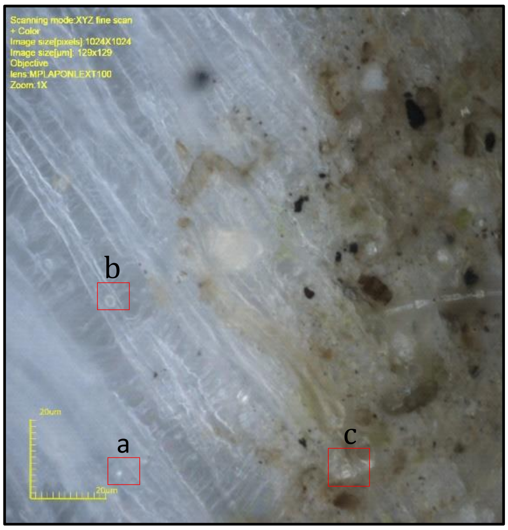
Nevertheless, these species differed in their capabilities to accumulate particulate matter and other pollutants, Shrubs which have large leaf surface areas are more exposed to higher quantity of particulate matter, whereas climbers may accumulate their PM deposition through interacting with other plants. The lowest deposition of herbaceous plants is related to smaller leaf surface area and high possibility of being exposed to weather conditions.

Leaf traits & PM accumulation in the analyzed species

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., [30]** and **Leonard et al., [57].**

To exemplify, Elaeagnus pungens Thunb., which has a leathery rough compositional surface, as it is clear in the 3D microscopic image (shown in Figure VI (a) (3D Laser Measuring Microscope), captured the highest total PM among the analyzed species with 83.6 $\mu g.cm^{-2}$ included three size fractions (large, coarse and fine) as it shown in Figure VI (b). Rough surface includes trichomes and grooves can increase the contact time with rain droplets as it can create dissimilar contact angles (θ) (representative of leaf wettability) between water droplet and leaf surface, that enriches larger sized particles adherence to leaf surface for a long time and delays their washing off. It was proved that surface roughness can substantially impact the fluid dynamics in convective flows through generating perturbations in the velocity profile which influence surface drag and turbulent mixing **[58].** Furthermore, leaves with ridges and grooves such as Dioscorea japonica allowed to capture considerable quantity of large-size particles (61.6 $\mu g.cm^{-2}$) as their size can perfectly adhere to surface grooves, meanwhile, fine size fraction (0.2-2.5µm) which recorded 15.2 $\mu g.cm^{-2}$ for both samples, can partially adhere to the same microfeatures with high possibility to be removed with air turbulence and precipitation which can flow them down to the surrounding soil, where it can be absorbed by plant's rhizosphere and roots **[59].**





(A) (B)

Figure VI. 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).

Considering **Dzierżanowski et al. [25], Sæbø et al. [27], PAULL et al. [59], and Fernàndez et al. [61]** findings, it can be concluded that the presence of leaf hair (trichomes) has been associated with positive PM accumulation in diverse analyzed plants as some leaf hairs present hydrophobicity, thus attract charged particles and heavy metals. Moreover, **De Nicola et al. [62]** found that trichomes can enlarge the surface area in which ambient particles may be deposited.

In this study, we also found that hair presence can enhance the PM accumulation on the leaves. The hairy surface of Cirsium vulgare allowed to accumulate substantial quantities of large, coarse, and size fractions of PM with 15.6 $\mu g.cm^{-2}$, 20.9 $\mu g.cm^{-2,}$ and 12.02 $\mu g.cm^{-2}$, 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, accumulation speed on a rough surface will be higher than on a smooth surface. **[62,63].**

According to **Mitchell et al. [64]**, PM is deposited mainly on leaf margins or along main leaf nerves, therefore, leaf margin can also enhance particulate matter accumulation. Elaeagnus pungens Thunb., undulate margin, with a wavy shape that curves up and down, for instance, can prevent deposited particles washing off or resuspension, while lobed margin which belongs to Dioscorea japonica and Cirsium vulgare can partially impedes PM falling from leaf surface with support of additional traits such as grooves and trichomes in each species.

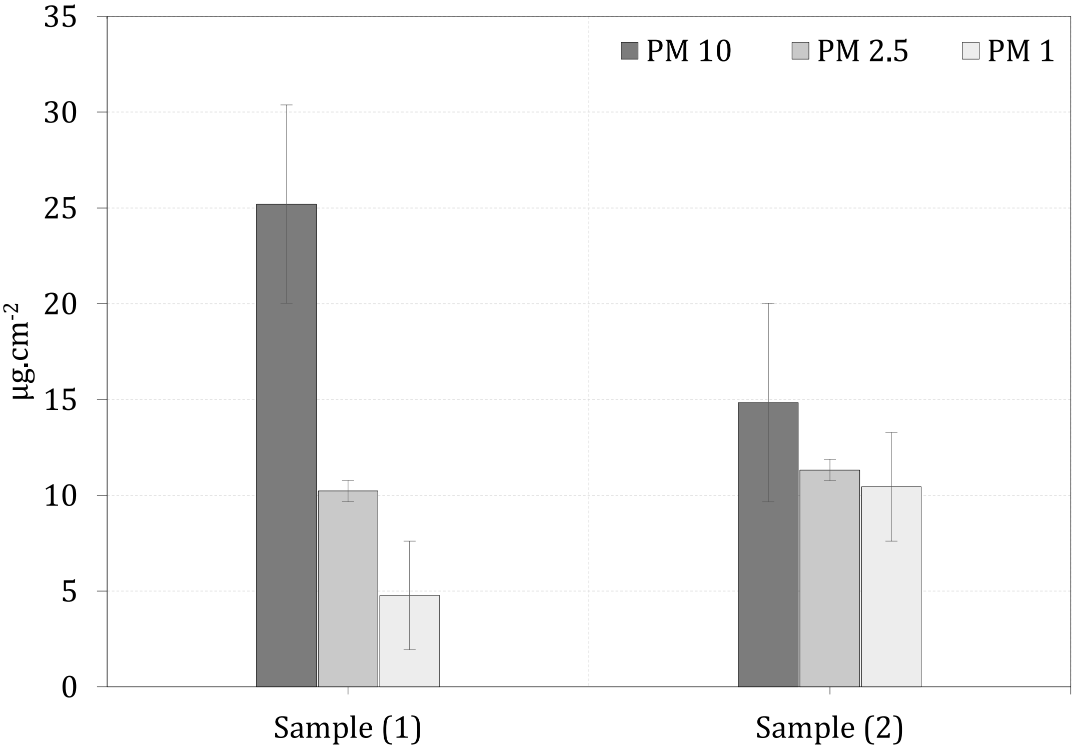


Figure VII. Accumulation of particulate matter (means± SE) ($\mu g.cm^{-2}$) within three different size fractions on leaves for Dioscorea japonica.

On the contrary, leaf surface area (LA) had no significant influence on PM accumulating process in the analyzed species. Although Cirsium vulgare has the largest LA reaching 34.39 cm^2 (average per leaf), it did not record the highest deposition of PM, in comparison between the three species, while the lowest surface area represented by Dioscorea japonica with a 1.6 cm^2(average per leaf) recorded high deposition in the three size fractions (large, coarse and fine) (Figure VII). As larger leaf area may contribute to 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 can lead to a negative correlation of the PM deposition.

On the contrary, **Weerakkody et al. [65]** noted that the small-leaved species showed comparatively low PM deposition, suggesting that this was due to their lower roughness and low capacity to with-stand PM contaminated air flow, thus lessening 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 the PM accumulation capabilities of the studied plants.

Consequently, leaves morphology 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 **[49,66].**

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 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. [67].**

Location of the analyzed species

The location of emission source may affect the quantity, size, and chemical composition of the accumulated PM. **Muñoz et al. [28] and Ottelé et al., [40]** found that plants can act as PM scavenger if it is located near an emission source, such as a main road, parking..etc. For example, large PM deposition of the analyzed shrub, may also correlated with its location which was close to two air-polluted resources (main road and car parking).

According to **Pope III & Dockery [68]**, the origin of large particles is from natural sources and anthropogenic activities, while the smallest particles mainly originate from transportation emissions, combustion, industrial processes, and photochemical reactions in the atmosphere.

The accumulated size particles on leaves of the analyzed species ranged between large and coarse particles, as selected locations are near the main road, car parking, and natural areas.

CONCLUSIONS

Shrubs can capture more particles than other analyzed species, as 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 a total of 179.02 $\mu g.cm^{-2}$ from ambient particulate matter.

Most of these particles accumulated on the plant's foliage belonged to coarse fraction size (2.5-10 μm) followed by large fraction (10 -100 μm), and finally fine fraction size (0.1 -2.5 μm). Elaeagnus pungens Thunb. captured the highest deposition of PM among the analyzed species with 83.6 $\mu g.cm^{-2}$.

Vegetation characteristics such as leaf margin and surface compositional morphology traits (roughness, grooves, and trichomes) as a taxonomic function have been addressed as significant factors associated with different PM depositions. On the other hand, leaf surface area had no direct correlation to PM accumulation.

Shrubs, climbers, and herbs, which were selected for this study, recorded high deposition of large and coarse 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 due to their smaller size, and the variable instantaneous momentum that imparts to fine particles, their movement on an erratic path known as Brownian motion (random and discontinuous movement) is the main factor to be deposited on vegetation leaves.

In conclusion, climbers' type has recorded the highest deposition of large particulate matter fraction (10-100 μm), while greatest deposition of coarse (2.5–10 μm) and fine (0.1 -2.5 μm) were captured by the selected shrub. On other hand, herbaceous plant species has the lowest portions in all quantified particulate matter.

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III-III

Efficiency of Evergreen E. Pungens Leaves as Biofilter For Ambient Particulate Matter Pollution in Summer Season

ABSTRACT

Particulate matter (PM) accumulation on leaves of Elaeagnus pungens shrub was examined. Two leaf samples were collected and analyzed using the gravimetric method. Leaf PM in different particle size fractions (PM_{10}, $PM_{2.5}$, PM_1) were accumulated effectively inside the foliage of the shrub through the experiment period of 14 days.

Total deposition of PM captured by thorny olive shrub was 83.6 $\mu g.cm^{-2}$ with the highest portion for coarse fraction (2.5 μm-10 μm) and the lowest was for the fine fraction (0.1 μm-2.5 μm).

Substantial factors for positive PM accumulation were leaf properties such as surface roughness and leaf margin. Shrubs proved to be an efficient plant species that can be used to decrease human exposure to anthropogenic pollutants in urban polluted areas.

INTRODUCTION

Particulate matter (PM) as a representative particulate pollutant is considered according to WHO, the most harmful carcinogenic pollutants affecting human health due to their small size that can penetrating the blood stream and affecting major organs in the human body.

Particulate matter is the solid or liquid atmospheric aerosol suspended in the air for long periods and transported over long distances, moreover, it includes four kinds: the total suspended particles (TSP), large particles (PM_{10}), coarse particles ($PM_{2.5}$), and fine particles (PM_1) with a diameter of ≤100μm, ≤10 μm, ≤ 2.5μm, and ≤1.0μm, respectively **[1-8].**

Several Environmental standards were established to regulate the Particulate Matter pollution globally, concerned the allowable concentration in the air and a limit on exceeding 50 $\mu g/m^3$ in an averaging period of 24 h for 35 times each year **[9, 10].**

Similarly, Japanese ministry of environment implemented emission restrictions, stated that the daily average for hourly values shall not exceed 0.10 mg $/m^3$ and hourly values shall not exceed 0.20mg/m^3**[11].**

Furthermore, various conventional complicated techniques and policies were applied to minimise the emission of Particulate matter through using different principles and methods, for instance, chemical methods which depend on wet and dry deposition.

Hence, there is a growing need for obtaining more cost-effective, simple and environmentally friendly mechanisms of Particulate matter pollution monitoring and remediation such as the environmental biotechnology mechanisms, especially, plants phytoremediation which involves transferring, removing, destroying, or stabilizing and accumulating contaminants through allowing the pollutants to adhering to the vegetation foliage and absorbing through roots **[12].**

Different studies have investigated the removal rate and resuspension of Particulate matter by plants leaves concluded that there are key factors that can enhance the PM phytoremediation through the vegetation foliage while addressing the main morphological characteristics of the leaf that can record significant results, for example, hair and wax. However, there is an absence of definitive basic data on the capturing ability of other types of leaves including leathery leaf type which was not investigated in the previous studies.

Therefore, the objective of this study is to assess the ability of Thorny olive leathery leaf to filter and capture PM, while addressing the substantial connections between PM deposition and 1) leaf morphological traits 2) leaf structure 3) plant type.

METHODS AND MATERIALS

Leaf samples and experimental site

Thorny Olive (Elaeagnus pungens) is a tough rapidly growing vine-like shrub that is resistant to drought and pollution. It has a 3-6 inches simple broadleaf evergreen with an ovate leathery hairless leaf, an alternate arrangement, and an undulate margin (North Carolina Uni. Extension Gardener Plant Toolbox, Table I.) **[13].**

It is located at Kyushu university campus (Fukuoka, Japan) (Latitude: 33°31'20" Longitude: 130°28'44'E) with a distance between the road and shrub ranging from 2 to 3 m.

The temperature was in the normal ranges of 26.8-33.8°C and the total monthly precipitation was recorded in August at 266.5mm in this location according to Japan Meteorological Agency **[14].**

Therefore, two young mature healthy leaf samples were collected from the shrub with a time gap of 10 days.

The sampling height was 1.5 m above ground level. Both leaf samples were placed in plastic bags, sealed, and labeled with numbers, and stored at 4°C in a clean laboratory refrigerator prior to analysis.

Table I. Structural characteristics of Thorny olive shrub (Elaeagnus pungens) (North Carolina Uni. Extension Gardener Plant Toolbox)

Common name	Botanical name	Leaf Margin	Leaf surface	Leaf shape	Leaf type
Thorny olive	Elaeagnus pungens	Undulate	Leathery hairless	Broadleaf ovate	Simple

Quantitative Gravimetric analysis of PM

The PM quantity was extracted using the Dzierżanowski et al. (2011) method **[15].** Both adaxial and abaxial surfaces of the leaves were washed with 250 mL distilled water (DW) for 1 min and after with 150 ml SDS (Sodium dodecyl sulfate) for 1 min.

Thereafter the resulted washing solutions were filtered through (a) polypropylene sieve (retention 106μm, Itoh seisakusho Manufacturing, Japan) (b) 10μm Omni pore membrane filter (Merck, Ireland) (c) 2.5μm glass microfiber filter (Whatman filter, cytiva, Japan) (d) 0.2μm PTFE membrane filter (Merck, Ireland), which was conducted using 25 mm vacuum filter holder with stopper support assembly connected to a vacuum pump to extract three fractions of PM: (i) 10–100 (large), (ii) 2.5–10 (coarse) and (iii) 0.2–2.5μm (fine).

The filters were dried before and after filtration for 30 min at 60 °C in the drying oven, then, stabilized in the weighing room for 60 min, and weighed using electronic balance (HT124R, Vibra, Japan).

The total leaf area of plant leaves was measured using ImageJ software, 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 morphological characteristics in the analyzed shrub and PM accumulation.

Calculations

The particulate matter quantity was measured through this equation **[15].**

$$\text{PM deposition}\left(\frac{\mu g}{cm^2}\right) = \frac{\mathbf{W}_{F+PM} - \mathbf{W}_F}{\mathbf{A}} \quad (1)$$

W_{f+PM}: Weight of filters with leaf PM deposition after filtration (µg).

W_f: Weight of blank filters before filtration (µg).

A: Total surface area of the leaf (cm^{-2}).

RESULTS

Quantitative assessment of leaf PM deposition

Significantly high amount of ambient PM was found on the foliage of the Thorny olive (Elaeagnus pungens) leaves that proved to be efficient plant at PM accumulation throughout the experiment period for 14 days.

In light of findings of Sæbø et al. **[16]**, the large size (coarse and fine) fractions represented the largest proportion of all accumulated particles**.** Similarly, in this study, coarse (2.5 -10µm) and large (10 -100 µm) fractions recorded the highest proportion for total PM deposition with 41.4µg.cm^{-2}, 22.8µg.cm^{-2}, respectively, whereas fine (0.1 -2.5µm) fraction recorded 19.3 µg.cm^{-2} (Figure I, a).

In terms of the different sampling times, Particulate matter deposition was higher in the second sample (after 14 days) for the three fractions (large, coarse and fine) than the first sample which recorded the lowest deposition of fine fraction with 4.5µg.cm^{-2}. (Figure I, b).

Furthermore, second sample recorded highest deposition of coarse fraction (2.5-10µm) followed by fine fraction (0.1 -2.5µm), and at last large fraction (10 µm -100 µm) with a percentage of 51.2%, 28% and 20.8%, respectively.

Whereas, in the first sample, likewise, coarse fraction (2.5 -10µm) recorded the highest proportion with 45.9%, followed by the large fraction (10 -100µm) with 38.7% and the lowest was the fine fraction (0.1 -2.5µm) with a percentage of 15.3%.

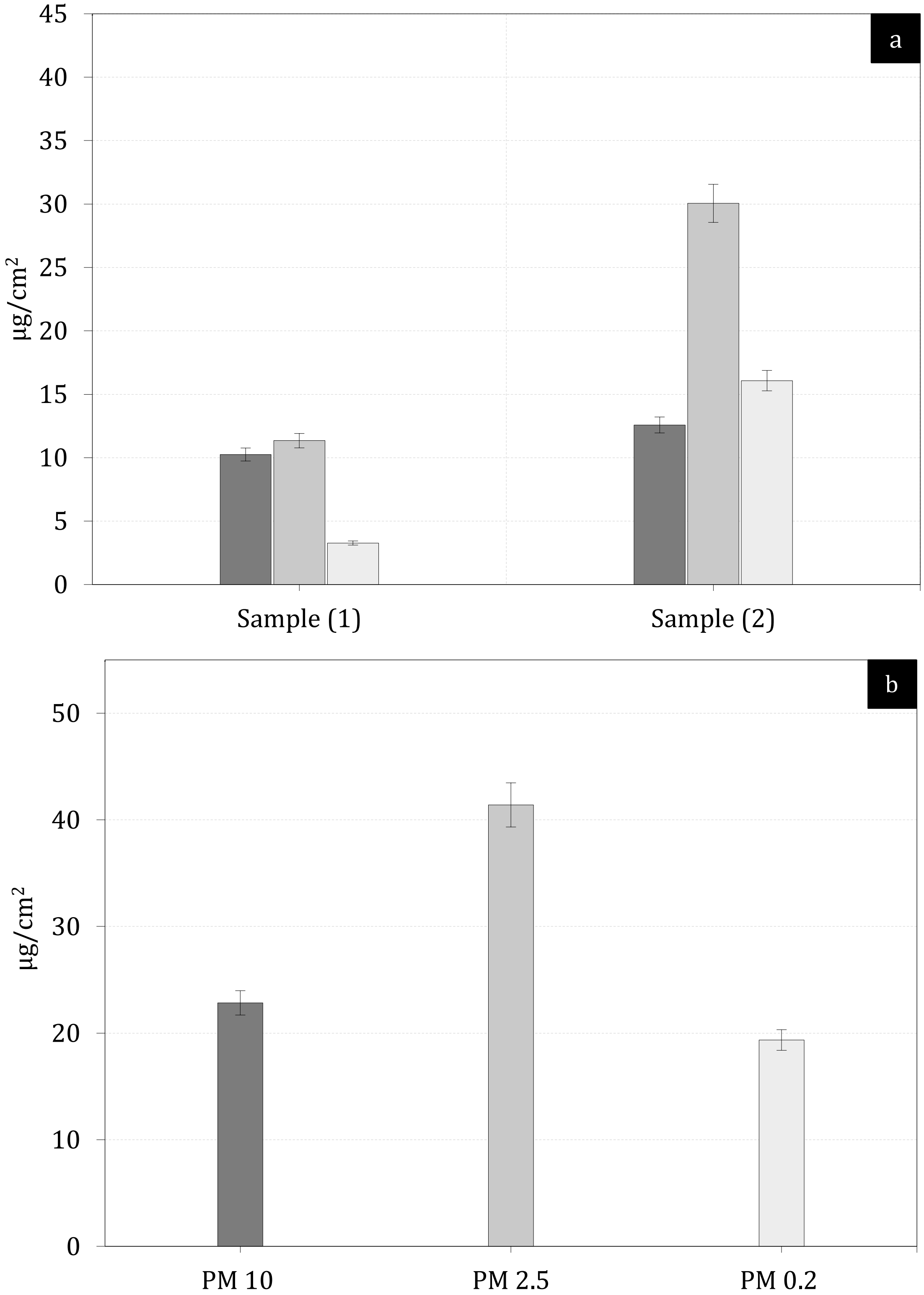

Figure I. Accumulation of particulate matter (µg.cm^{-2}) on leaves of Thorny olive (a) Total PM accumulation for different three fractions (b) represent the PM accumulation for different samples with time gap (14 days), Error bars are SD.

Association between PM deposition and leaf morphological structural traits

Morphological characteristics of leaf such as leaf margin and rough surface has been addressed as key factors associated with enhancing the PM phytoremediation phenomenon in plants **[17, 18].**

This study clearly showed that roughness and leaf margin were substantial factors that helped in capturing particulate matter in Thorny olive (Elaeagnus pungens) that has leathery rough surface (Figure II).

Leaf surface roughness can change the transition from laminar to turbulent airflow which increases the deposition velocity across it comparing to smooth leaf surface **[19].** Moreover, leaf surface roughness can create different contact angles (θ) between a water droplet and the leaf surface, as a result, the contact time will increase between rainwater and the leaf surface which will delay PM washing **[20].**

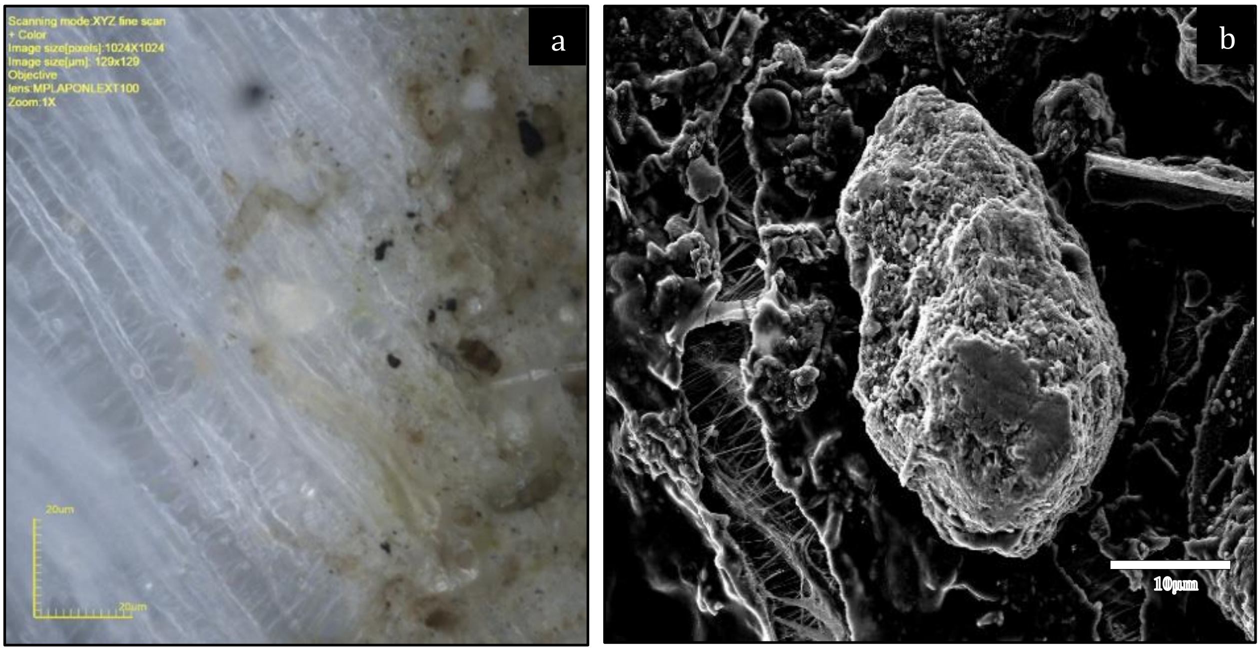

Figure II. Particle matter accumulation on Elaeagnus pungens (A) Morphological structure of the leaf surface (3D Laser Measuring Microscope -100x) (b) Scanning Electron Microscope image of particulate matter on Elaeagnus pungens filters after filtration.

Additionally, leaf margin can enhance particulate matter accumulation, Thorny olive (Elaeagnus pungens) undulate margin, with wavy shape that curves up and down, can prevent the particulate matter falling from the leaf surface, which increases the time period of PM adherence on the leaf surface.

CONCLUSIONS

Throny olive shrub (Elaeagnus pungens) can capture large-size particles (coarse and fine fraction), whereas fine fraction can be easily removed by air turbulence and precipitation which can distract the stable deposition as a result of their small size.

Morphological traits, such as leaf surface roughness and leaf margin can influence the accumulation process positively.

Total deposition of PM captured by thorny olive shrub was 83.6 µg.cm $^{-2}$, mainly, the highest proportion was for coarse fraction, followed by large fraction and finally for fine fraction.

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III-IV

Particulate Matter Removal Capability of Two Native Species in Autumn Season

ABSTRACT

To decrease particulate matter rates in urban areas, plants can be used as green purifiers for different pollutants. Two samples were collected and analyzed from Acer palmatum Thunb. and Toxicodendron succedaneum to address PM removal capability of vegetation in Autumn season.

The analyzed species were proved to capture considerable amount of PM. Trees were able to accumulate less quantity than shrubs with 60.5 $\mu g.cm^{-2}$ and 235.3 $\mu g.cm^{-2}$, respectively. The largest potion was covered by coarse size fraction (2.5-10µm), whereas fine size fraction (0.1-2.5µm) recorded the lowest portion of all deposited PM on the leaves of the analyzed species.

Both species exhibited same pattern in samples accumulation, second sample was higher than first sample. This study found than lanceolate shape and compound leaf type were key factors in enhancing PM capturing in the analyzed species, while large surface area and leaf margin which had no significant effect on PM capturing process.

Vegetation can be considered as PM scavengers if located near emission source, nevertheless, type of pollution source may affect quantity of deposited PM.

INTRODUCTION

Air pollution concentrations are dramatically increasing in urban regions. Different pollutants of air pollution cause complicated health issues and decrease the life expectancy of affected people every year according to **WHO, [1]**.

Predominantly, particulate matter which consists of heterogeneous mix of liquid and solid particles composed of different organic and inorganic compounds and originates mainly from anthropogenic activities, natural resources, transportation and industrial processes as main sources, is considered as the most harmful pollutant **[2-4].**

Long exposure to smaller diameters of particulate matter that can penetrate functional systems, may trigger chemical and biological alterations in human body, aggravating air-related diseases such as cardiac, cardiopulmonary and respiratory diseases and cause 0.8 million premature deaths yearly around the world **[5-10].**

In order to decrease toxic effects of PM in the ambient air, numerous techniques and policies were applied. Removal potential of vegetation was proved in the pervious studies (i.e. **Nowak et al., [11]; Escobedo et al. [12]; Cavanagh et al. [13]; Ottelé et al. [14]; Sternberg et al. [15]; Sæbø et al., [16]; Przybysz et al., [17]; Leonard et al., [18]).**

Through analyzing multiple trees and shrubs, **Sæbø et al. [16], Dzierżanowski and Gawroński [19] and Nawrot et al. [20],** verified the ability of large leaf surfaces species to capture PM in different seasons, especially in growing season.

Some studies such as **Przybysz et al., [17] and Pikridas et al., [21]** and confirmed their capability in winter season, when air exchange is limited and PM rates are the highest, because of extensive using of fossil fuels. However, these studies were applied to evergreen species as their foliage is available for PM deposition all year round **[22-24].**

Therefore, there is a lack in understanding of PM accumulation in deciduous species which are more exposed to lose their PM accumulation with fallen leaves. In autumn season, environmental parameters may also affect PM capturing in vegetation, nevertheless, some species can capture large quantity of particulate matter before losing their leaves.

The aim of the present study is to analyze PM removal capacity in two native deciduous species in autumn season, while considering role of leaf traits in enhancing PM accumulation.

METHODS AND MATERIALS

PM Experimental analysis

Two different species in habit and location were selected to analyze PM removal capacity in vegetation in Autumn season which applied to standard weather in Fukuoka prefecture (Figure I).

During two weeks (experimental period), the monthly mean daily temperature was in the normal ranges of 9.6-17.6°C and the c in this location according to **Japan Meteorological Agency** (Fukuoka WMO station (ID:47807)) **[25].** Twenty leaves from each species were collected, placed in paper bags in laboratory refrigerator prior to chemical analysis at 4̊C.

Figure I. Sampling locations of the analyzed species at Kyushu university in Fukuoka, Japan, (a) Toxicodendron succedaneum (b) Acer palmatum Thunb. [26]

Table. I. Characteristics of the analyzed species (North Carolina Uni. Extension Gardener Plant Toolbox)**[27]**

Common name	**Botanical name**	**Leaf Margin**	**Leaf surface**	**Leaf shape**	**Leaf type**	**Leaf Arrangement**
Japanese Maple	Acer palmatum Thunb.	Serrate	Leathery hairless	Palmasect	Simple	Opposite
Japanese Hazenoki tree	Toxicodendron succedaneum	Entire	Glossy, leathery, Hairless	Lanceolate (Palmately)	Compound	Opposite

Using gravimetric method, the collected samples were washed for 1 min by (i) deionized water and (ii) Sodium dodecyl sulfate (SDS)($CH_3(CH_2)_{11}OSO_3Na$) (Merck, Germany) for deep extraction of three fractions of PM (10–100 (large), 2.5–10 (coarse) and 0.2–2.5μm (fine).

Thereafter, turbid washed solutions were filtrated using filtration system consisted of a) Polypropylene sieve (retention 106 µm, Itoh seisa kusho Manufacturing, Japan) (b) 10 µm membrane filter (Merck millipore Ltd, Ireland) (c) 2.5 µm membrane filter (Whatman filter, Cytiva, Japan) (d) a 0.2 µm PTFE membrane filter (Merck Milli pore Ltd, Ireland), which were connected to 25 mm vacuum filter holder with stopper support assembly and a vacuum pump.

Used filters were dried before and after filtration for 30 min at 60 °C in drying chamber, stabilized in the weighing room for 60 min, and weighed using electronic balance (HT124R, ViBRA, Japan). The total leaf area was measured using ImageJ software.

CALCULATIONS

The particulate matter quantity was measured through this equation.

$$\text{PM deposition}\left(\frac{\mu g}{cm^2}\right) = \frac{W_{F+PM} - W_F}{A} \qquad (1)$$

W_{f+PM}: Weight of filters with leaf PM deposition after filtration (µg).

W_f: Weight of blank filters before filtration (µg).

A: Total surface area of the leaf (cm^2).

RESULTS

Quantitative assessment of leaf PM deposition

The analyzed plants species were able to accumulate significant quantity of PM with total of 295.8 $\mu g.cm^{-2}$, coarse size fraction (2.5-10µm) dominated the largest portion of all accumulated PM with 123.7 $\mu g.cm^{-2}$ (41.8%), followed by large size fraction (10-100µm) with 100.03 $\mu g.cm^{-2}$(33.8%).

While fine size fraction (0.1-2.5µm) recorded the lowest deposition with 72.01 $\mu g.cm^{-2}$ (24.3%) (shown in Figure II). Similar conclusions were found by **Sæbø et al.,[16]** and **Dzierżanowski et al. [28],** the large size fraction covered the largest proportion of all deposited PM. On contrary, **Freer-Smith et al., [24]** recorded substantial accretion of fine size fraction compared with fine and coarse size fractions.

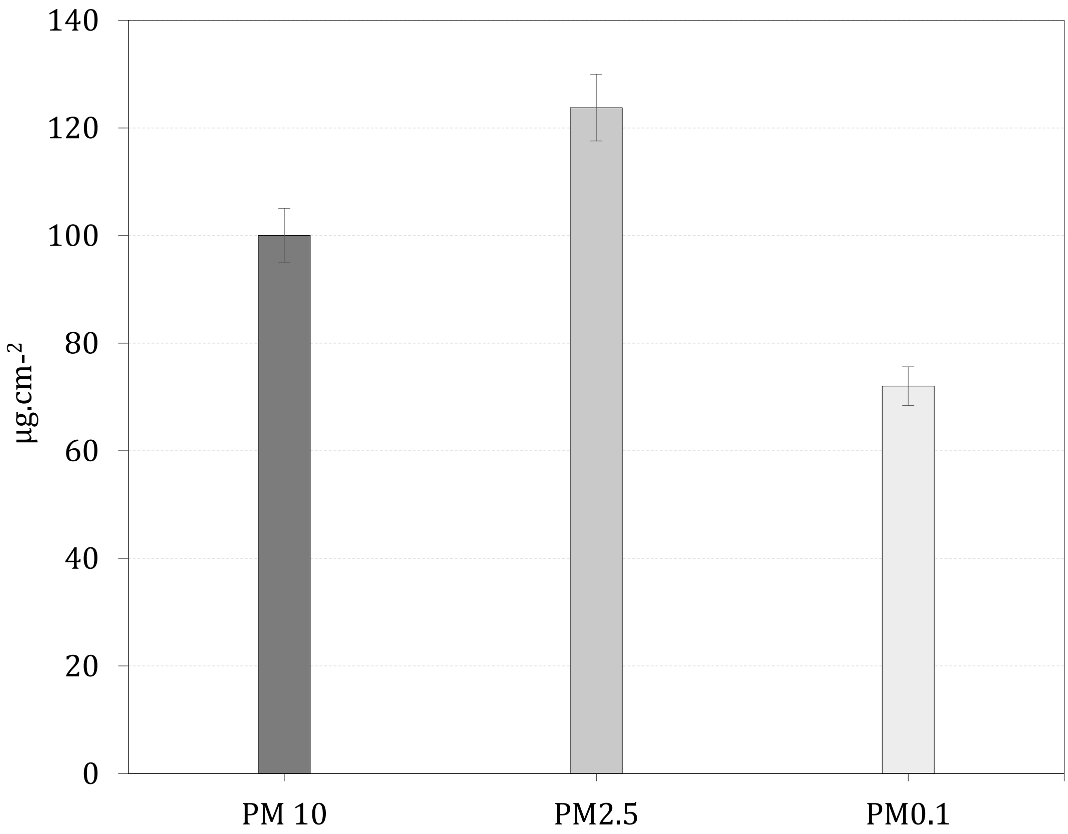

Figure II. Total PM accumulation(µg.cm^{-2}) on leaves of the analyzed species in three fractions (large, coarse, and fine), Error bars are standard errors.

Nevertheless, species differ in their capacity to remove PM from the ambient air **[28-31].** Despite the fact that trees need specific conditions such as prevailing soil conditions, space, sub-surface infrastructure and sunlight **[32].**

Trees are used as main purifiers of air pollutants in urban regions due to their large surface area and ability to increase turbulence and thus enhancing PM accumulation [33]. However, **Dzierzanowski et al. [28]** found that large trees are less efficient than low strata plantings in terms of Accumulation.

In this study, the analyzed shrub with lower leaf surface area was proved to remove more PM than trees by 235.3 µg.cm^{-2} and 60.5 µg.cm^{-2}, respectively.

The highest portion of PM was recorded by coarse size fraction (2.5-10µm) for both species with 97.6 µg.cm^{-2} (33.0%) for Toxicodendron succedaneum and 26.1µg.cm^{-2} (8.8%) for Acer palmatum Thunb. (Figure III).

The reason may related to low wash-off rates represented by shrubs which allows to retain PM permanently because of hydrophobicity of sticky or waxy leaves during precipitation events **[34].**

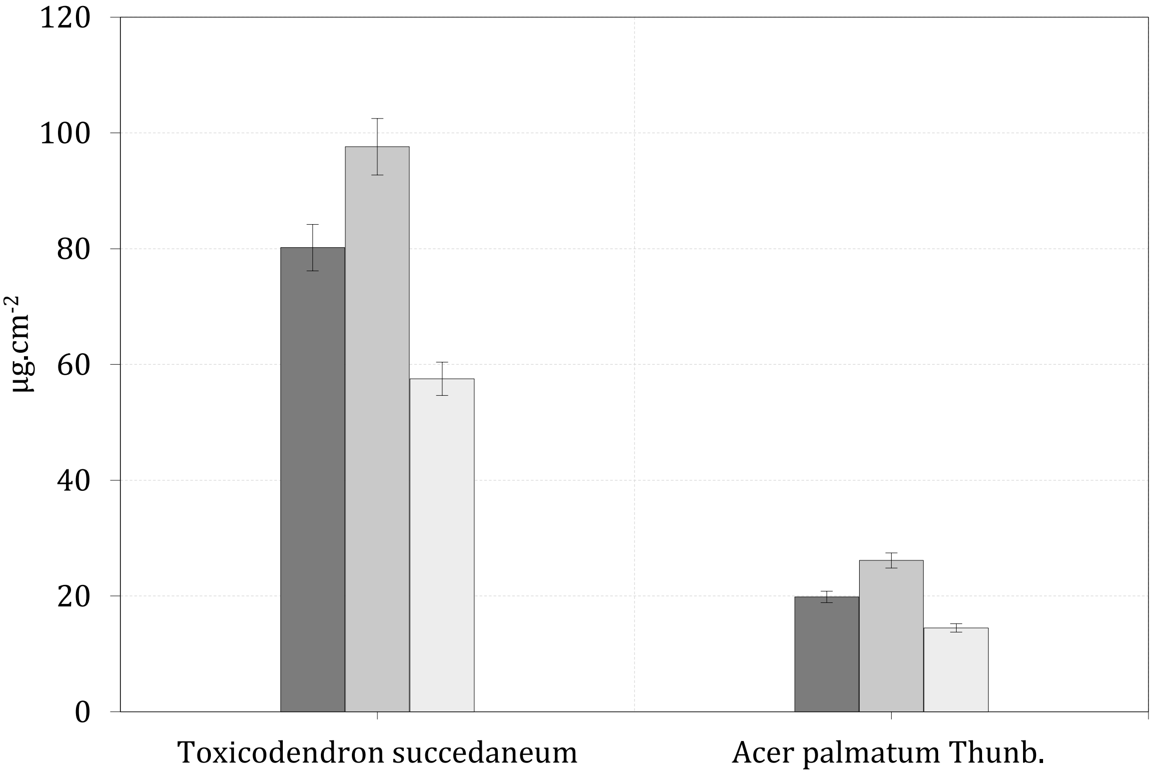

Figure III. Difference in total PM accumulation (µg.cm^{-2}) in three fractions (large, coarse and fine) among Acer palmatum Thunb. and Toxicodendron succedaneum, Error bars are standard errors (SE)

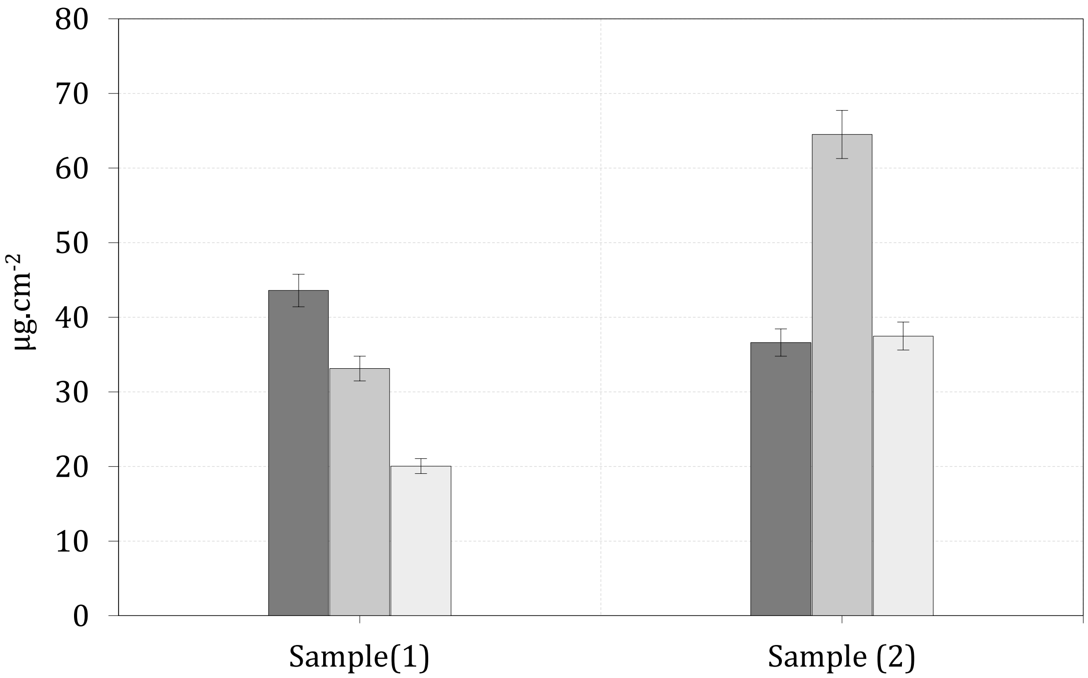

Figure IV. Total PM samples accumulation (µg.cm^{-2}) in three fractions (large, coarse and fine) among on leaves of Toxicodendron succedaneum, Error bars are standard errors (SE)

In term of different sampling time, second sample showed higher deposition than first sample of all deposited PM on leaves of Toxicodendron succedaneum with difference 41.8 μg.cm^{-2}. Large size fraction(10-100μm) recorded the largest deposition in first sample with 43.6 μg.cm^{-2}(18.5%), while coarse size fraction was the highest of second sample accumulated PM with 64.5 μg.cm^{-2} (27.4%) (Figure IV).

Acer palmatum Thunb. displayed same pattern of Toxicodendron succedaneum, second sample represented higher deposition than first sample with 40.0 μg.cm^{-2} and 20.5 μg.cm^{-2}, respectively. Coarse size fraction (2.5-10μm) was the largest portion in both samples with 10.1 μg.cm^{-2}(16.7%) for first sample and 16.1 μg.cm^{-2}(26.6%) (Figure V).

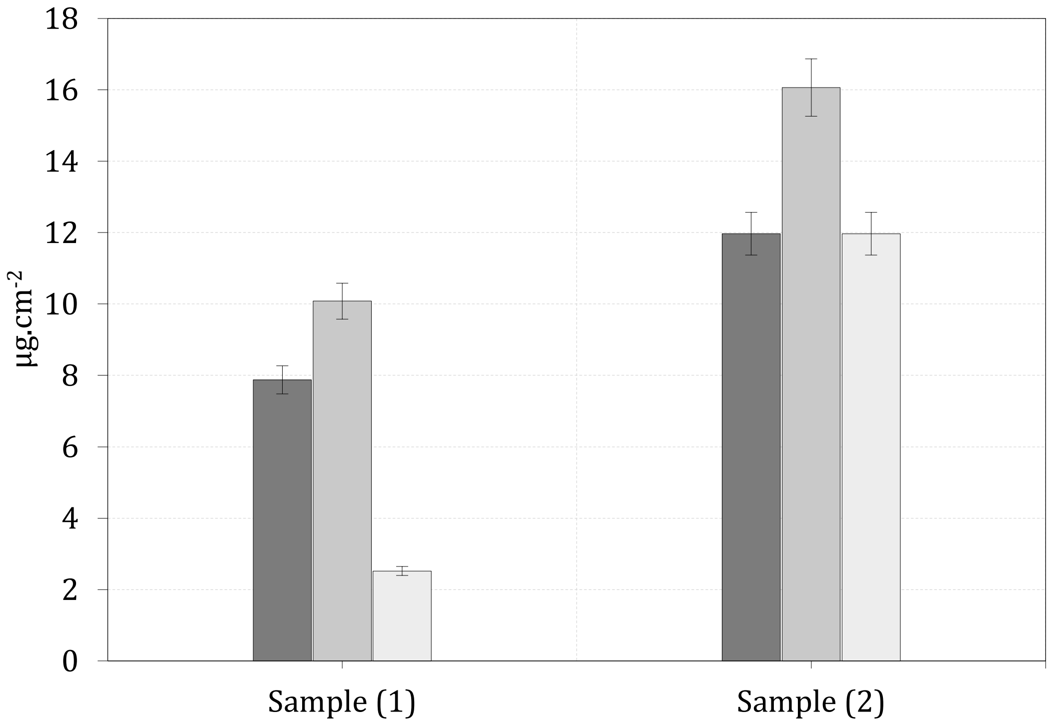

Figure V. Total PM samples accumulation (μg.cm^{-2}) in three fractions (large, coarse, and fine) among on leaves of Acer palmatum Thunb., Error bars are standard errors (SE)

Association between PM deposition and leaf morphological structural traits

There was positive relation between macrostructural traits and PM accumulation in the analyzed species. According to **Popek et al. [29]**, larger leaf area is associated with increasing PM capturing on foliage of vegetation, however, in the present study, Acer palmatum Thunb. which has larger surface area (15.9 cm^2), recorded lower deposition than Toxicodendron succedaneum with smaller surface area (5.2cm^2). It was observed by **Freer-Smith et al. [24]; Weerakkody et al. [35, 36]** that smaller sized leaves are more capable to accumulate greater PM deposition compared with large sized leaves, due to their reduced tendency to interact with air turbulence thus lowering the turbulence surrounding the leaf boundary, in contrast of large leaves that have larger boundary layer **[18].**

Leaf shape was reported to be major factor in enhancing PM accumulation, according to **Leonard et al., [18]**, lanceolate shaped leaves, which are broadest below the middle, recorded more PM than other shaped leaves, similarly, Toxicodendron succedaneum with lanceolate shaped leaves accumulated larger PM deposition than Acer palmatum Thunb. with palmasect shaped leaves.

On other hand, leaf margin was found to has no significant effect on PM accumulation process. In general, leaf margin tends to prevent washing off or resuspension of PM through weather conditions, such as air turbulence and precipitation.

An additional factor may the reason to collect more particles, compound leaf type (more than two distinct leaflets). Through weather conditions, 20% of PM may be washed off from leaves by precipitation, some particles may fall to different leaves in the same structure, which allows to keep fallen particles on leaves of the subjected species **[37].**

Location of the analyzed species and PM accumulation

The location of emission source may affect chemical composition, quantity, and size of the deposited PM. Two sites were included in the present work, near car parking and near main road. The analyzed species near car parking captured more PM deposition than near main road (shown in Figure VI). There is no difference in the accumulated PM size among the analyzed species, three different size fractions (large, coarse and fine) were accumulated on the leaves of Toxicodendron succedaneum and Acer palmatum Thunb. with higher percentage for large and coarse size fractions, while fine size fraction recorded the lowest deposition for both species. The origin of large particles is natural sources and anthropogenic activities, whereas coarse and fine particles mainly originate from transportation emissions, industrial processes, and photochemical reactions in the atmosphere **[38].**

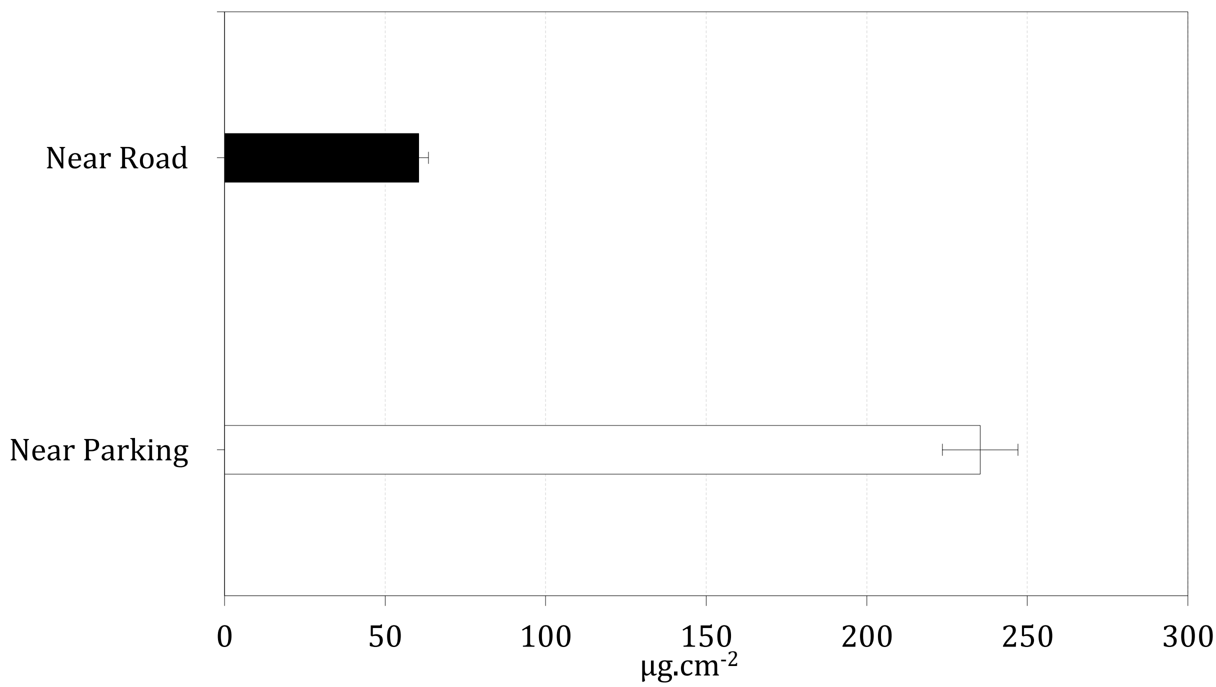

Figure VI. Difference in total PM accumulation (μg.cm^{-2}) among sites of emission sources, Error bars are standard errors (SE)

CONCLUSIONS

Vegetation in different seasons can effectively capture PM. In this study, the tested species (T. succedaneum and Acer palmatum Thunb.) accumulated significant quantity of PM in autumn season with total of 295.8 $\mu g.cm^{-2}$.

However, there is difference between species in PM removal capacity, shrubs were demonstrated to be more effective than trees. Coarse size fraction (2.5-10μm) dominated the largest portion of all deposited PM with 123.7 $\mu g.cm^{-2}$ (41.8%). While fine size fraction recorded the lowest portion with 72.01 $\mu g.cm^{-2}$(24.3%) for both species.

Leaf traits such as lanceolate shape and compound leaf type were found to enhance PM accumulation in the analyzed species. On other hand, leaf surface area and leaf margin had no significant influence. Type of emission source may affect quantity of accumulated PM, species near parking captured more particles than near main road.

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CHAPTER IV

Environmental Stress Effect on Vegetation Phytoremediation Capability

Accretion of Ambient Particulate Matter on Leaves of Arundinaria Gigantea

ABSTRACT

Different species of vegetation were demonstrated to scavenge large quantities of PM. Although their smaller leaf surface area, the analyzed grass accumulated substantial deposition of PM with 65.9 $\mu g.cm^{-2}$.

According to emission source location (main road), most of the accumulated particles were coarse and fine particles resulted from transportation, while large sized particles recorded the lowest deposition with 13.2 $\mu g.cm^{-2}$. Arundinaria gigantea showed an increase in the second sample deposition with 17.7 $\mu g.cm^{-2}$.

Leaf shape, surface area and morphology of the tested species were key factors in PM accumulation process improvement. Signs of environmental urban stress were showed on the analyzed species leaves (necrosis spots, chlorotic patches).

Further analysis should be considered concerns morphological analysis of leaves to address effects of urban stress on leaves microstructure, moreover, to address potential alterations in biological mechanisms of stressed plants.

INTRODUCTION

In urbanized areas, air pollution is considered as the main reason of plethora of different health and environmental issues. Specifically, particulate matter (PM) which varied in terms of origin, chemical composition, and size, is defined a heterogenous mixture of particles with diameter 0.001–100 µm that consist of organic and inorganic aerosol suspended in ambient air **[1,2].**

Short-term exposure to ambient PM is directly correlated with high mortality rates due to cardiovascular, respiratory and venous thromboembolic disease **[3].** Smaller sized particles in aerodynamic diameter can penetrate major organs (liver, spleen, kidney, brain) and initiate systemic inflammatory alterations, which can lead to life-threatening complications in blood coagulability due to toxic chemicals carried by the particles (e.g. polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and heavy metals) **[4-6].**

In order to eliminate PM concentrations, vegetation was demonstrated as an effective purifier to air pollutants via phytoremediation which is plant-based, environmentally friendly biotechnology to metabolize, detoxify and degrade indoor and outdoor air pollutants **[7,8].** Nevertheless, plant species differed in PM removal capacity.

Due to their large total leaf area, the architecture of their crowns resulting from the complex formation of foliage and shoot induces turbulent air movement, which positively influences PM removal capacity, trees were stated as the most efficient group for PM scavenging **[9-11].**

Woody shrubs have significant capacity to accumulate large quantities of PM, because of their low wash-off rates and hydrophobicity of sticky or waxy leaves which allows to retain PM permanently during natural rain events **[12].** However**,** large leaf surface area species such as trees, shrubs and climbers were hypothesized to increase local air pollution concentrations through reducing near-surface air exchange and pollutant dispersion **[13-16].** Low strata planting can be planted as sufficient alternative to lessen the recirculation of pollutants **[15-17].** In particular, herbaceous species that can also accumulate fallen PM of other species.

The main characteristics of the soil of polluted areas, especially close to streets and roads, are different compared with other places, physical, chemical and biological alterations were observed by **Pavao-Zuckerman, [18] and Bretzel et al., [19].** For instance, low content of organic matter and nutrients, excessive content of trace elements, correlated with structural degradation, so that vegetation in these areas are more susceptible to high degrees of environmental stress **[19].**

However, some herbaceous species can tolerate these conditions and provide a practical mitigation strategy despite the fact that they can capture less PM than other species **[20].** Pervious comparative studies that investigated native herbaceous plants are few and still there is a lack of knowledge about their PM removal capacity **[21].** Therefore, the aim of this study is to address capability of Arundinaria gigantea to remove PM while considering associations among leaf traits and PM accumulation process.

MATERIALS & METHODS

Plant types and experimental sites

Figure I. Samples site location in Kyushu university, Fukuoka, Japan **[22].**

To analyze PM deposition on the sampled leaves (two samples with time gap 14 days) from the selected species (location shown in Figure I). Both samples were washed with deionized water and SDS for 1 min each. Thereafter, resulted solutions were applied to filtration process consisted of four filtration tools (i-106 µm sieve, ii- 10 µm membrane filter, iii- 2.5 µm glass microfiber filter, iv- 0.2 µm PTFE membrane filter). Total leaf area was measured using ImageJ software. Allowing the amount of PM (in three fractions) to be expressed as $\mu g.cm^{-2}$.

CALCULATIONS

The particulate matter deposition was quantified through the following equation **[2].**

$$\textbf{PM deposition} = \frac{W_2 - W_1}{A} \qquad (1)$$

W_2: Weight the filter with the particulate matter after filtration

W_1: Weight of the filter before filtration

A: Total Surface area of leaf in cm^2.

RESULTS

Particulate matter deposition on the analyzed species

Capability of herbaceous plants was demonstrated in numerous studies**, Weber et al. [23]** found that 16 species of herbaceous plants were able to accumulate significant amount of PM from road sources. While **speak et al. [20]** quantified the annual PM_{10} removal capacity in 325 ha of green roof of herbaceous vegetation, remediation of 2.3% (±0.1%) of 9.18 tonnes PM_{10} emission in the same area. Moreover, Festuca rubra (the analyzed herbaceous species) displayed an aptitude for air pollution reduction closes to trees.

On contrary, **Currie and Bass [24]; Leonard et al. [25] and Weerakkody et al. [26, 27]** found grass species accumulated less quantity compared to trees and shrubs and **Dochinger [28]** stated that PM deposition rates of deciduous species without leaves are equal to the PM removal capacity of grass species showing low capability comparing with other species. This may related to high tendency for linear leaves ˝grass like‶ to bend easily with air turbulence due to their narrow stems or petioles, in addition to their simple leaf arrangements with larger gaps between their leaves, that may create lower turbulence around the leaf, thus lowering PM capturing levels **[27].**

The present study, grass species showed considerable PM deposition with 65.9 μg.cm^{-2}. Confirming findings of **Weber et al., [23]** who observed larger deposition of PM captured by all species on all sites were coarse size particles (2.5-10 μm). Similarly, coarse size particles (2.5-10μm) recorded the highest portion with 27.24 μg.cm^{-2}(41.3%). Followed with fine size particles (0.2-2.5μm) with 25.6 (38.7%). While the lowest deposition was recorded by large size particles (10-100μm) with 13.2 μg.cm^{-2} (19.9%) (shown in Figure II).

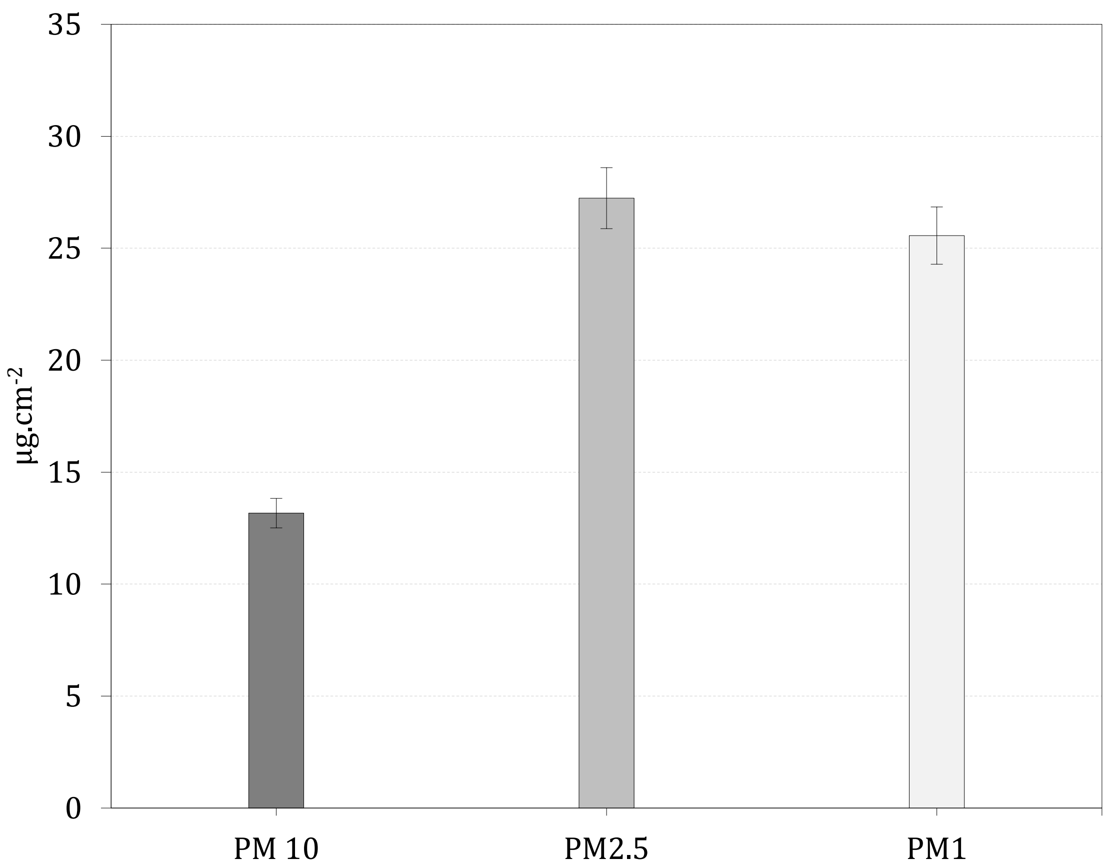

Figure II. Total PM deposition(μg.cm^{-2}) on leaves of Arundinaria gigantea, error bars are mean±SE

According to **Chow et al., [29],** the origin of coarse and fine particles is from transport emissions, combustion, and industrial processes. While large particles can originate from anthropogenic activities and natural sources. This may explain the structure of accumulated PM in the analyzed species which located near main road. In term of different sampling time, second sample recorded higher depiction than first sample with 41.9 $\mu g.cm^{-2}$ and 24.1 $\mu g.cm^{-2}$, respectively. Fine size particles (0.2-2.5μm) dominated the highest potion in all second sample deposition with 19.4 $\mu g.cm^{-2}$ (29.4%), whereas coarse size particles were the highest in first sample deposition with 12.7 $\mu g.cm^{-2}$ (19.3%).

Large size fraction (10-100μm) was the lowest deposition in both samples with 5.2 $\mu g.cm^{-2}$ (7.9%) for first sample and 7.9 $\mu g.cm^{-2}$ (12.1%) for second sample (Figure III).

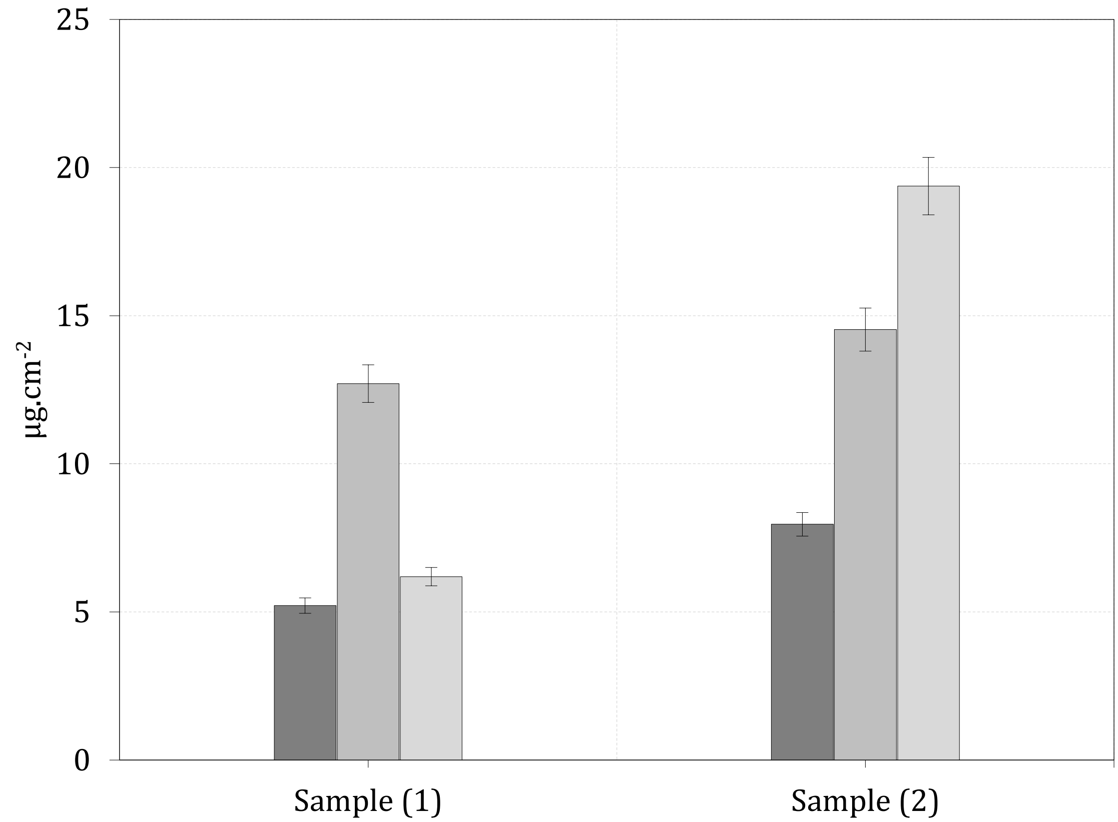

Figure III. Total PM samples deposition($\mu g.cm^{-2}$) on leaves of Arundinaria gigantea, error bars are mean±SE

Effects of leaf morphological structural traits on PM deposition

Leaf functional traits are reliable markers of PM accumulation enhancement **[8,30].** We found that the tested leaves traits such as leaf shape, rough surface, and hair presence had substantial effect on PM scavenging. Lanceolate shape was reported as the most effective shape associated with positive PM accumulation according to **Leonard et al., [25].** Rough surface and hair presence can change transition from a laminar to a turbulent airflow, moreover, velocity of PM deposition on rough surface is higher than smooth surface **[31].**

Leaf surface area had also clear effect on PM capturing process, with 15.3 cm^{-2} as an average area per leaf, PM removal capacity was improved in the analyzed species. As the long, narrow shaped leaves may be more likely hit by particles in the polluted air than large broad leaves, which have thicker boundary leaf layers (LA) (refers to air surrounding the leaf) that may contribute to increase leaf tendency to resist the deposition of particles **[25, 27, 32].**

As a result of urban stress, various changes may appear on vegetation planted in highly polluted areas. For example, PM deposited on the foliage may disturb light rays which influences chlorophyl production negatively **[33,34].** Smaller sized particles can clog the stomata, thus decreasing gas exchange **[35].** Accumulated PM on leaf can also rise leaf temperature **[36].** Furthermore, PM with pH values of ≥ 9, may trigger leaf tissues injury to leaf tissues directly or indirectly through altering the soil pH **[37]. Honour et al. [38]** found that herbaceous plants' exposure to PM pollutants may cause species-specific changes in growth and phenology, with a consistent trend of increased senescence and deferred flowering.
The analyzed species showed various symptoms of urban stress, such as loss of chlorophyll pigmentation, tip abscission and chlorotic patches and severe marginal and upper surface injury, moreover shrinkage in leaf size comparing with healthy mature leaves (Figure IV).

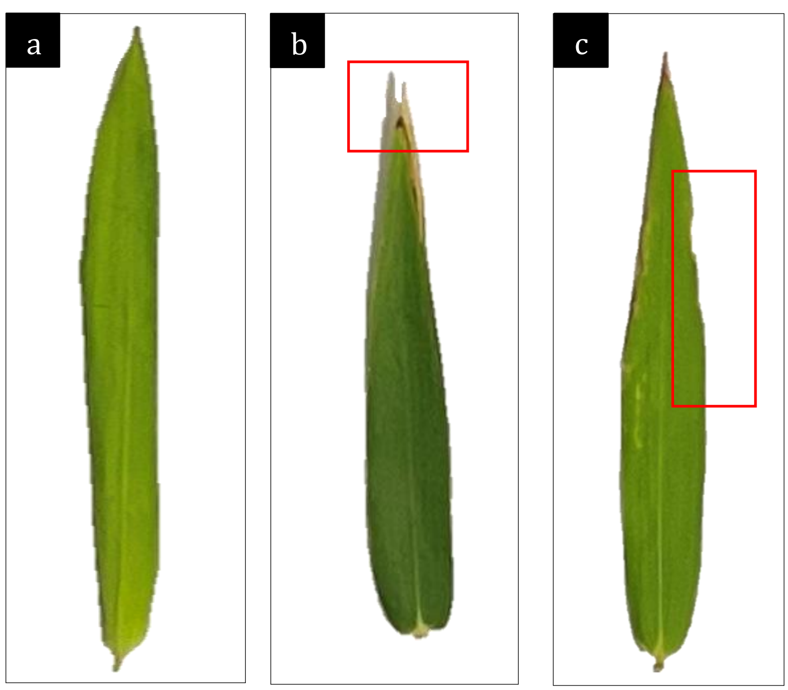

Figure IV. Environmental stress symptoms on the analyzed species leaves (a) loss of chlorophyll pigmentation, (b) tip abscission and chlorotic patches (c) Severe marginal and upper surface injury

CONCLUSIONS

Nevertheless, herbaceous species showed clear signs of urban stress such as necrosis, grasses can accumulate particulate matter significantly comparing with other species. Arundinaria gigantea accumulated total of 65.9 $\mu g.cm^{-2}$. Coarse size particles dominated the highest portion of all deposited PM with 27.2 $\mu g.cm^{-2}$ (41.3%).

While the lowest deposition was recorded by large size particles with 13.2 $\mu g.cm^{-2}$ (19.9%). The second sample recoded higher deposition than first sample with 41.9 $\mu g.cm^{-2}$ and 24.1 $\mu g.cm^{-2}$, respectively. Leaf traits such as leaf shape (lanceolate), leaf surface area, rough surface, and hair presence had significant influence on PM accumulation process.

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CHAPTER V

CONCLUSIONS

In our first paper, we found that PM scavenging was effectual with large surface area and smaller sized rough leaves vegetation. Nevertheless, weather factors may influence PM quantity. This suggests changes in planting and protection strategies to enhance remediation ability.

Further analysis should be included concern morphological structure and chemical composition of deposited PM on different types of vegetation.

In our second topic, we have demonstrated that Autumn season was more effective than summer season to accumulate significant deposition of PM. Moreover, low strata plantings such as shrubs and climbers may accumulate more PM than trees due to higher capability to capture particles from different emission sources such as air and soil. Larger sized particles dominated the highest potion of accumulated PM.

Addressed results can be used in various applications and studies investigating suitable plants species in urban polluted areas to achieve maximum phytoremediation rates.

In our third topic, our findings indicate that besides accumulated PM that can affect plants remediation capability. As PM can cause chemical, biological, and structural alterations in stressed plants and some species showed substantial tolerance whereas, urban stress symptoms (necrosis and chlorosis) were clear on other plants.

There were additional factors included, environmental, structural, and spatial conditions. To illustrate, emission source type and location may influence size and quantity of accumulated PM inside vegetation foliage.

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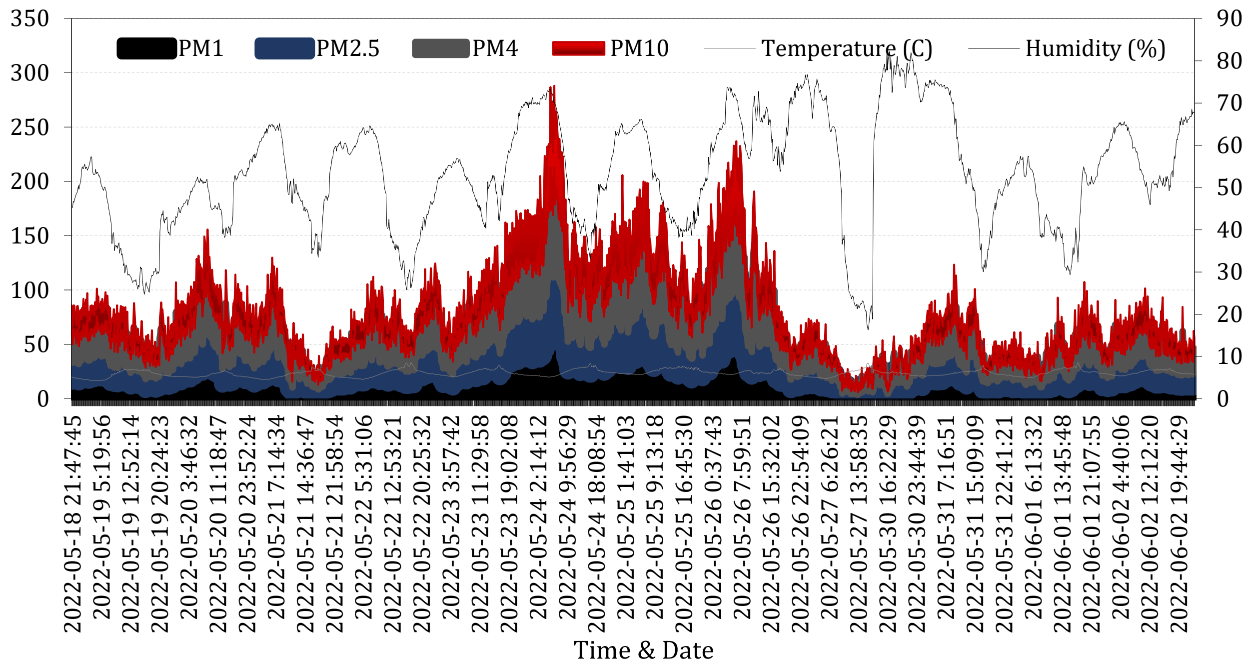
APPENDIX A

Environmental parameters in sampling sites

➢ **Spring season**



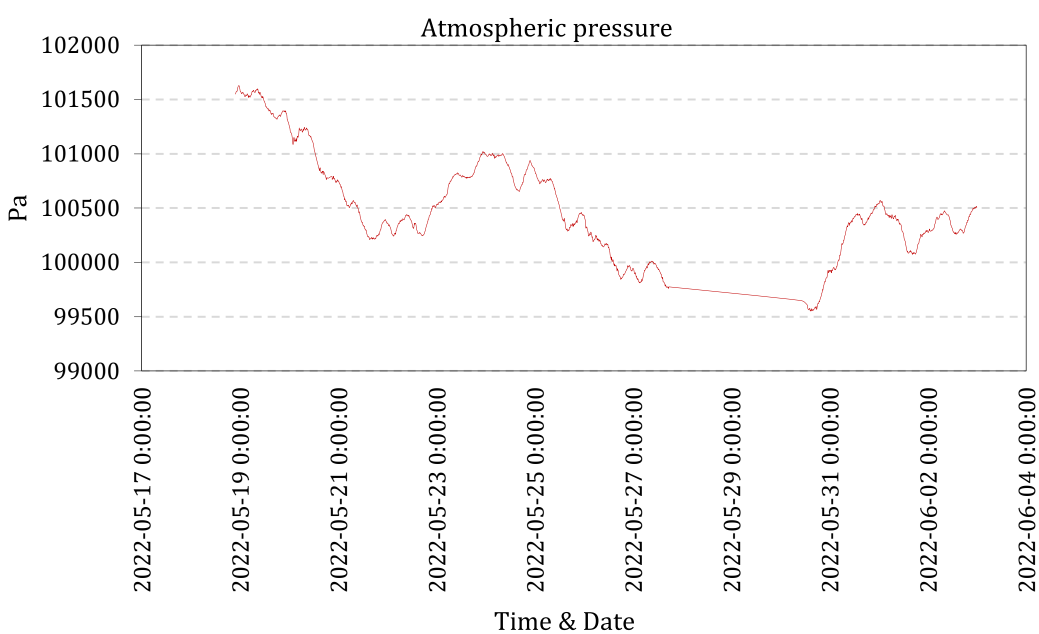
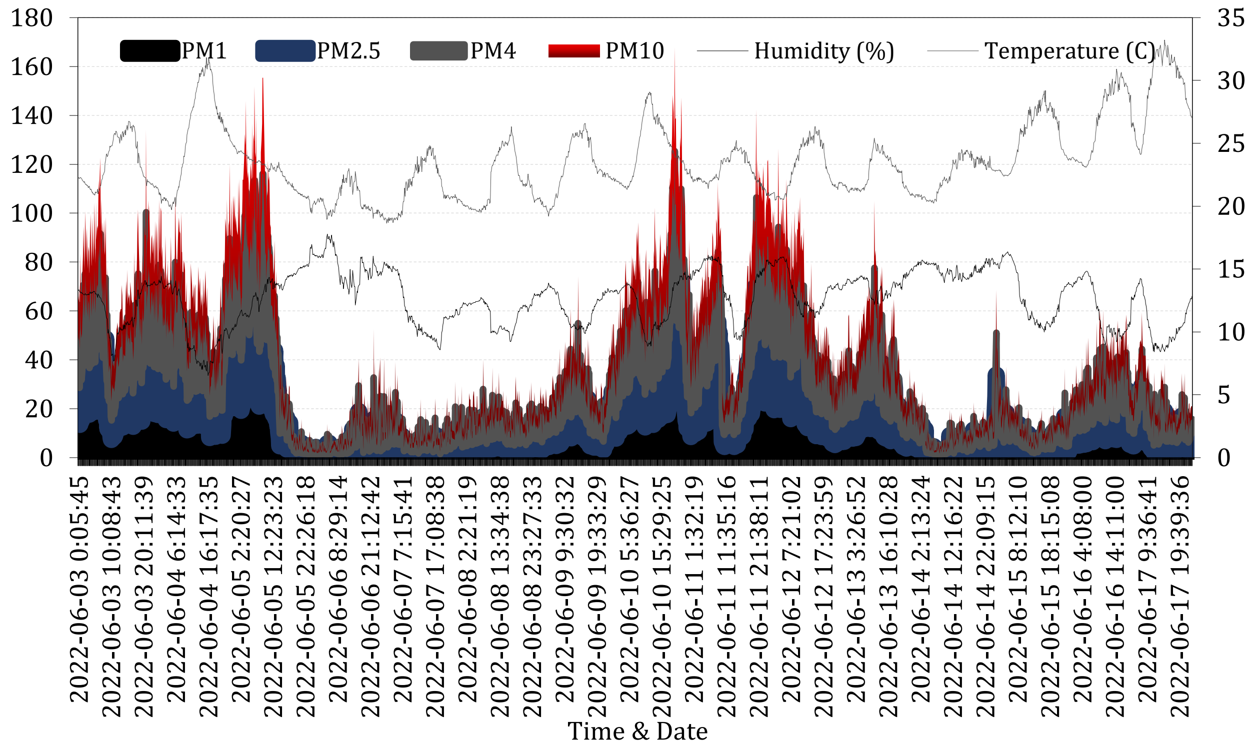


Figure I. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} (µg. m^{-3}), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 18th May till 2nd June 2022.

- **Summer season**



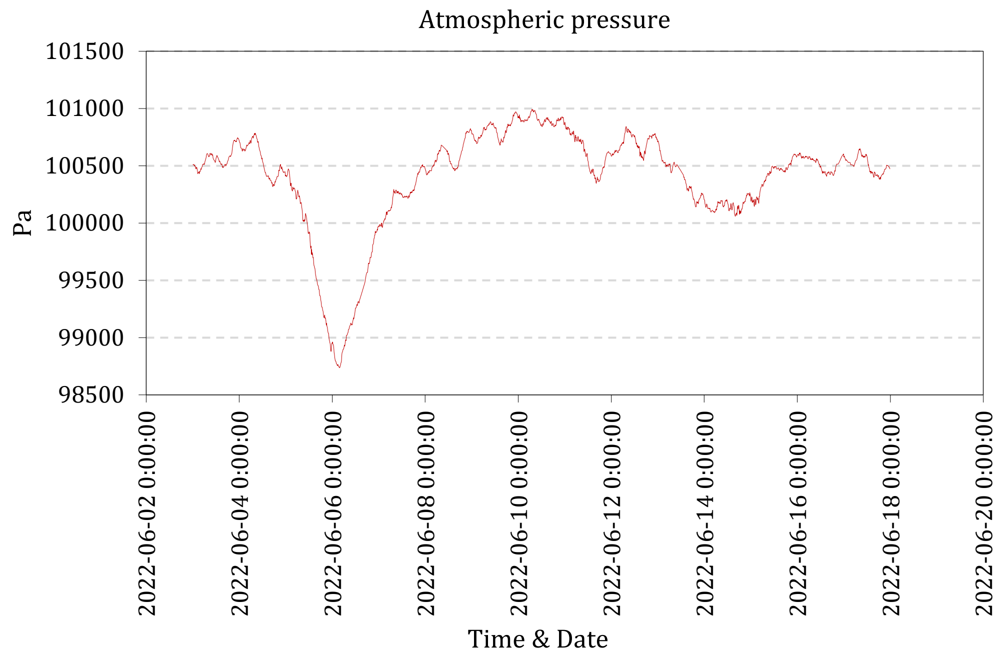
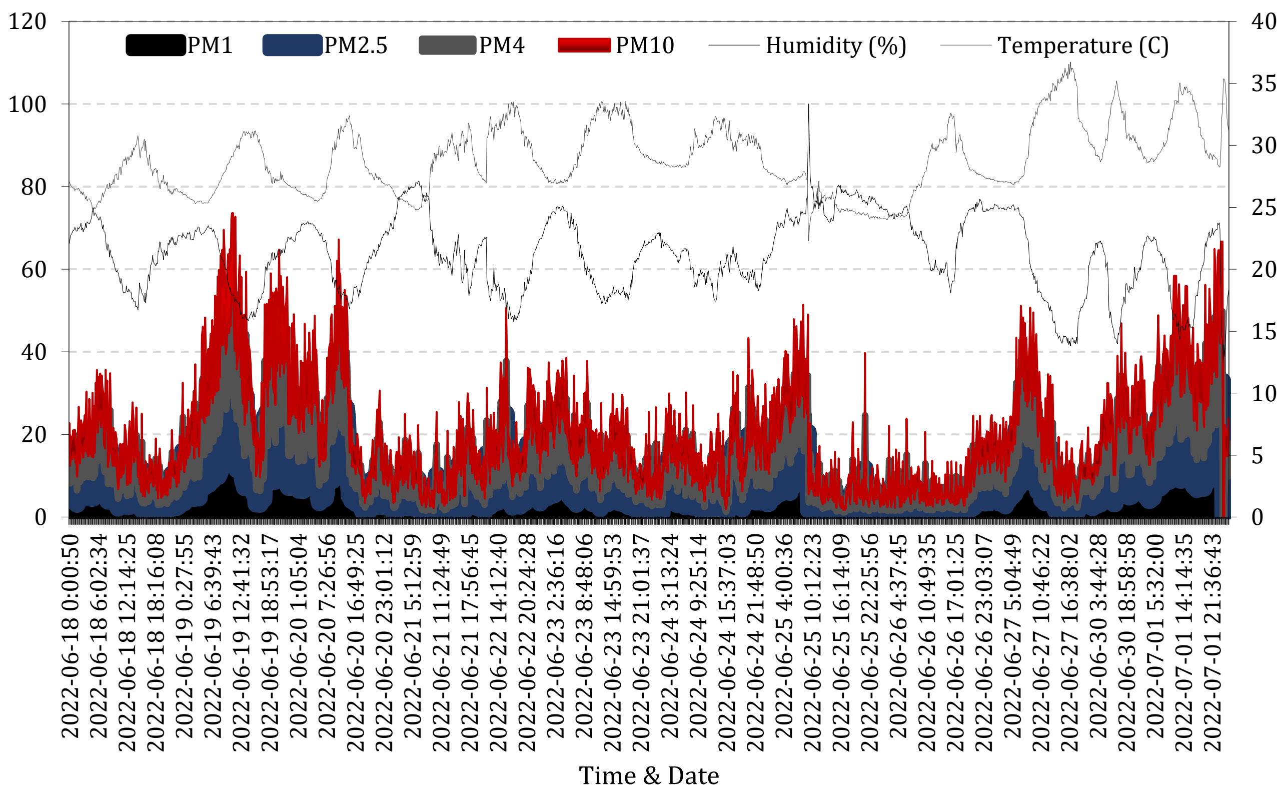


Figure II. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} (µg. m^{-3}), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 3rd June till 17th June 2022.



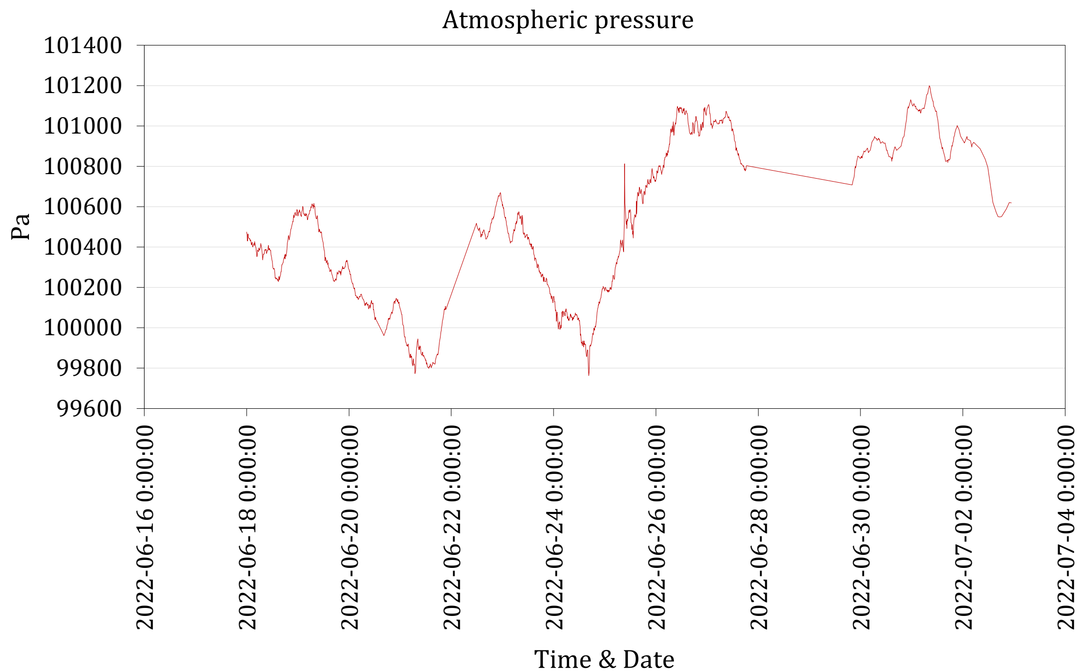
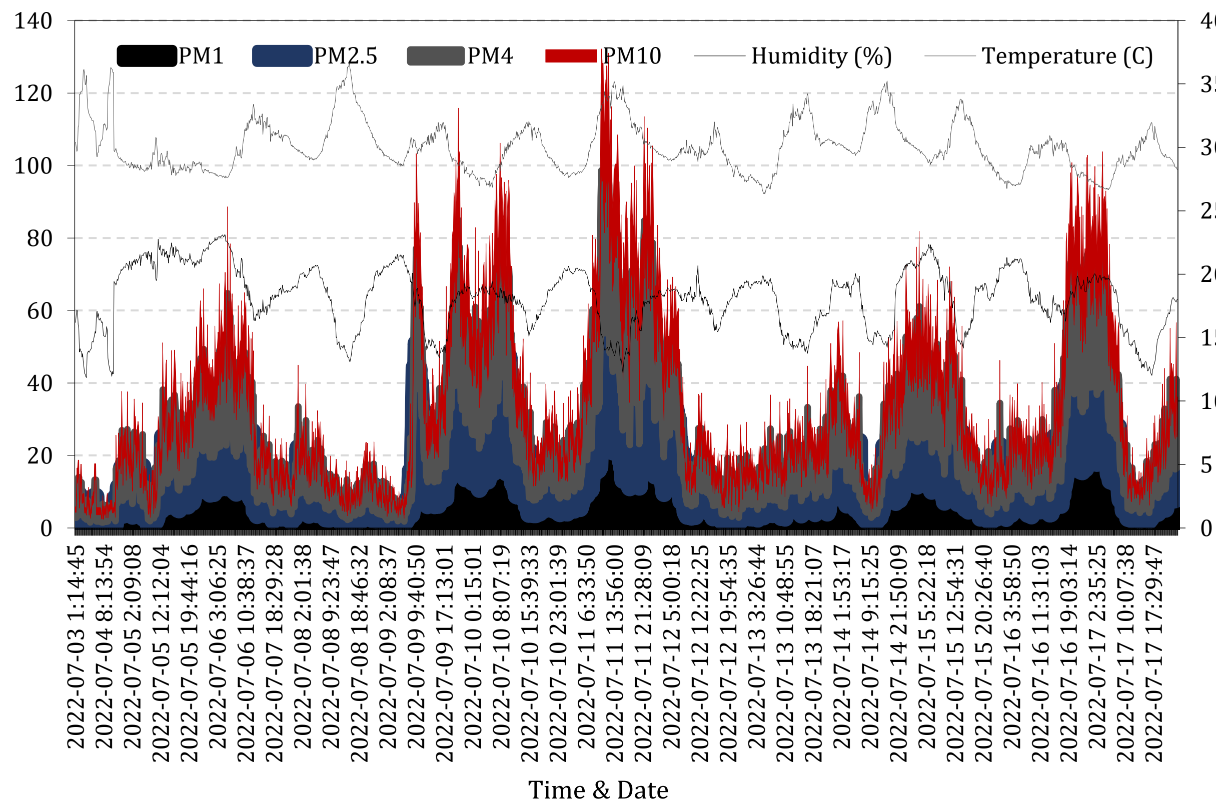


Figure III. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} (μg. m^{-3}), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 18th June till 1st July 2022.



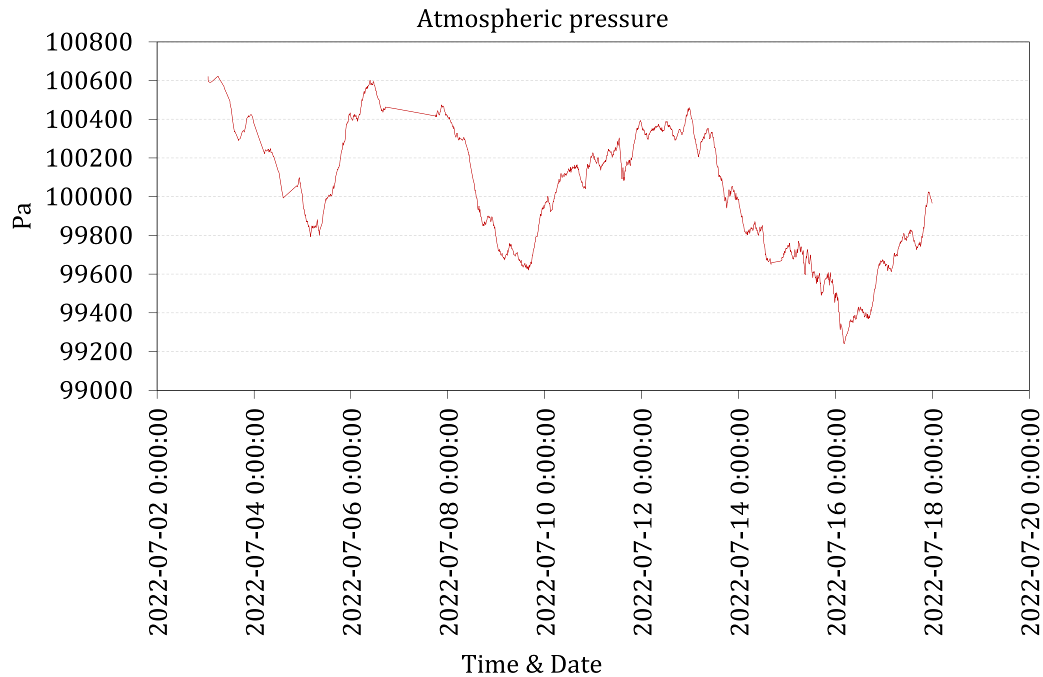
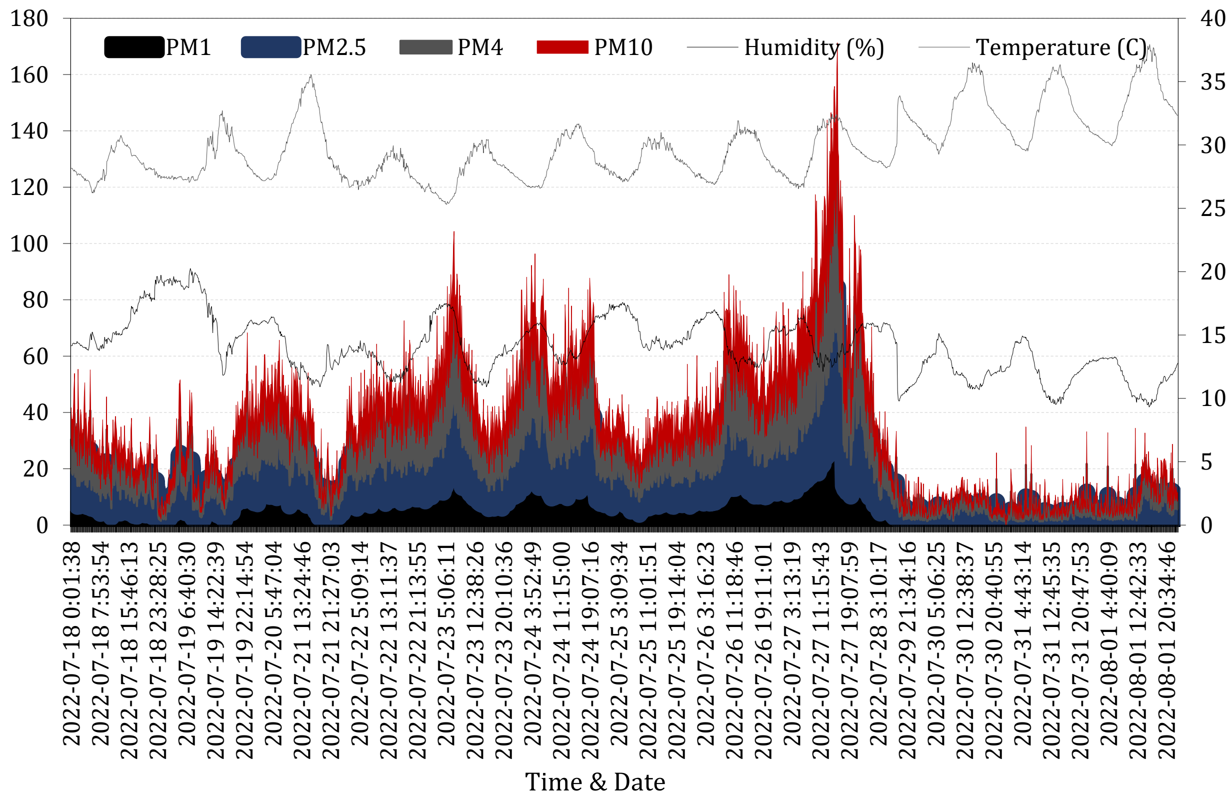


Figure IV. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} ($\mu g.\ m^{-3}$), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 3rd July till 17th July 2022.



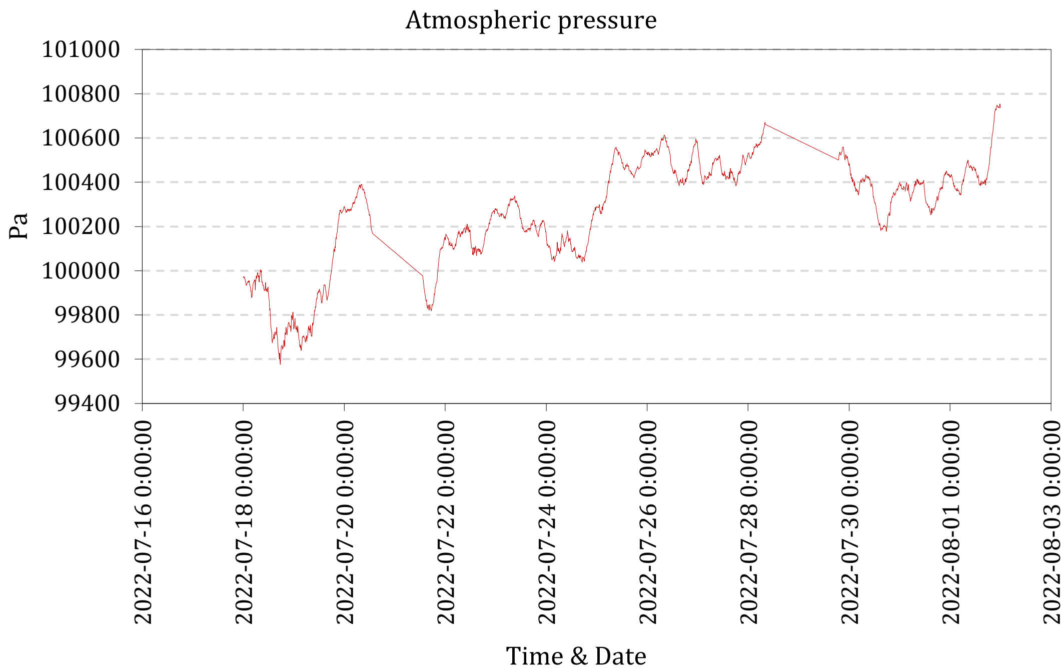
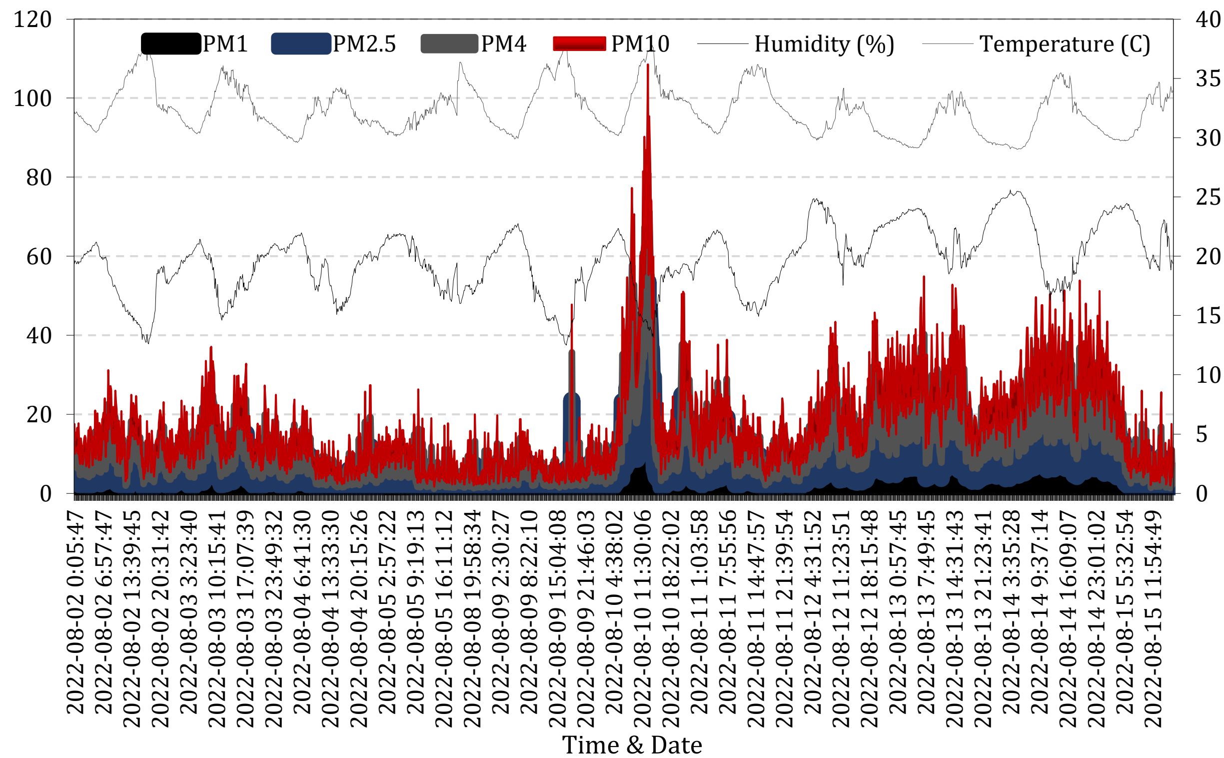


Figure V. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} ($\mu g.\ m^{-3}$), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 18th July till 1st August 2022.



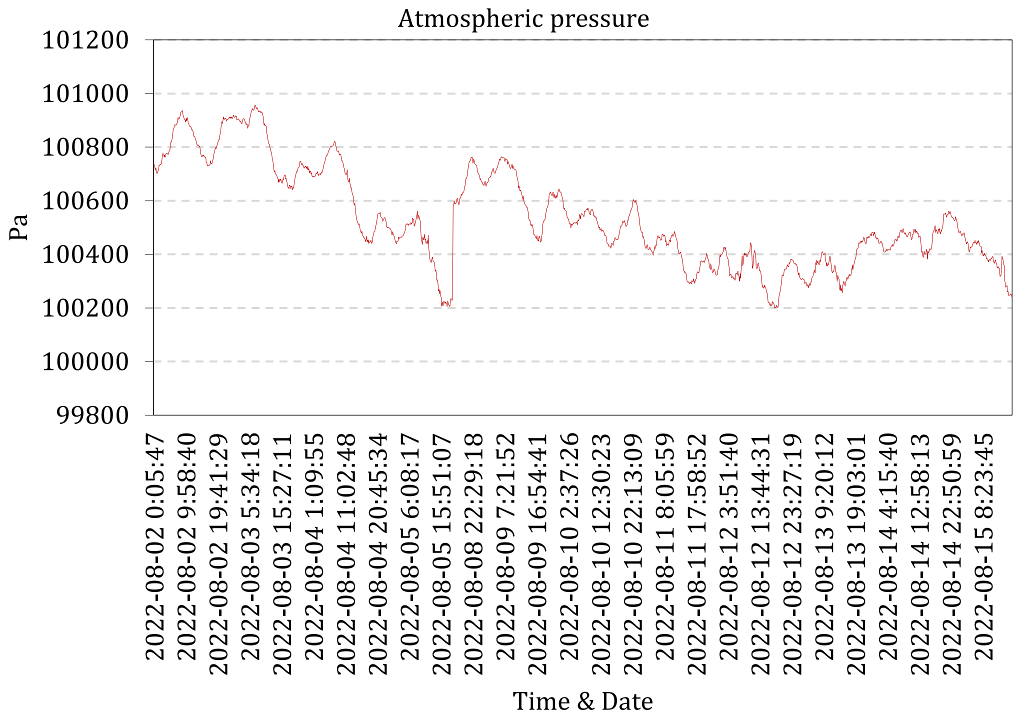
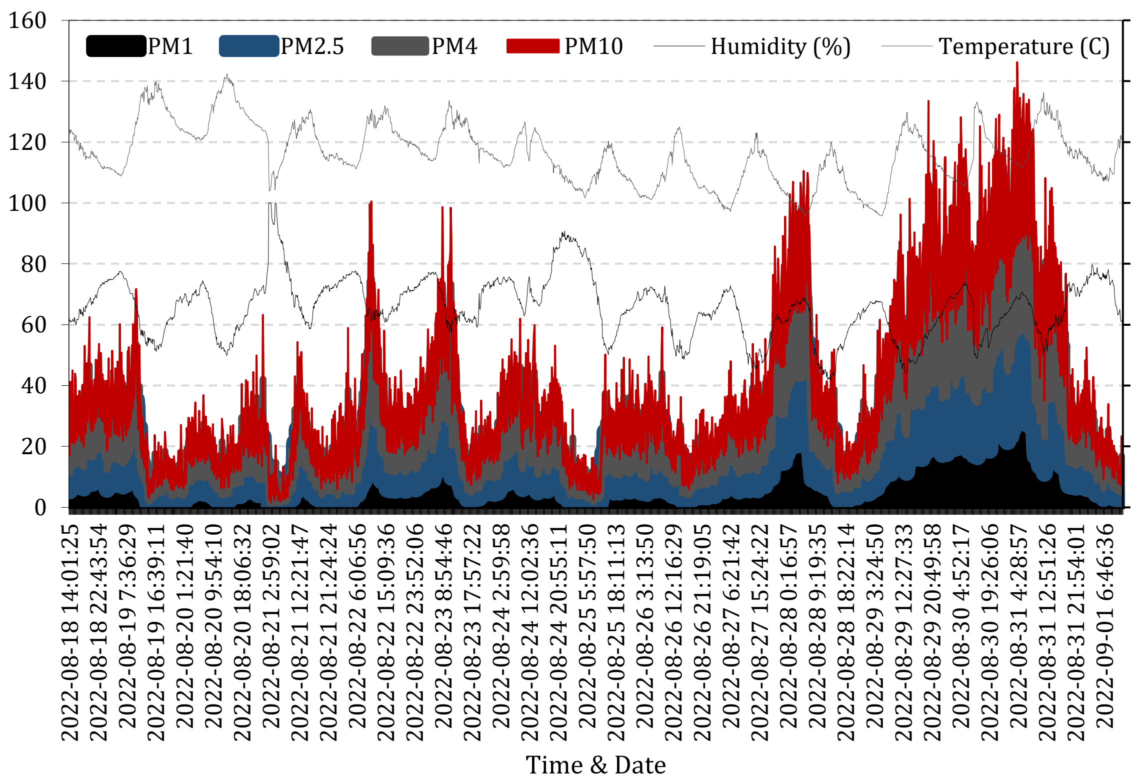


Figure VI. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} (μg. m^{-3}), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 2nd August till 15th August 2022.



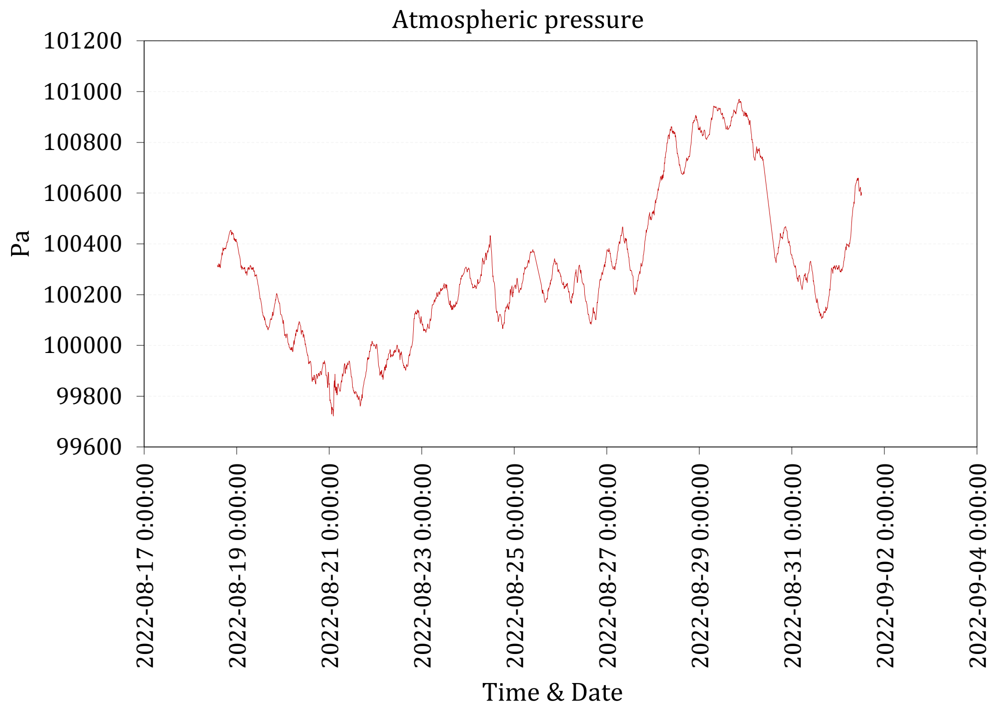


Figure VII. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} (µg. m^{-3}), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 17^{th} August till 1^{st} September 2022.

- **Autumn season**

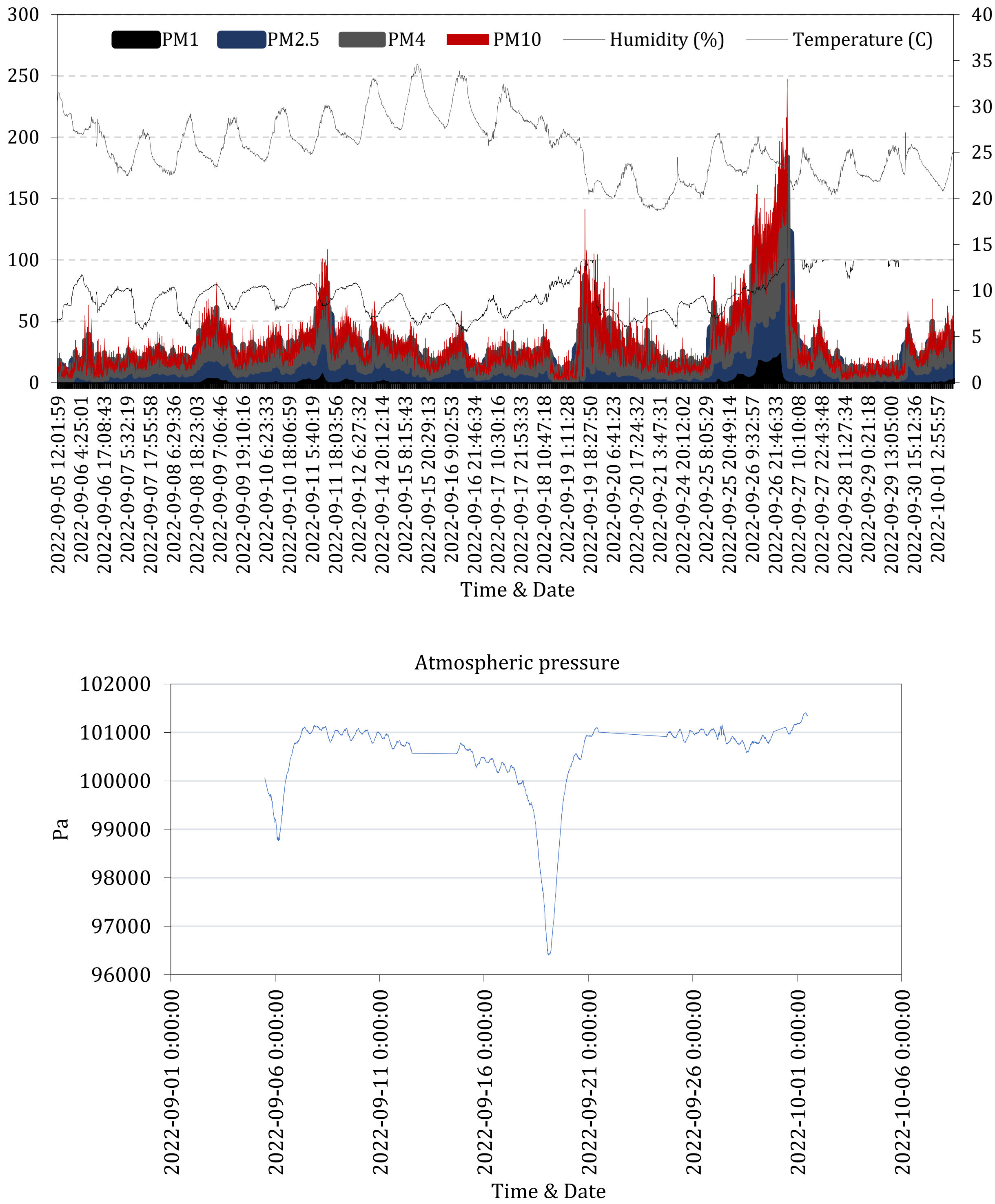


Figure VIII. Daily concentrations of PM_1, $PM_{2.5}$, PM_4 and PM_{10} (µg. m^{-3}), temperature (C), humidity (%) and pressure (Pa) from data sheet sensor illustrated from 5th September till 1st October 2022.