THE IMPACT OF LIVING WALLS IN THE REDUCTION OF ATMOSPHERIC PARTICULATE MATTER POLLUTION

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A thesis submitted in partial fulfilment of the requirement of Staffordshire University for the degree of Doctor of Philosophy

May 2018

I dedicate this thesis to my loving parents who have been a great inspiration throughout this challenging journey

"When you catch a glimpse of your potential, that's when passion is born."

— Zig Ziglar

"When trees burn, they leave the smell of heartbreak in the air."

— Jodi Thomas

Abstract

Traffic-generated particulate matter (PM) air pollution remains a serious threat to human health and the environment. Whilst the ability of vegetation to reduce PM pollution is widely recognised, little or no information is available on the value of living walls in this respect. This thesis explored their potential to serve as near-road/rail PM filters, and evaluated the optimal conditions required to maximise their PM capture efficiency, using both field and laboratory-based techniques. This study revealed living walls could immobilise substantial levels of PM associated with both road and rail traffic. Of three living walls studied, one, located along a busy road, was shown to remove substantial levels of PM_1 ($122.08 \pm 6.9 \times 10^7$) and PM_{10} ($4.45 \pm 0.33 \times 10^7$) per 100 cm^2; the highest level of $PM_{2.5}$ removal ($9.9 \pm 5 \times 10^7$) was recorded from a wall located near a busy train station. The best PM-removing species, and the important leaf traits, were identified using living wall plants and natural/synthetic leaf models. There was a considerable inter-species variation in PM accumulation and the PM densities on some species were x50 and x65 higher compared to others. The influence of leaf size on PM accumulation was found to be dominant over other examined characters (leaf size, shape, and micromorphology); smaller leaved species with a high Leaf Area Index were identified as the most efficacious. Analysis of captured PM from rail and road traffic showed the ability of living wall plants to immobilise a wide range of elements which are known to be hazardous to health. Simulated rainfall demonstrated the potential for rain to wash particulates off leaves and renew capture surfaces to ensure their continuous functioning; wash-off removed between 48.4% and 92.5% of particles depending on the species. An impact of planting design on the PM reduction performance of living wall species was demonstrated by the superior capture efficiency resulting from increasing the topographical heterogeneity of plantings. This research enhances our understanding of the benefits of living wall systems in relation to PM pollution mitigation, and consequent improvement of human wellbeing.

Acknowledgements

First and foremost my heartfelt gratitude goes to my supervisors for their remarkable support; I could not have wished for a better supervisory team to complete my PhD research. I would like to express my sincere gratitude to Professor John Dover, former principal supervisor and current external supervisor, for his tremendous support, invaluable comments and guidance. I would not have come this far without his constant encouragement, enthusiasm, patience and flexibility as an advisor, while also providing me with opportunities to enhance my skills and expertise in the field. I would like to express my heartfelt gratitude to my principal supervisor Dr. Paul Mitchell, for his untiring guidance, insightful comments and constant support throughout this thesis. His instructions, enthusiasm and constant support stimulated and inspired me to continue this challenging journey with determination. My sincere gratitude also goes to Dr Kevin Reiling, my second supervisor for all his immense guidance and motivation to steer me in the right direction in this challenging research drive. His invaluable suggestions widened my research from various perspectives.

I gratefully acknowledge Dr. Ruth Swetnam, for her valuable instructions given in leaf-morphology analysis using GIS, and Dr Dave Skingsley for his support in chemical analysis trials in FTIR spectroscopy. I am extremely grateful to Nemec Cascade Garden Ltd., Czech Republic, for donating a living wall system for this study, especially to Benjamin Raich, Project Manager, for his enthusiastic support. I gratefully acknowledge Network Rail, New Street Station, Birmingham, UK for providing permission to sample plant material from the living wall managed by them. I am also grateful to Kevin Hobbs, Head of Research and Development, Hillier Nurseries Ltd., Romsey for donating some plants, specially propagated for this research and to Barry Clarke, Hillier Nurseries Ltd. for help identifying the species. The help of James Bentley, and Adam Van Winsum, from Staffordshire University Estates are gratefully acknowledged for their support in maintaining living walls used in this study. Dr Md Asaduzaman, Joy Harding, Dhammika Sriyananda and Dr Sameera Viswakula are greatly acknowledged for their valuable support given for the statistical analysis of data.

I am extremely grateful to Dr Alison Davidson, Caroline Newman, Ian Hopkins, Rosie Duncan, Peter Martin and the technical skills specialists' team for providing technical support for this research and for their enthusiastic support in various other means. The enthusiastic support received from Christine Dover and Sean Farrel, to create industry and media links to this research via Green Wall Centre was inspiring and greatly acknowledged. A very big thank you to my fellow PhD colleagues, specially, Eleanor Atkins, Dilhani De Silva, Esme Hookway, Elecia Henry, Simon Phillips, Sara Behforootan, Soheil Komilian and Mark Broadhead, and to my dear friends Vindhya de Silva and Nimashi Fernando for their invaluable support in lab/field work and for being with me in all the ups and downs faced throughout this journey.

Finally, yet importantly, my heartfelt gratitude goes to my beloved parents, sisters and brother for supporting me emotionally and financially throughout this most challenging journey; without their support this project would have not reached fruition.

Table of content

List of Figures

List of Tables

Abbreviations

AQMA	Air Quality Measured Areas
BC	Black Carbon
BSE	Back Scattered Electrons
CE	Cleansing Efficiency
CiTTyCAT	Cambridge Tropospheric Trajectory model of Chemistry and Transport
COPD	Chronic Obstructive Pulmonary Disease
CLRTAP	Convention on Long-range Transboundary Air Pollution
CTAG	Comprehensive Turbulent Aerosol Dynamics and Gas chemistry
DCP	Drop Contact Angle
DEFRA	Department for Environment, Food and Rural Affairs
DfT	Department for Transport
Dp	Particle diameter
EC	Elemental Carbon
EDX	Energy Dispersive X-ray
EEA	European Environment Agency
ESEM	Environmental Scanning Electron Microscopes
EU	European Union
FTIR	Fourier-Transform Infrared Spectroscopy
GI	Green Infrastructure
GIS	Geographical Information System
GLA	Greater London Authority
GLM	Generalised Linear Model
GLMM	Generalised Linear Mixed-effect Model
HDPE	High Density Polyethylene
IARC	International Agency for Research on Cancer
INCA	Integrated Calibration and Application
IPCC	Intergovernmental Panel for Climate Change
LAI	Leaf Area Index
LES	Large Eddy Simulation
LM	Linear Model
LV	Low Vacuum
MCI	Mild Cognitive Impairment
MISCAM	Micro Scale Air Pollution Model

MVF	Microvascular Function
OC	Organic Carbon
OECD	Organisation for Economic Cooperation and Development
PAC	Particulate Abatement Capacity
PAH	Polycyclic Aromatic Hydrocarbons
PCB	Polychlorinated Biphenyl
PCDD/F	Polychlorinated dibenzo-p-dioxins and dibenzofurans
PE	Polyethylene
PM	Particulate Matter
PNC	Particle Number Concentration
RCP	Royal College of Physicians
RCPCH	Royal College of Paediatrics and Child Health
SE	Secondary Electron
SEM	Scanning Electron Microscope
SIDS	Sudden Infant Death Syndrome
SIRM	Saturation Isothermal Remanent Magnetization
SQL	Structured Query Language
TSP	Total Suspended Particles
UFORE	Urban Forest Effects Model
UNECE	United Nations Economic Commission for Europe
UPVC	Unplasticised Polyvinyl Chloride
Vds	Deposition Velocity
VGS	Vertical Greenery Systems
VOC	Volatile Organic Compound
WHO	World Health Organisation

Peer-reviewed publications produced from this research

Weerakkody, U., Dover, J.W., Mitchell, P., Reiling, K. (2017). Particulate matter pollution capture by leaves of seventeen living wall species with special reference to rail-traffic at a metropolitan station. *Urban Forestry and Urban Greening*. 27:173-186.

Weerakkody, U., Dover, J.W., Mitchell, P., Reiling, K., (2018). Evaluating the impact of individual leaf traits on atmospheric particulate matter accumulation using natural and synthetic leaves. *Urban Forestry and Urban Greening*. 30: 98-107

Weerakkody, U., Dover, J.W., Mitchell, P., Reiling, K., (2018). Quantification of the traffic-generated particulate matter capture by plant species in a living wall and evaluation of the important leaf characteristics. *Science of the Total Environment.* 635:1012-1024

Weerakkody, U., Dover, J.W., Mitchell, P., Reiling, K., (2018). The impact of rainfall in remobilising particulate matter accumulated on leaves of evergreen species grown on a green screen and a living wall. Submitted to *Urban Forestry and Urban Greening* (in press).

The co-authors of these articles were supervisors of this reseacrh project and contributed via supervisory guidance and proof-reading.

Structure of Thesis

This document has been structured such that the findings of the research are presented in a format suitable for publication in internationally peer-reviewed journals, with slight modifications to align with a thesis structure, while still complying with university research degree regulations. Chapter 1 is the introductory chapter which comprises literature review, rationale for the research, and aims of the thesis. Chapter 2 briefly outline the generic methods used in this research and Chapter 3 details a pilot study conducted to optimise the methods. Chapter 4 and Chapter 5 are written based on the experiments which explored the potential for living walls to immobilise PM associated with rail and road traffic respectively. Some findings on the impact of leaf morphological traits on reduction of PM discussed in Chapters 4 and 5 were further evaluated in Chapter 6 based on the experiments used manipulative experimental designs. Chapter 7 evaluates the ability of rainfall to remobilise captured PM, to understand their continuous functioning as PM traps. Overall findings and their significance are then discussed in Chapter 8 reflecting upon the aims and objectives of this thesis.

Chapter 1	**Chapter 2**	**Chapter 3**
Introduction	Generic methods	Pilot study - developing a protocol for leaf section selection

Chapter 4	**Chapter 5**	**Chapter 6**	**Chapter 7**
Particulate matter pollution capture by leaves of seventeen living wall species with special reference to rail-traffic at a metropolitan station	The potential of living wall species to immobilise PM associated with road-traffic	Evaluating the impact of individual leaf traits on atmospheric particulate matter accumulation using natural and synthetic leaves	The impact of rainfall in remobilising particulate matter accumulated on leaves of evergreen species grown on a green screen and a living wall

Chapter 8
General Discussion and Conclusion

Chapter 1 : Introduction

This research project explores the value of living walls in the reduction of atmospheric particulate matter (PM) pollution. This chapter will provide an insight to the problem of PM pollution and an introduction to living wall systems, including a brief overview of associated vertical greenery systems (VGS). The definitions of living walls, PM, green infrastructure (GI) and other important terms will be provided in the relevant sections of the chapter. Current knowledge regarding the ability of vegetation to mitigate PM pollution will then be reviewed. The chapter will conclude by giving the rationale of the present study and its aims and specific objectives.

1.1 Atmospheric particulate matter pollution

1.1.1. Outdoor air pollution

Seinfeld and Pandis (2006) defined air pollution as "*a situation in which substances that result from anthropogenic activities are present at concentrations sufficiently high above their normal ambient levels to produce a measurable effect on humans, animals, vegetation or materials*". In 2014, an estimated 92% of the world's population was living in areas where the outdoor air pollution levels exceeded acceptable thresholds, as specified by the World Health Organisation (WHO) air quality standards, with annual global premature deaths caused by outdoor air pollution estimated at 3.7 million (WHO, 2014). The Organisation for Economic Cooperation and Development (OECD) estimated that, over the next four decades, approximately 9 million premature deaths globally, per annum, will be caused by outdoor air pollution (Balit, 2016). Atmospheric PM (less than 10 µm in diameter) and gaseous pollutants such as ozone (O_3), nitrogen dioxide (NO_2) and sulphur dioxide (SO_2) are mainly responsible for this mortality (Balit, 2016; WHO, 2014). According to global statistics, traffic-based fuel combustion, heat and power generation, industrial emissions, municipal and agricultural waste incineration, and polluting fuels generated from residential activities are considered to be the main sources of these ambient air pollutants (WHO, 2016). In the UK at present, and based on the strong links to cardiovascular and respiratory diseases, the main concerns are particulate pollutants, oxides of nitrogen (NO_x), ozone and ammonia (Balit, 2016). According to Royal College of Paediatrics and Child Health (RCPCH) and Royal College of Physicians (RCP) estimations, 40,000 premature deaths per annum in the UK are attributable to outdoor air pollution which is largely caused by transport (fossil fuel combustion in cars and lorries) and power stations (RCP, 2016). The annual cost of air pollution to UK society has been estimated at £20 billion, while estimates for the European Union (EU) reach as high as €240 billion (RCP, 2016).

1.1.2. Atmospheric particulate matter

In recent WHO assessments, PM has been classified as the most harmful air pollutant to human health due to the higher prevalence of morbidity caused by PM in comparison to other air pollutants. (WHO, 2014). Dockery and Pope (1994) defined PM as "*an air-suspended mixture of both solid and liquid particles*". Particulates which are directly emitted to the atmosphere from the source are known as primary PM and those resulting from chemical and physical transformations of particles/gases in the atmosphere are known as secondary PM (Fig.1.1) (Fauser, 1999). Whilst atmospheric PM can originate from natural or anthropogenic sources (Weber *et al.*, 2014), primary particles are mainly generated through anthropogenic activities such as road transport, industrial activities, stationary combustion sources such as coal/biomass burning, construction work, and cigarette smoke. Naturally generated primary particles originate from land and sea as soil-based or marine aerosols (Kelly and Fussell, 2012). The best known examples of secondary PM are the nitrate and sulphate particulates which result from the oxidative transformation of nitrogen dioxide and sulphur dioxide in combustion processes, especially from road transport (Kelly and Fussell, 2012).

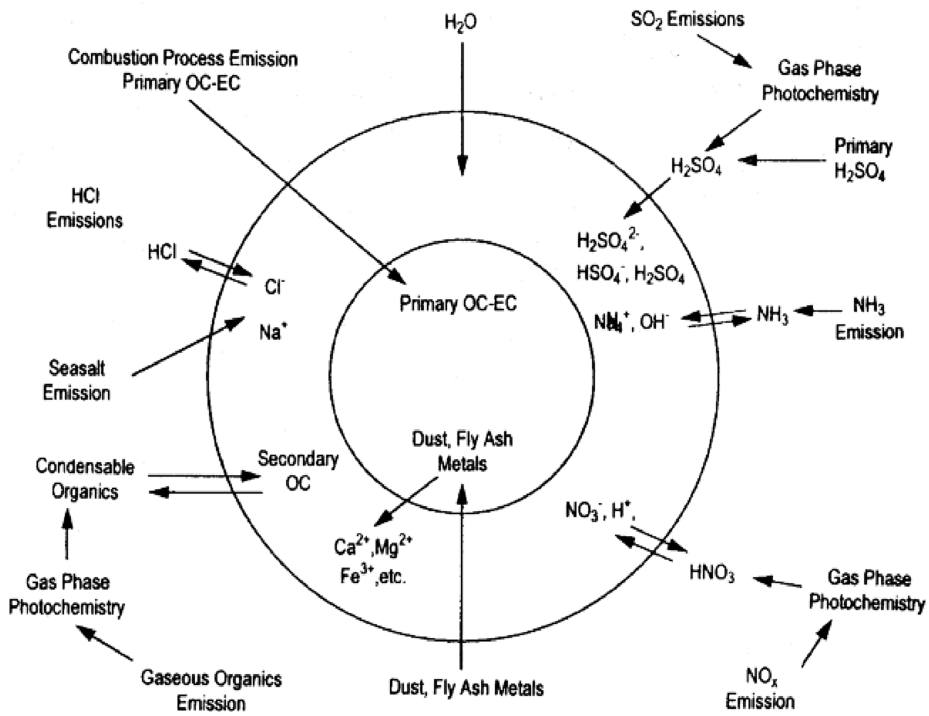


Figure 1-1 Reaction pathways in the formation of atmospheric particulate matter (from Pilinis and Pandis, 1995) OC – organic carbon, EC – elemental carbon

The behaviour of PM varies according to its aerodynamic diameter (Wang *et al.*, 2006a); the latter is defined as the diameter of a particle that has the same settling velocity as a smooth sphere with unit density of 1 kg L^{-1} (Fauser, 1999). Three different size fractions of PM are of concern based on their inhalation, deposition, and toxicity: coarse particles/PM_{10} (aerodynamic diameter ≤10μm), fine particles/$PM_{2.5}$ (aerodynamic diameter ≤ 2.5μm) and ultrafine particles/$PM_{0.1}$ (aerodynamic diameter ≤0.1μm) (Chow *et al.*, 2006; Solomon *et al.*, 2012). According to Grochowicz and Korytkowski (1996), particles in the fine and ultrafine size ranges account for 30% of the total mass of PM and 99.9% of the total number of PM. Particle number concentration (PNC) and their surface area are found to be more effective in evaluating finer particles, particularly ultrafine particles, than the conventional expression of particle mass (Sager and Castranova, 2009). The sources of these different PM sizes are summarised in Table 1.1.

Table 1.1 Sources of particulate matter in different size fractions

PM size fraction	Source	References
Coarse PM	**Natural sources** volcanos windstorms soil particles debris of forest fires sea salt biological origins: Fungal spores spores of myxomycetes bryophyte/pteridophyte pollen grains bacteria **Anthropogenic PM** farming mining, road dust burning of biomass/coal black smoke building construction some secondary particles	Kelly and Fussell, 2012 Solomon *et al.*, 2012 Morris, 1995 Li *et al.*, 2012 Solomon *et al.*, 2012 Kelly and Fussell, 2012
Fine PM	vehicle emissions - gasoline and diesel combustion of biomass and coal industrial processes: cement, paper, steel agricultural work Secondary PM: chemical reactions between nitrates, sulfates, ammonia and organic aerosols	Chow *et al.*, 2006 Kelly and Fussell, 2012 Solomon *et al.*, 2012 Stern *et al.*, 2014 Wang *et al.*, 2006 Solomon *et al.,* 2012 Kelly and Fussell, 2012
Ultrafine PM	vehicle emission combustion sources Secondary PM: photochemical reactions in the atmosphere Biological origin: Viruses	Chow *et al.*, 2006 Franck *et al.*, 2011 Li *et al.*, 2012

The chemical composition of particulates can vary depending on local environmental factors including: geology, meteorological conditions (e.g. humidity, wind speed, temperature fluctuations, precipitation), and anthropogenic activities in the area (Laongsri, 2012). PM in urban areas consists of numerous different organic substances and complex mixtures of inorganic compounds (Barros *et al.*, 2010; Spencer *et al.*, 2008). Inorganic substances generally encompass nitrate, ammonium, sulphates, and metal compounds (Barros *et al.*, 2010; Gray *et al.*, 1986; Spencer *et al.*, 2008) and account for 25% - 50% of the total mass of PM (Gray *et al.*, 1986). The organic substances in PM can include polycyclic aromatic hydrocarbons (PAH), n-alkanes, polychlorinated dibenzo-p-dioxins, carboxylic acids, dicarboxylic acids, polychlorinated biphenyls (PCBs), dibenzofurans (PCDD/Fs), and hundreds of other compounds (Barros *et al.*, 2010; Dzierzanowski *et al.*, 2011; Holzinger *et al.*, 2010).

In urban settings, fine particulates are mainly comprises organic compounds (Chai *et al.*, 2005). In the large industrial city of Beijing, the proportions of organic carbon (OC), elemental carbon (EC), water soluble ions, and trace elements were estimated at 14.7%, 1.6%, 45.0%, and 11.4% respectively (Li *et al.*, 2012). Harrison and Yin (2000) reviewed and summarised the chemical make-up of atmospheric PM in some large cities around the world (Table. 1.2) and later (Yin and Harrison, 2008) evaluated the chemical composition of PM in urban environments in the UK using a mass closure model.

Of the total mass of PM_{10}, Yin and Harrison (2008) reported 23.7% as organic compounds, 8.0% as EC, and 13.4% as Fe-rich dust; for $PM_{2.5}$ they found 26.1% as organic compounds, 11.2% as EC and 5.9% as Fe-rich dust. In urban locations in the UK, organic carbon, elemental carbon, Al, Cl, NH_4^+, SO_4^{2-}, NO_3^-, crustal and biological materials were found in bulk volumes as major elements, and some metals (Pb, Cd, Hg, Ni, Cr, Zn and Mn) and some other organic compounds were found in trace quantities (Laongsri, 2012; Yin and Harrison, 2008). EC is mostly primary PM resulting from combustion processes and largely found in the fine fraction with a diameter less than 1 µm (Laongsri, 2012).

Table 1.2 Chemical composition of PM (%) in large cities around the world as reviewed by Harrison and Yin (2000) and modified by Laongsri (2012).

Sampling site	PM	Total Mass (µg m^{-3})	EC	OC	Organics	TC	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Crustal	Minerals	Other	Reference
Birmingham, UK	$PM_{2.5}$	15.8	11.2	-	26.1	-	24.0	21.2	4.0	-	8.4	-	5.2	Yin and Harrison,
	PM_{10}	23.9	8.0	-	23.7	-	16.0	18.5	9.3	-	20.8	-	3.7	2008
Birmingham, UK	PM_{10}	25.7	18.0	20.0	-	38.0	17.0	6.0	2.0	6.0	-	-	31.0	Harrison et al., 1997
	$PM_{2.5}$	22.5	-	-	-	50.0	26.1	6.6	1.8	9.9	-	5.6	0.0	
Leeds, UK	$PM_{2.5-15}$	13.3	-	-	-	13.3	7.4	5.8	8.2	2.5	-	62.8	0.0	Clarke et al., 1984
	PM_{15}	35.5	-	-	-	33.8	19.2	7.9	4.2	6.8	-	28.1	0.0	
Southern California	$PM_{2.5}$	37.0	5.0	-	26.5		20.9	9.8	0.4	9.0	2.5	-	23.9	Chow et al., 1994
(urban)	PM_{10}	37.0	3.5		20.0		13.2	12.7	1.3	5.7	18.2	-	19.3	
Edison California	$PM_{2.5}$	49.6	6.0	31.4	44.0	37.4	6.0	3.0	-	2.0	35.0	-	0.0	Chow et al., 1996
	PM_{10}	52.5	5.7	19.7	27.6	25.4	6.3	3.0	-	2.0	46.1	-	4.3	
	$PM_{2.5}$	-	3.9	14.9	20.9	18.8	34.1	1.1	-	13.0	-	4.3	22.8	
Eastern U.S.	$PM_{2.5-10}$	-	-	-	-	-	4.9	-	-	1.8	-	51.8	41.5	U.S. EPA, 1996
	PM_{10}	-	3.3	6.1	8.5	9.4	27.8	1.2	-	10.7	-	19.6	18.9	
	$PM_{2.5}$	-	14.7	27.8	38.9	42.5	10.8	15.7	-	7.5	-	14.6	0.0	
Western U.S.	$PM_{2.5-10}$	-	-	-	-	-	3.1	-	-	0.8	-	69.3	26.8	U.S. EPA, 1996
	PM_{10}	-	5.1	21.4	30.0	26.5	4.6	24.0	-	6.7	-	36.3	0.0	
Lahore Parkistan	TSP	607	2.9	13.1	-	16.0	3.0	2.1	-	1.2	-	16.4	61.3	Smith et al. 1996
Hong Kong	RSP	66.2	-	-	-	57.1	14.4	2.8	2.3	3.3	-	6.1	14.0	Qin et al., 1997
Beijing China	$PM_{2.5}$	66	4.7	-	24.2	-	24.9	9.7	0.3	14.3	1.7	-	20.1	Huang et al., 2005

Crustal elements of PM are mainly contained in soil-based particles and in road dust associated with traffic emissions (e.g. Ba, Cu, Ni, Zn, Pb, Si, Ca, Fe, Al, K, Zn, and Ti) (Lin *et al.*, 2005; Swietlicki *et al.*, 1996). Numerous ions such as SO_4^{-2}, NO_3^-, Ca^{2+}, NH_4^+, Na^+, K^+, Mg^{2+}, Cl^-, and NO_2^- can be found in the water-soluble fraction of PM, and sulphate can also be found as parts of OC particulates and toxic metal particles (Li *et al.*, 2012). According to Karanasiou *et al.* (2007), Fe, Ca, Al, Mg and Ba metals are mainly found in the particles with 4.7 - 5.8 µm diameter and only minor quantities found in the particles with 0.65 - 1.1 µm diameter, whereas, Cr, Ni, Mn, Sb, As and K showed a bimodal distribution in particles with

0.65 - 1.1 µm diameter and in 4.7 - 5.8 µm diameter. According to Li *et al.* (2012), an estimated 95% of the total elemental composition was accounted for by Fe, Al, Mg, Ca, Na, and K, and only 0.5% was accounted for by minor PM components such as As, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sb, Se, Tl, V, and Zn (Li *et al.*, 2012).

In addition to their detrimental health effects (see Section 1.1.2.1), PM pollutants are also known to reduce atmospheric visibility, alter the radiative properties of the atmosphere (Han *et al.*, 2012), and change the urban thermal environment (Yan *et al.*, 2016b). PM in the atmosphere can alter radiation and thermal balance by absorbing solar and thermal radiation reflected from the earth, thereby influencing climate (Bellouin *et al.*, 2005; Gustafson *et al.*, 2011). Due to the presence of climate forcers such as black carbon (BC), the Intergovernmental Panel on Climate Change (IPCC) identified PM as a direct contributor to of global warming; while some other constituents in PM such as organic carbon (OC), ammonium, sulphate, and nitrate were considered to have a cooling effect (IPCC, 2013). The annual cost to UK society due to PM pollution has been estimated at £16 billion (RCP, 2016).

1.1.2.1. Health effects of PM

PM pollution has been linked to various health effects, emergency room visits, and hospital admissions (Atkinson *et al.*, 2001; Halonen *et al.*, 2008; Pope and Kanner, 1993; Schwartz, 1996; Shaughnessy *et al.*, 2015). Coarse particles are also known as thoracic particles as they can reach the human lower respiratory system (Brunkeef and Holgate, 2002) and mainly deposit in the primary bronchioles (Kelly and Fussell, 2012). Fine particles are known as respirable particles as they can reach the narrower spaces in lungs and enter the alveoli and terminal bronchioles (Brunkeef and Holgate, 2002; Kelly and Fussell, 2012). Ultrafine particles are more important as they are capable of penetrating the air-blood barrier in the deeper respiratory system, enter the systemic circulation (Franck *et al.*, 2011), organs, and even cross cell membranes and influence intracellular functions (Kelly and Fussell, 2012; Riddle *et al.*, 2009; Solomon *et al.,* 2012). High reactive surface areas, high alveoli deposition, and reduced ability in clearance make ultrafine particles more harmful compared to other particle size fractions (Franck *et al.*, 2011).

The European Environment Agency (EEA) (2017) estimated that long-term exposure to $PM_{2.5}$ was responsible for 428,000 premature deaths in Europe in 2014 (Guerreiro *et al.*, 2017). They also estimated that 82–85% and 50–62% of the urban population in the EU were exposed to levels of $PM_{2.5}$ and PM_{10} (respectively) far in excess of the recommended standards, in the 2013–2015 period. The association between respirable PM and health effects has revealed that even very low levels of exposure to PM can cause health issues (Shaughnessy *et al.*, 2015). The increased rate of mortality in infants and older people due to the sudden upsurge in atmospheric PM during the London fog episode in 1952, and findings of contemporary studies in other countries with high atmospheric PM levels have shown a strong link between PM pollution and increased mortality (Woodruff *et al.*, 2006). Out of 40,000 annual deaths estimated to have been caused by outdoor air pollution in the UK, 29,000 were caused by PM (RCP, 2016). The death risk associated with a 10 $\mu g.m^{-3}$ increase of PM_{10} is 0.5% (Laden *et al.* 2006) and for an increase of 10 μg.m^{-3} of $PM_{2.5}$, it is 6%, 8% and 4% for cardiopulmonary diseases, lung cancer, and other causes respectively (Pope III *et al.*, 2011). Although their exposure dosages are negligible in terms of mass, ultrafine particles usually dominate respirable PM in terms of reactive surface area (Franck *et al.*, 2011; Oberdorster *et al.*, 1994; Sager and Castranova, 2009). Feng and Yang (2012) found a positive association between cardiovascular diseases and chronic exposure to PM across 2,231 contiguous U.S counties in the 2007 – 2009 period, and Karottki *et al.* (2014) found a statistically significant inverse relationship between outdoor PNC (PM_{10}, $PM_{2.5}$ and $PM_{0.1}$) and microvascular function (MVF) which is a widely used indicator for cardiovascular hazard. Cardiovascular effects are known to be linked more with $PM_{2.5}$ released from combustion sources (e.g. traffic) and crustal elements (Stanek *et al.*, 2011). Schwartz (1996) reported 5%, 8% and 6% increase in hospital admissions per 50 mg m^{-3} increase in PM_{10}, due to cardiovascular disease, chronic obstructive pulmonary disease (COPD), and pneumonia (respectively) using a multi-city study in the U.S.

Some chemicals in PM, PAHs in particular, have potential geno-toxic and carcinogenic effects (Fauser, 1999; Ji *et al.*, 2010; Maitre *et al.*, 2011). It was estimated there was an approximate 10-15% increase in risk of lung cancer by living in a city with high PM levels compared to a cleaner city (Beeson *et al.*, 1998; Pope *et al.*, 2002). Short- and long-term exposure to PM is also responsible for other chronic respiratory effects such as decreasing lung function, exacerbation of COPD, increases in the symptoms of bronchitis, exacerbation of asthma, oxidative stress in macrophages by iron-related PM, and allergies (Anderson *et al.*, 2013; Braun-Fahrlander *et al.*, 1997; Gehring *et al.*, 2010; Gilmour *et al.*, 1996; Parker *et al.*, 2009; Pierse *et al.*, 2006; Pope and Kanner, 1993; Raizenne *et al.*,1996; Walters *et al.*, 1994). Numerous studies have reported a strong correlation of daily PM levels with hospital admissions, lung function test results, and respiratory symptoms (Chestnut *et al.*, 1991; Pope *et al.*, 1991; Shaughnessy *et al.*, 2015). Raizenne *et al.* (1996) reported a significant relationship between PM exposure levels and pulmonary functions of children in the UK and Canada. Post-neonatal mortality associated with PM pollution (Laden *et al.*, 2006) is mainly caused by respiratory problems (Ha *et al.*, 2003) and Sudden Infant Death Syndrome (SIDS) (Woodruff *et al.*, 1997).

Brook *et al.* (2010) summarised the health effects identified as following short-term exposure of PM such as progression of atherosclerosis, systemic inflammations, decrease of MVF, endothelial dysfunctions, pulmonary inflammations, changes in cardiac autonomic function, oxidative stress, and complications in blood coagulation. In addition to cardiopulmonary diseases, particulate pollution causes adverse effects on reproductive (e.g. preterm delivery, foetal measurements) and neurological health (Kelly and Fussell, 2012). Ranft *et al.* (2009) found an association between chronic exposure to traffic-generated PM and mild cognitive impairment (MCI). According to Seaton *et al.* (1995) ultrafine particles can be retained in alveolar macrophages and influence phagocytosis. Once they enter the blood stream they can be transported to the liver, heart and kidney (Kreyling *et al.,* 2010) where they may cause systemic inflammatory changes (Brunekreef and Holgate, 2002). They can also enter the brain via the olfactory nerve which may cause various central nervous system disorders depending on their chemical composition and toxicity (Solomon *et al.*, 2012). Iron-rich particles in the brain are particularly important in this respect as they are known to cause oxidative brain damage which potentially leads to neurodegenerative conditions such as Alzheimer's and Parkinson's diseases (Allsop *et al.*, 2008; Maher *et al.*, 2013).

1.1.2.2 Traffic-generated PM

Traffic-generated PM constitutes a significant portion of atmospheric PM (Pant and Harrison, 2013; Ranft *et al.*, 2009) and is considered to be the most toxic class (WHO, 2005). Approximately 25% of global $PM_{2.5}$ and PM_{10} in the atmosphere (Karagulian *et al.*, 2015) and 50% of PM_{10} in Europe (Künzli *et al.*, 2000) is considered to be generated by road-traffic. The largest proportion of traffic-generated PM is emmitted from diesel exhaust and this is considerably higher compared to petrol engines (Kelly and Fussell, 2012). In some large industrialised cities in the world, DEPs (diesel exhaust particles) account for the majority of atmospheric PM (up to 90%) due to their greater number of diesel-fuelled cars (Shah *et al.*, 2004). In addition to vehicle exhaust, traffic-derived PM contains various non-exhaust emissions released from engine wear-and-tear, brake pads, tyre wear, abrasives, clutch wear, and dust from the road surface (Mulawa *et al.*, 1997; Thorpe and Harrison, 2008; Wåhlin *et al.*, 2006). Traffic-derived ultrafine PM is given special attention due to its higher toxicity and greater abundance compared to other particle sizes (Lin *et al.*, 2005). The chemical composition of these particles is elemental carbon, high molecular weight organic components including PAHs, heavy metals, oxides of nitrogen, sulphates, and ammonia; most of these are carcinogens (Fauser, 1999; Sharma *et al.*, 2005). The International Agency for Research on Cancer (IARC) classified diesel exhaust as a Group 1 (carcinogenic to humans) carcinogen (Silverman *et al.*, 2012).

In the UK, especially in London, traffic-generated pollution has become the major source of PM in air (Fig. 1.2) (DEFRA, 2017). Approximately 39% of the cars in the UK (DfT, 2017) and 50% in EU are diesel powered (RCP, 2016). Traffic volume is predicted to increase further in the 2015-2040 period, in the UK, by 19–55% (DfT, 2015). However, the petrol:diesel mix may change following the 'Volkswagen scandal' (Environmental Protection UK, 2018) and petrol:electric hybrids and electric vehicles may be introduced more rapidly. Previous concerns about air pollution due to air acidification and black smoke from coal burning have now changed to concerns about PM and NO_x emissions from transport and secondary pollutants generated in the atmosphere (RCP, 2016). In addition to road transport, railway traffic is one of the important PM sources in the UK due to diesel and electric train emissions; other than from train exhaust, fine and ultrafine PM are mainly released via wheel friction, engine wear-and-tear, friction with overhead cables, and brakes (Thornes *et al.*, 2016).

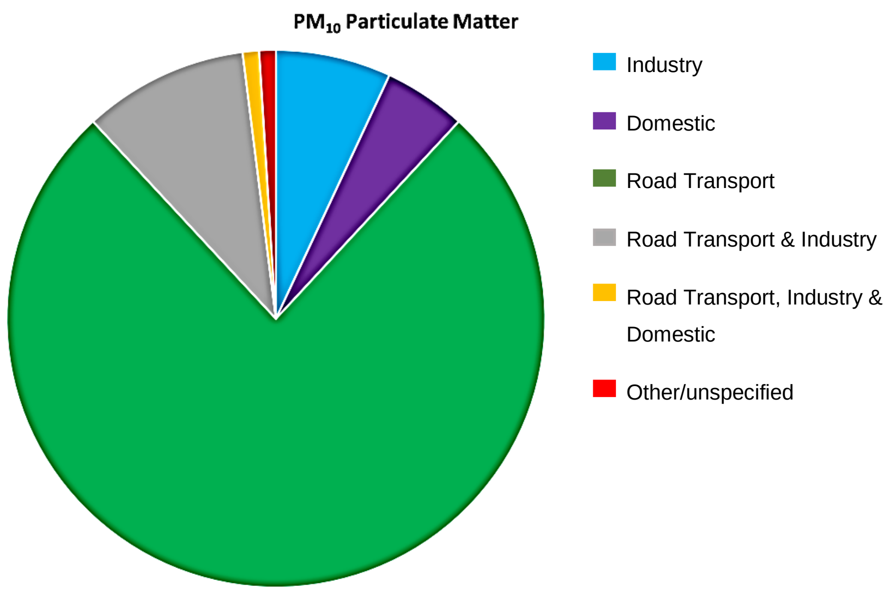


Figure 1-2 The proportional source contribution of PM_{10} in the UK based on analysis conducted in Air Quality Measured Areas (AQMA) (DEFRA, 2017).

1.1.3 PM pollution mitigation

Air pollution mitigatory measures are aimed at minimising the concentration of pollutants in the atmosphere either by limiting the emissions directly at point source or minimising the total pollutant input to the local environment (RCP, 2016). The majority of these mitigatory approaches commonly act on most outdoor air pollutants whilst some specifically address PM, such as PM traps in diesel-fuelled vehicles (RCP, 2016). Considering the serious threat of outdoor air pollution on human health, in 2005, WHO declared air quality guidelines for important air pollutants, including PM; limits for $PM_{2.5}$ were defined as a daily mean of 25 $\mu g.m^{-3}$ and an annual mean of 10 $\mu g.m^{-3}$ and limits for PM_{10} were defined as a daily mean of 50 $\mu g.m^{-3}$ and an annual mean of 20 $\mu g.m^{-3}$ (WHO, 2016). WHO estimated that an annual reduction of particulates (below 10 µm in diameter) from 70 $\mu g.m^{-3}$ (world average level 71 $\mu g.m^{-3}$) to 20 $\mu g.m^{-3}$ could reduce air pollution-related deaths by 15% (WHO, 2014). In addition to WHO guidelines, under '*National objectives for the protection of human health',* the European Union, adopted air quality guidelines in Directive 2008/50/EC: for PM_{10}, a daily mean of 50 $\mu g.m^{-3}$ (not to be exceeded more than 35 times a year) and an annual mean of 40 $\mu g.m^{-3}$; for $PM_{2.5}$, an annual mean of 25 $\mu g.m^{-3}$ (EEA, 2016). However, according to RCP (2016) irrespective of these specified guidelines, any smaller level of PM exposure carries a risk to human health and there is no actual safe level. Several different mitigatory approaches such as emission reduction, enhancing atmospheric dispersion and building high emission sources away from current pollution hotspots or highly populated areas have been taken both nationally and internationally to combat PM pollution (Pugh *et al.*, 2012).

The air pollution legislation in EU followed a 'twin-track approach' to meet the air quality standards by reduction of $PM_{2.5}$ exposure and including controls on emission levels. In addition to local pollution control approaches, there are several international interventions such as the Convention on Long-range Transboundary Air Pollution (CLRTAP), the 1979 United Nations Economic Commission for Europe (UNECE), and the Gothenburg Protocol (Guerreiro *et al.*, 2017). The UK Health Alliance on Climate Change has proposed six main strategies that, together, address all important air pollutants: “enhance cross-departmental collaboration (mainly the health sector and the Government) in reducing air pollution and climate change; discontinue the use of coal power by 2025; extend the clean air zones further to other cities and expand the already existing ones; improve the air pollution monitoring in the areas with large vulnerable populations; and retain or enhance EU air quality regulations” (Balit, 2016).

Despite all national and international initiatives taken, PM levels in the UK (Pugh *et al.*, 2012) and many large cities around the world (WHO, 2016) (Fig. 1.3) exceed recommended air quality standards. Current strategies of emission reduction and building clean air zones are long-term targets and are unlikely to bring a short-term improvement in reduction of city PM levels generated from transport. This has been exacerbated by the difference between lab testing of car emissions and real world emission levels indicating the scandal of ‘cheat’ devices used by Volkswagen (Environmental Protection UK, 2017).

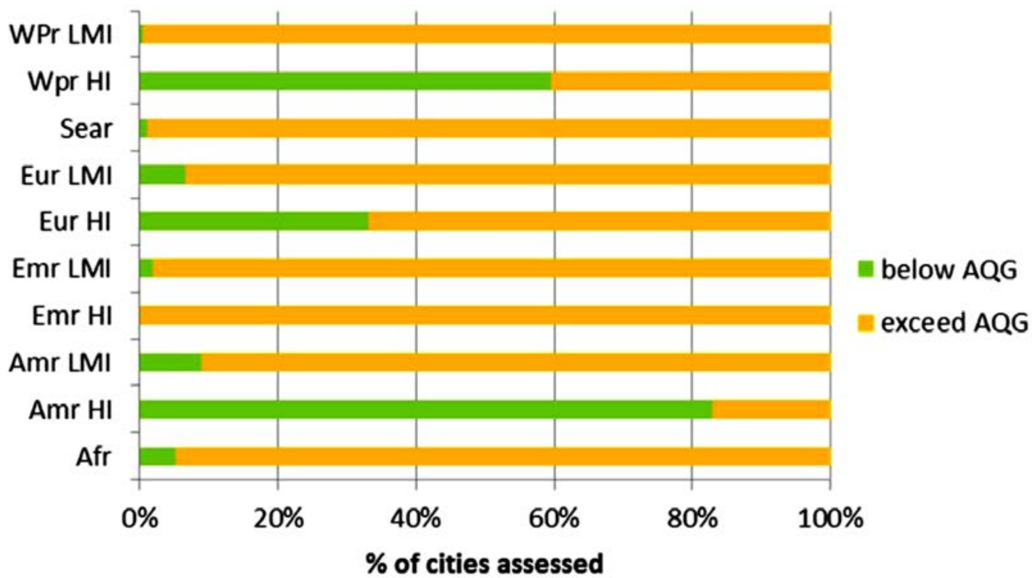


Figure 1-3 Annual mean PM (PM_{10} and $PM_{2.5}$) levels in cities around the world with reference to WHO air quality guidelines (AQG)* (WHO, 2016)

* Afr: Africa; Amr: America; Emr: Eastern Mediterranean; Eur: Europe; Sear: South-East Asia; Wpr: Western Pacific; LMI: Low- and middle-income; HI: high-income

Achieving air quality targets and improving human health need effective short-term strategies such as increasing surface deposition (Pugh *et al.*, 2012). Since vegetation is known to act as a sink for PM (Beckett *et al.*, 2000b; Slinn, 1982; Freer-Smith *et al.*, 2005) the potential of urban green infrastructure in reducing PM pollution has recently being highlighted (Abhijith *et al.*, 2017; Dover, 2015; Jayasooriya *et al.*, 2017).

1.2. An introduction to living wall systems as a component of Green Infrastructure

1.2.1. Green walls – an overview

Green walls are vertical greenery systems (VGS) (Wong *et al.*, 2010a), are an important component of Green Infrastructure (GI) and are potentially valuable in highly urbanised settlements. Green Infrastructure can be defined, as *"a strategically planned and delivered network comprising the broadest range of high quality green spaces and other environmental features. It should be designed and managed as a multifunctional resource capable of delivering those ecological service and quality of life benefits required by the communities it serves and needed to underpin sustainability. Green Infrastructure includes established green spaces and new sites and should thread through and surround the built environment and connect the urban area to its wider rural hinterland."*(Natural England, 2009). Dover (2015) defined the concept of green walls as growing plants on the unused space on walls, and differentiated them from green roofs and ground vegetation, which take up space. Vertical greenery was historically known since the Hanging Gardens of Babylon were constructed in 500 BC (Manso and Castro-Gomes, 2015; Wong *et al.*, 2010b). Whilst many environmental values of GI for the integrity of urban ecosystems are well recognised (Lafortezza *et al.*, 2013; Naumann *et al.*, 2011; Pakzad and Osmond, 2016), finding land space for trees has been challenging due to increasing population pressure, rapid expansion of cities, and the economics of tree maintenance. Urban settings are hostile to planting trees due to various other challenges such as prevailing soil conditions, potential building shading effects, and sub-surface infrastructure (Johnston and Newton, 2004). Since green walls can overcome these limitations, they have become popular, particularly in the context of city development (Pérez-Urrestarazu *et al.*, 2015).

1.2.2. Types of green wall systems

Green walls are diverse in form, and definitions have been based on their designs and the type of plant incorporated. Hence, previous researchers have developed various classification systems, but with several overlaps based on their characteristics:

- Köhler (2008) and Dunnett and Kingsbury (2008) classified green walls into two groups as 'green facades' and 'living walls' based on the type of vegetation used in the systems.
- Ottelé *et al.* (2011) categorised them as direct and indirect walls based on having an air filled gap between the system and wall.
- Manso and Castro-Gomes (2015) proposed a classification based on their construction characteristics (Fig. 1.4).
- Dover (2015) used a simplified version of Ottele *et al.* (2011)'s of direct and indirect walls, based on having direct or indirect connection between vegetation and wall, and further classified using vegetation charactersitics.

- Sadeghian (2016) classified them into 3 groups as wall-climbing, hanging-down, and modular green walls combining the characters of growing media, construction, and vegetation.

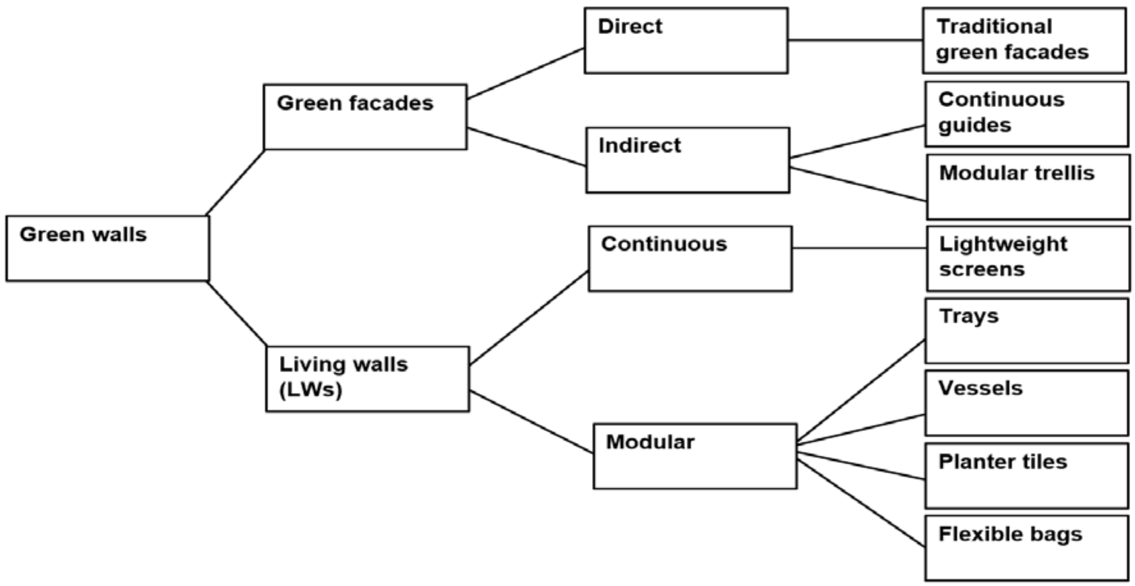


Figure 1-4 Classification of green walls based on the construction characteristics (Manso and Castro-Gomes, 2015)

Based on the context or purpose of development, there are several overlaps in these classifications. As the focus of this thesis is mainly based more on vegetation characteristics and planting design than their construction features, the classification used in Dover (2015) has been adopted (Fig. 1.5). Plants which were directly rooted on the walls, or climbers which were rooted on the ground but directly attached to the walls or vertical surfaces, were considered as direct green walls, whereas plants grown on supporting structures which were attached to the wall or vertical surfaces without having a direct contact between vegetation and the wall surface were classified as indirect walls. Based on the type of vegetation used and the design of the system, indirect green walls can be further categorised into green facades and living walls. In addition to direct and indirect walls, hedges are also a type of green wall with its own support mechanism (Dover, 2015).

1.2.2.1. Green facades and green screens

Green facades are VGSs that use climbing or hanging species rooted in the ground, planters or on the roof, which are growing upward or downward by twisting around a supportive structure (e.g. wire mesh) that keep the vegetation detached from the wall (Fig. 1.6) (Dover, 2015; Pérez *et al.*, 2011). However, in some research, surface climbers which directly attach to walls have also been considered as green facades based on the classification used (Manso and Castro-Gomes, 2015). Free-standing green facades which are not attached to a building facade are known as green screens (Chiquet, 2014) which is a special case of facade greening where climbing species are pre-grown on wire mesh and are installed utilising very little ground space and used with or without artificial irrigation (Dover, 2015; Pérez *et al.*, 2011). Green screens are suitable in both outdoor (typically *Hedera helix*) and indoor applications (typically *Philodendron* sp.). Green facades generally use one or few species of climbers; as a consequence of their lower diversity, lower vegetation density, and simple supporting structures they require less maintenance and protection compared to living walls (Ottelé *et al.*, 2011) and hence they are a relatively cheap indirect greenery system (Perini and Rosasco, 2013). However, use of stainless steel as a supportive structure, in many types of green facade, increases their environmental burden compared to other systems and replacing stainless steel with low impact recyclable material such as hard wood and high density polyethylene (HDPE) can potentially reduce their environmental footprint (Ottelé *et al.*, 2011).

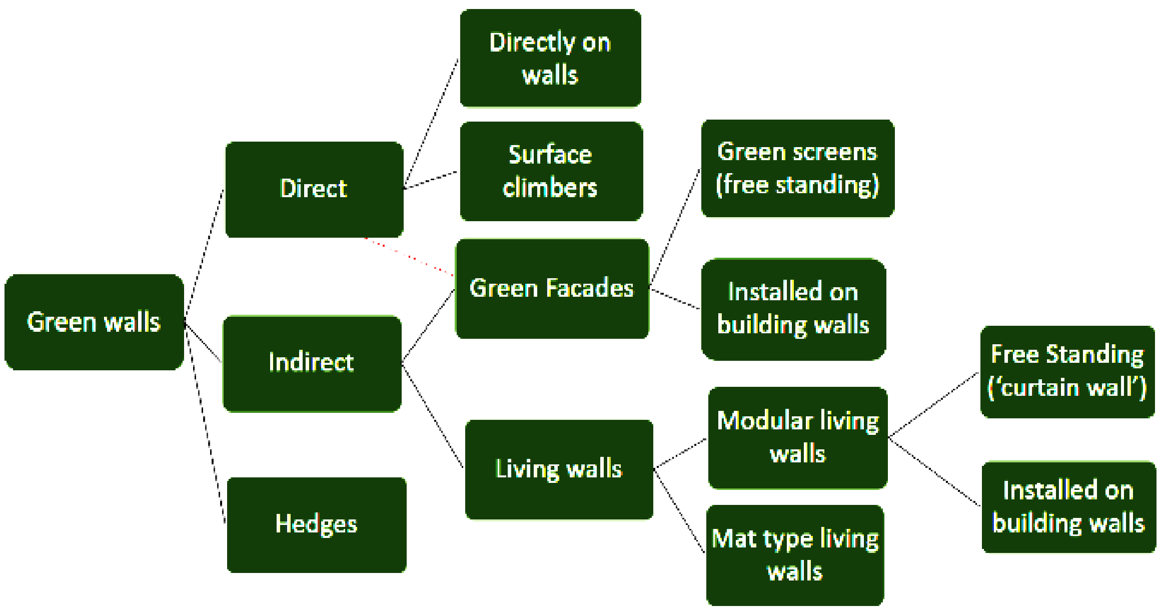


Figure 1-5 Classification of green walls based on the vegetative characteristics and attachement to the wall (after Dover, 2015).

Figure 1-6 A green screen manufactured by Mobilane ® located on Staffordshire University's campus in Stoke-on-Trent, UK.

1.2.3. Living walls

Living walls are vertically grown, artificially irrigated, mostly hydroponic systems which facilitate the growth of a wide variety of plant species, with a potential for greater artistic expression than simply using climbing species (Dover, 2015). Living walls are relatively new innovations and are the most advanced type of green wall systems designed to integrate VGS into taller buildings (Manso and Castro-Gomes, 2015). Living walls allow a rapid, and uniform, coverage of large vertical surfaces, and can be adapted to all types of buildings, including skyscrapers. There are two main types of living wall: the mat type ('Patrick Blanc systems') (Blanc, 2008) and the modular type (Fig.1.7) based on their different design characteristics (Dover, 2015). Mat type systems are also known as continuous living walls, where plants are grown on a hydroponically-maintained felt-based blanket attached to a structural frame, whereas, in modular living walls, plants are grown in modules carrying a growing medium such as soil, rockwool, coco-coir, Grodan©, or peat (Fig. 1.8) (Dover, 2015; Feng and Hewage, 2014; Pérez *et al.*, 2011). Modular systems have the advantage of being planted *in-situ* after the wall installation or by attaching pre-vegetated panels to the structural frames (Feng and Hewage, 2014; Pérez-Urrestarazu *et al.*, 2015). There are numerous living wall companies established globally, with a range of design approaches to the implemenation of VGS (depending on the manufacturer). Modular panels are made out of various materials such as synthetic fabric, plastic, concrete, or metal (Pérez *et al.*, 2011). Modular living walls can also be free-standing, consuming little ground space, without being attached to a building wall (Dover, 2015).

However, compared to other VGSs, living walls are expensive and have high maintenance requirements (Feng and Hewage., 2014). On the other hand, according to a life cycle analysis conducted by Ottele *et al.* (2011) and Feng and Hewage. (2014), modular living wall systems are durable and environmentally sustainable compared to other indirect VGSs due to their use of low impact material usage (e.g. HDPE and unplasticised polyvinyl chloride - UPVC). In contrast, felt-based, mat-type systems have a serious environmental burden due to their high environmental footprint including high energy and water consumption (Ottele *et al.*, 2011; Feng and Hewage., 2014). The use of non-recyclable material (typically PVC) in these systems further increases their environmental burden of wastage which can be potentially reduced by alternative material usage (Feng and Hewage., 2014) and by integrating the systems into the building envelope (Ottele *et al.*, 2011).

1.2.4. Benefits of living walls and green facades

Although VGSs were initially recognised for their aesthetic values (Perini *et al.*, 2012) they have a great potential for delivering environmental benefits/ecosystem services (Dover, 2015; Perini *et al.*, 2011; Manso and Castro-Gomes, 2016); both living walls and green facades deliver similar benefits, the extent depending on the diversity and density of the vegetation used (Pérez-Urrestarazu *et al.*, 2015; Ottelé *et al.*, 2011). Living walls are aesthetically more appealing compared to all other systems due to their more flexible planting regime and higher species diversity (Pérez-Urrestarazu *et al.*, 2015).

Figure 1-7 Images of a) Patrick Blanc Living wall system (mat wall) attached to the Athenaeum Hotel in Piccadilly, London; b) the free-standing 'curtain' modular living wall at the Westfield Centre, Shepherd's Bush, London © John Dover; c) the modular living wall at the Waitrose supermarket in Bracknell, Berkshire © John Dover

Thermal benefits and associated energy saving effects of VGSs have been comprehensively studied, in diverse climatic conditions. When installed on buildings, VGSs have been found to provide both cooling and insulating benefits (Cheng *et al.*, 2010; Feng and Hewage, 2014; Manso and Castro-Gomes, 2016; Perini *et al.*, 2011; Wong *et al.*, 2010a) together with associated cost savings due to lower energy consumption from air conditioning and heating systems (Coma *et al.*, 2017); reduced energy consumption also reduces CO_2 production (Sheweka and Magdy, 2011). According to Alexandri and Jones (2008), energy saving by green walls can range from 35% to 90% depending on the climatic conditions, the amount of vegetation used and their location in/on the building. In addition, by absorbing most of the solar radiation and preventing it reflecting back to the lower atmosphere, these green wall systems can also help to reduce the 'urban heat island effect' (Chen *et al.*, 2014; Gago *et al.*, 2013; Onishi *et al.*, 2010; Taha, 1997).

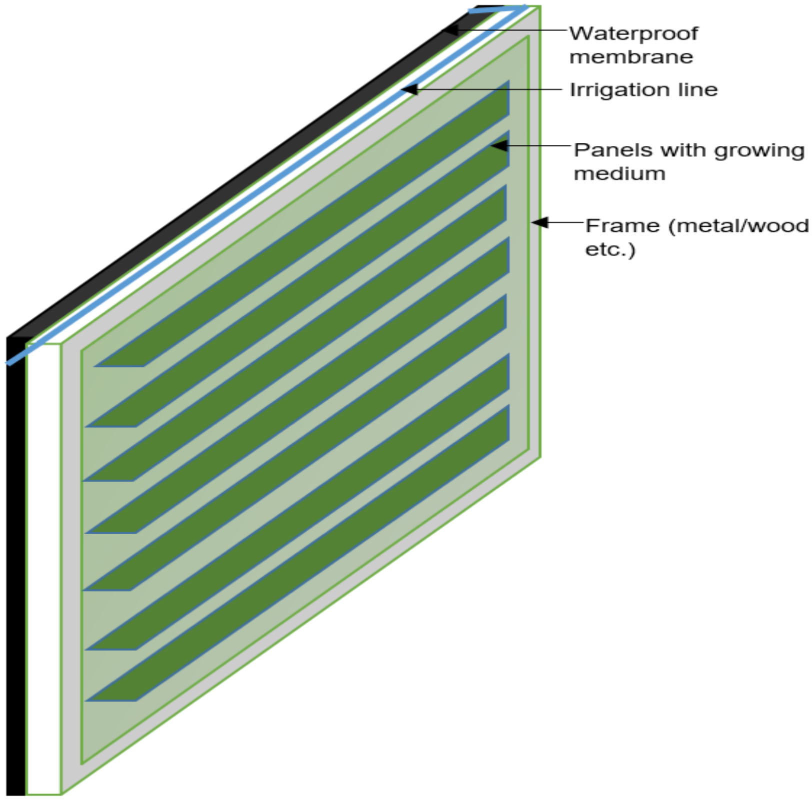


Figure 1-8 Diagrammatic representation of the basic structure of a modular living wall system

Most of the land in urban areas is devoted to human needs and biodiversity values of cities have declined in most cases (Pauchard *et al.*, 2006); green walls have been recognised in urban reconciliation ecology for their great potential for improving urban biodiversity by providing habitats for urban wildlife (Collins *et al.*, 2017; Francis and Lorimer, 2011). According to Chiquet (2014), even the ivy walls with relatively low coverage are found to be important in biodiversity and green walls can be suitable for different species depending on the maturity of the vegetation and the volume. The biodiversity value of living walls is particularly important due to their high floral diversity providing habitats for more species; moreover, they help pollination by attracting insects and birds (Chiquet, 2014; Collins *et al.*, 2017; Francis and Lorimer, 2011; Köhler, 2008; Ottelé *et al.*, 2011; Perini *et al.*, 2011; Roehr and Laurenz, 2008). Chiquet *et al.* (2013) and Köhler (1993) reported ecosystem services of VGSs for birds by providing habitats, food, and nesting sites in the UK (in both living walls and green facades) and Berlin (in green facades) (respectively). Mårtensson *et al.* (2016) conducted a field experiment and showed the ability of living walls to grow edible evergreen plants in the Scandinavian climate, which can be directly beneficial to both human and urban wildlife.

Reduction of urban air pollution is one of the applications of green walls of particular interest at present; they are known to improve both outdoor and indoor air quality by reducing harmful air pollutants such as NOx, volatile organic compounds (VOCs) (e.g. formaldehyde, benzene etc.), SO_2, CO_2 and PM (Cheng *et al.*, 2010; Coward *et al.*, 1996; Lee and Sim, 1999; Lohr and Pearson-Mims,1996; Wolverton,1993).

By reduction of greenhouse gases in the atmosphere, living walls and green facades support adaptations to climate change (Sheweka and Magdy, 2011; Matthews *et al.*, 2015). A study conducted by Thottathil *et al.* (2011) demonstrated that green facades can efficiently remove sulphates, nitrates, and particulates from the atmosphere. Although tall trees can increase air pollution in street canyons by obstructing the air flow, using a green wall scenario in the Urban Forest Effects Model (UFORE), Pugh *et al.* (2012) demonstrated that VGSs had the potential to reduce NO_2 and PM_{10} by 40% and 60% respectively. Several studies have revealed a great potential of green facades (mainly using *Hedera helix*) to remove PM pollutants from air (details are reviewed in section 1.3) (Dover and Phillips, 2015; Perini *et al.*, 2017; Ottelé *et al.*, 2010; Sternberg *et al.*, 2011). Prior to the inception of this research, little or no information was available on the value of living walls; whilst Perini *et al.* (2017) described the VGS used in their study as a facade, the design had elements of living walls by having two shrubs interspersed with two climbers (details are reviewed in section 1.3).

Whilst surface-climbing plants such as ivy have a largely undeserved reputation for damaging buildings (Mishra *et al.*, 1995; Mouga and Almeida, 1997; Viles *et al.*, 2011) indirect VGSs are recognised for their bioprotective role by shielding the building facade from acid rain, weather changes and damp (Ottele, 2011; Pérez-Urrestarazu *et al.*, 2015; Sternberg *et al.*, 2011). Living walls and dense green facades also act as noise barriers (Berardi *et al.*, 2014; Köhler, 2008; Salmond *et al.*, 2016) and were estimated to reduce noise by 5 dB(A) when using *Parthenocissus tricuspidata* (Boston ivy) and by 2–3 dB(A) when using *Rubus sp.* (Köhler, 2008). Reduction of rain water runoff and facilitating sustainable urban drainage (Czemiel Berndtsson, 2010; Roy *et al.*, 2012) and discouragement of graffiti in cities (Mir, 2011) are other important environmental services delivered by these systems. Beside these environmental benefits, in addition to direct impacts on human health by decreasing air pollution related diseases, green walls also improve mental health of urban dwellers by reducing environmental stress (Dunnett and Kingsbury 2004; Köhler 1993; Ulrich and Simons 1986). In general, the ecosystem services of living walls are more pronounced compared to facade greening or any other VGS due to their higher species diversity (Feng and Hewage, 2014) .

1.3. The ability of vegetation to mitigate PM pollution

1.3.1. PM dry deposition on vegetation: the mechanism

PM can be passively removed from the atmosphere by wet deposition via precipitation and by dry deposition on land cover (McDonald *et al.*, 2007); moist, rough, or electrically charged surfaces are favoured in this respect (Pye, 1987). Davidson and Wu (1990) defined PM dry deposition as "*transport of particulate contaminants from the atmosphere onto surfaces in the absence of precipitation*"; and identified the key influential factors for this process: atmospheric characteristics, nature of the deposition surface, and properties of the PM. Sehmel (1980) summarised possible attributes within each of these factors to explain the variability in deposition velocities of the particles (Table. 1.3).

Roupsard *et al.* (2013) compared the effectiveness of PM dry deposition on different surface materials and demonstrated that vegetation is more effective than smooth building surfaces in this respect; the level of PM deposition on synthetic grass (i.e. simulating some physical characteristics of vegetation e.g. a rough surface with turbulent flow) was 10 to 30 times higher than on cement or glass for fine particulate matter. The ability of urban parks and gardens to filter out dust particles has been discussed since the 1950s (Hennebo, 1955). Vegetation can remove atmospheric particulates by absorbing them through the stomata (some ultrafine particles behave as gaseous molecules and some have been found on sub-stomatal cavity surfaces) (Thompson *et al.*, 1984; Fowler, 2002) and by four main processes of dry deposition (Fig.1.9): sedimentation under gravity, diffusion by Brownian motion, turbulent transfer resulting in impaction, and interception (Chamberlain and Little, 1981; Wang *et al.*, 2006). In sedimentation, particle deposition is driven by gravity and the collection efficiency depends on the deposition surface and properties of the particles (Legg and Powel, 1979). When the inertia of particles is too high to follow the air flow deviations around an object they collide with the surface and deposit via impaction (Petroff *et al.*, 2008a). When particles with small inertia, which thus follow the airflow, pass over plant surfaces with less than half a diameter distance between the centre of the particle and the deposition surface they collide and deposit via interception (Fuchs, 1964). Diffusion via Brownian motion transfers the PM along a concentration gradient which is only significant close to the deposition surface (Davidson and Wu, 1990; Roupsard *et al.*, 2013).

Table 1.3 Example properties of the atmosphere-surface-contaminant system influencing dry deposition

Atmospheric properties	Surface properties	Properties of particulates
Flow separation	Canopy structure	Brownian and eddy diffusivities
Micrometeorological interactions with the surface: Friction velocity Roughness height	Chemical/biological reactivity	Chemical reactivity
Relative humidity	Electrostatic properties	Density
Solar radiation	Geometry of roughness elements	Diameter
Temperature	Leaf area index	Diffusio-phoretic properties
Turbulence intensity	PH effects	Electrostatic properties
Wind speed	Penetration of canopy by contaminant	Hygroscopicity
	Prior deposition loading	Momentum
	Terrain characteristics	Shape, size
	Thermal properties	Solubility
	Wetness	Thermal properties

(after Sehmel, 1980 - modified and cited in Davidson and Wu, 1990) © Pergamon Press. The properties of gas contaminants have been removed from the original table to improve clarity

PM dry deposition onto a surface (in vegetation this can be a canopy, a tree, or individual leaves) involves three main steps: aerodynamic transport, boundary layer transport, and interactions with the deposition surface (Davidson and Wu, 1990). The atmospheric boundary layer is the air layer which is directly adjacent to the deposition surface (Burkhardt and Grantz, 2016). Aerodynamic transport is the transport of particles from the lower atmosphere to sublayers of the boundary layer by eddy diffusion (diffusion through turbulent transfer) and sedimentation, and boundary layer transport is the transport of particles across the atmospheric boundary layer via all four processes of dry deposition (Davidson and Wu, 1990).

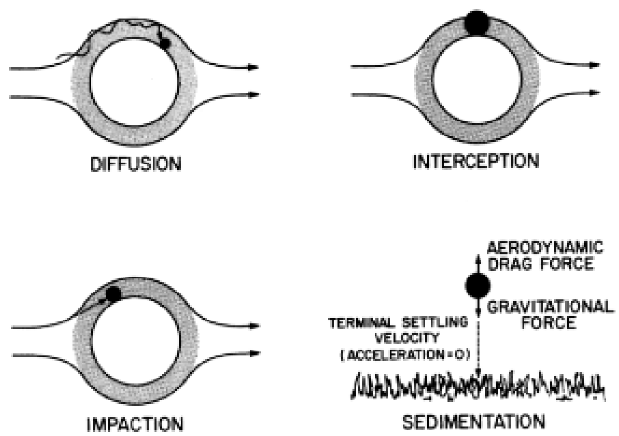


Figure 1-9 Diagrammatic representation of the main processes of PM dry deposition on a cylindrical object (Davidson and Wu, 1990).
The cylinder surrounded by the shaded area represents the deposition surface; arrows represent the sublayers of boundary layer; the black, solid circular, object represents a particle. *Please note that the air sublayers of the boundary layer are variable in thickness around an object and the arrows are only indicative of the general air flow.*

According to Roupsard *et al.* (2013), particulate deposition on vegetation is a uni-dimensional, and vertical, process; the total resistance to the deposition of particulates can be from the collective effects of aerodynamic, boundary, and surface resistance. Meteorological conditions in the atmosphere have a strong influence on aerodynamic resistance, and both aerodynamic and boundary layer resistance are influenced by atmospheric friction and shear velocity (caused by drag forces) (Bruse, 2007; Petroff *et al.*, 2008a). Surface resistance to PM deposition is influenced by the particle diameter and the micro-roughness or micro-topography of the plant (Slinn, 1982).

Larger PM sizes tend to deposit more rapidly compared to smaller PM sizes (Smith, 1977; Davidson and Wu, 1990). According to the model of 'theoretical particle deposition velocities' developed by Slinn (1982) (Fig.1.10), particles larger than PM_{10} are mostly deposited via sedimentation and the influence of sedimentation gradually lessens with the reduction of particle diameter due to decreasing mass; hence, this process is not important for particles with diameter less than 1 µm. Particles between 1µm and 0.1µm are mostly deposited via interception and some through impaction (Slinn, 1982). Particulate deposition per unit area of vegetation is higher closer to pollution sources (Weber *et al.*, 2014) and this may be due to the higher levels of impaction and interception on the vegetation due to the high concentration of pollutants near emission sources. Ultrafine particles behave in a manner similar to gas molecules and are mostly deposited via diffusion (Roupsard *et al.*, 2013). Fine particle deposition is heavily influenced by atmospheric stability (Wesely *et al.*, 1985; Lamaud *et al.*, 1994) and phoretic effects (thermosphoresis, diffusio-phoresis, and electrostatic) (Petroff *et al.*, 2008a). Deposition velocity, “the coefficient used to quantify the transfer of aerosol particles by dry deposition in the environment” (Roupsard *et al.,* 2013) was often used in modelling approaches to evaluate PM dry deposition on surfaces (Roupsard *et al.*, 2013b; Petroff *et al.*, 2008b; Slinn, 1982). According to Fowler *et al.* (2004) the influence of electrophoresis or thermal gradients were not considered in the deposition velocity model of Slinn (1982) which might have influenced the predicted behaviour of particles between 1 µm and 0.1 µm (i.e. charged metal-based particles).

The aerodynamic transport and boundary layer transport of different particle size fractions has been comprehensively studied under different meteorological conditions (Legg and Powel, 1979; Slinn, 1982; Petroff *et al.*, 2008b). Wind speed, wind turbulence, humidity and rainfall have considerable influence on PM deposition on vegetation (Litschke and Kuttler, 2008; Tomasević *et al.*, 2005). Croxford *et al.* (1996) and Fritschen and Edmonds (1976) found a significant positive relationship between PM deposition on forests and wind speed due to the increased levels of turbulence caused by high wind speed. The same effect was revealed by using trees in wind tunnels and exposing them to different types of particulates, such as NaCl droplets with a mean diameter of 1.28 µm (Beckett *et al.*, 2000a), mean diameter of 0.8 µm (Freer-Smith *et al.*, 2004), diameter in the 0.05 µm -15 µm range (Blanusa *et al.*, 2015) and uranyl acetate particles with a mean diameter of 0.82 µm (Ould-Dada, 2002). Wind tunnel approaches use precise measurements of wind velocity, particle diameter, and pollutant concentrations; nevertheless, the unidirectional air flow used in small laboratory wind tunnels are unlikely to be encountered in the ‘real world’ (Hwang *et al.*, 2011; Yan *et al.*, 2016a). Hwang *et al.* (2011) overcame these drawbacks by simulating an omni-directional airflow in a laboratory, and used free standing trees with a wide spacing (similar to an urban setting) to assess the influence of wind. Ultrafine size soot particles (similar to typical diesel engine particles in the range of 0.005 to 0.05 µm) and the deposition mechanism of Brownian diffusion were used in this study. The results of this study confirmed the earlier findings of the postive infulence of increasing widspeed on PM deposition. The porosity of vegetation and its drag force are also known to be affected by wind speed (Tiwary *et al.*, 2008) which, again, have an impact on PM deposition rates depending on the nature of the built environment (Abhijith *et al.*, 2017). The humidity of the environment has an influence on the deposition of hygroscopic particles (particles which absorb moisture and expand in size such as sulphate and nitrate particles). The deposition velocity of such particles increases with their growing particle mass (due to the impact of increased relative humidity), this process is limited at their saturation point i.e. the maximum size particles get to following absorption of water (Winkler, 1988).

Particulate trapping on leaves could be temporary since there is a possibility of remobilisation (rebound) (Fig. 1.10), e.g. wash-off by rain or rebound by wind (Currie *et al.,* 2008; McPherson *et al.,* 1994; Pye, 1987; Terzaghi *et al.,* 2013). McPherson *et al.* (1994) modelled the particulate interception of PM_{10} by urban trees and found that 50% of the captured particles were re-suspended under typical meteorological conditions. According to Gregory (1973) particles can be retained on surfaces depending on their texture and the particle mass, and high mass particles and sticky surfaces are favourable in this respect. In contrast, recent studies reported that smaller particle sizes are encapsulated by sticky waxy surfaces and tend to be retained on leaves without resuspension or being washed-off, whilst larger particles are loosely bound to the surfaces and tend to be readily washed-off or resuspended by wind (Ottelé *et al.*, 2010; Terzaghi *et al.*, 2013). According to Witherspoon and Taylor (1969), resuspension of PM by wind and rain after an hour from initial deposition can vary widely between species; for oak 91% of particulates were resuspended compared with just 10% from pine needles. Although increasing wind speed is described as favourable in increasing PM deposition velocity (Beckett *et al.*, 2000a; Hwang *et al.*, 2011), re-suspension is also higher at high wind speed (Gregory, 1973). However, as different modelling approaches used different assumptions and parameters, the actual PM deposition and retaining rates with reference to different particle size fractions is still unclear (Petroff *et al.*, 2008a).

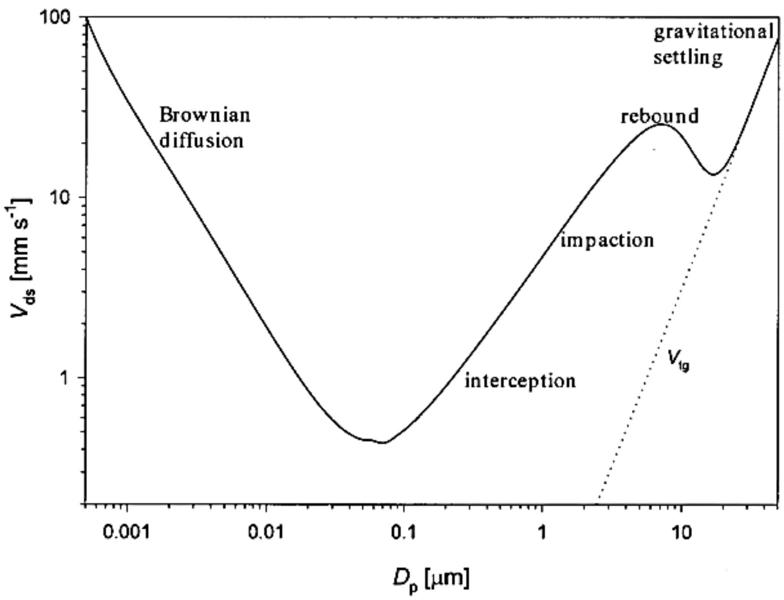


Figure 1-10 The variation of PM deposition velocity on short vegetation in relation to particle size (Slinn, 1982). Vds: deposition velocity; Dp: particle diameter

1.3.2. PM capture variation among different types of vegetation

1.3.2.1 PM deposition on trees and canopies

Particulate pollution removal by vegetation is explained as “one-dimensional vertical deposition on a homogeneous layer of vegetation in the form of a forest or field” (Janhäll, 2015) and this might not be an appropriate definition for urban vegetation (excluding parks and large gardens) as it mostly exists as individual trees, bushes, hedges, or other vertical greenery systems. Several studies have revealed the ability of trees to capture and retain PM under various environmental conditions (Beckett *et al.* 1998; Beckett *et al.* 2000b; Blanusa *et al.* 2015; Chen *et al.* 2017; Freer-Smith *et al.*, 2003; Fowler *et al.* 2004; Hofman *et al.* 2014; Jin *et al.* 2014; McDonald *et al.* 2007; Nowak *et al.* 2006; Randrup *et al.* 2001; Yang *et al.* 2015a). Of all the different types of vegetation, trees are considered to be the most effective type in capturing PM mainly due to their larger surface areas compared to shorter vegetation (Beckett *et al.*, 2000b; Fowler *et al.*, 2004; Jayasooriya *et al.*, 2017). However, Madders and Lawrence (1981) emphasised the importance of using a suitable planting design, choice of species, and appropriate location in tree planting to reduce pollutants. According to Manning and Feder (1980), tree canopies are particularly important as their great surface roughness enhances PM deposition by increased turbulence. Dochinger (1980) compared the ability of deciduous canopies (red oak *Quercus rubra* and sugar maple *Acer saccharum*) and evergreen coniferous canopies (red pine *Pinus resinosa* and blue spruce *Picea pungens*) in the reduction of dust and Total Suspended Particles (TSP) without particle size categorisation. The apparent tree cleansing efficiency (CE), which is the difference between dust collected in an open area and under a canopy as a percentage of total dust in an open area, was given as 27% and 38% for deciduous canopies and coniferous canopies respectively.

Using a Pollution Flux model, Nowak (1994) estimated that trees in Chicago captured 234 tons of PM_{10} in 1991; they also found large trees captured more PM than smaller trees due to the larger surface area of the former (i.e. trees with dbh above 76 cm captured 70x more particulates than trees with less than 8 cm dbh). However, the potential of high vegetation density to reduce PM dispersion (by obstructing the air flow) was not considered in this model and may have had an influence on the results (Janhäll, 2015). Beckett *et al.* (2000b) calculated the capture efficiency and deposition velocity of PM on broad-leaved tree species and conifers using a gravimetric approach in which particulates were collected by washing material off the leaves using water, followed by filtering and weighing the residue (hereafter referred to as "wash-off and filter method"). Of the five species tested (Corsican pine *Pinus nigra,* Leyland cypress *Cupressocyparis leylandii*, field maple *Acer campestre,* whitebeam *Sorbus aria* and hybrid poplar *Populus deltoides*), coniferous species with leaf needles (e.g. *P. nigra*) and the species with hairier leaves (e.g. *S. aria*) showed higher capture efficiency. However, ignoring or underestimating the water soluble fraction of particulates was given as a limitation of this method (Beckett *et al.*, 2000b); according to Li *et al.* (2012) the water soluble fraction accounts for some 45% of the total weight of particulates. Since there is unlikely to be an even distribution of water soluble PM fraction on leaves of different species of plants, the findings on relative capture rates can also be affected by this method. The water solubility of smaller PM size fractions, which are more important in relation to health, is an important consideration as they may dissolve faster compared to larger diameter PM. Insoluble PM held on the epicuticular waxes or surface microstructures of leaves may not have been washed-off in this approach and, hence, may not be weighed (Cheetham *et al.*, 2012). Freer-Smith *et al.* (2003) compared the deposition velocities and capture efficiencies of PM on seven species of trees (oak *Quercus petraea*, alder *Alnus glutinosa*, ash *Fraxinus excelsior*, sycamore *Acer pseudo-platanus*, Douglas fir *Pseudotsuga menziesii*, weeping fig *Ficus nitida* and eucalyptus *Eucalyptus globulus*) exposing them to NaCl particles with 1 µm diameter in a wind tunnel. *P. menziesii a c*oniferous species with narrow, small, leaf-needles had a higher PM capture efficiency and a higher deposition velocity compared to the other six species with broad leaves. Whilst wind tunnel approaches can provide precise measurements on wind velocity and the concentration of PM exposed, they do not approach the realities experienced in the 'real world' (see Section 1.3.1) where both the chemical and physical properties of atmospheric PM are more complex and their aerodynamic behaviour and deposition velocities are unlikely to be simulated by uniform sized NaCl particles (Davidson and Wu, 1990; Slinn, 1982; Yan *et al.* 2016). Hwang *et al.* (2011) also showed a higher effectiveness of needle-leaved species to remove PM (Japanese red pine *Pinus densiflora,* Japanese yew *Taxus cuspidata*) compared to broad-leaved species (sycamore *Platanus occidentalis,* keaki *Zelkova serrata*, and maidenhair tree *Ginkgo biloba*) using a modified wind tunnel approach with omnidirectional wind conditions (see Section 1.3.1) and airborne soot particles. Interestingly, among broad-leaved species, the deposition velocity for *P. occidentalis* was higher than that of *Z. serrata* or *G. biloba* suggesting an important role of surface roughness and leaf micro-structure in removing ultrafine soot particles.

McDonald *et al.* (2007) simulated the deposition velocity of PM_{10} on trees (considering total tree cover) using an atmospheric transport model in the West Midlands of England and Glasgow in Scotland. They estimated a 19% reduction of PM_{10} in the West Midlands would result from an increase in tree cover from 3.7% to 16.5%, and a 6% reduction of PM_{10} in Glasgow for an increase in tree cover from 3.6% to 8%. Tallis *et al.* (2011) estimated an annual removal of 852 to 2,121 tons of PM_{10} by urban tree canopies in a 157 kha area in the area covered by the Greater London Authority (GLA) using a modified version of UFORE. Ram *et al.* (2012) also studied the levels of suspended PM (larger than 0.2 µm in diameter) accumulated on plant canopies of banyan tree *Ficus benghalensis and* Indian fire tree *Polyalthia longifolia* in Kolkata City, India using the 'PM wash-off and filter method' and demonstrated that plant canopies can effectively capture atmospheric particulates.

Many of these studies noted the effectiveness of evergreen conifers to capture and retain PM (Beckett *et al.* 2000b; Dochinger, 1980; Freer-Smith *et al.*, 2003; Hwang *et al.* 2011; Ram *et al.,* 2012); besides, as the needles of conifers are retained over winter they can immobilise particulate pollutants throughout the year (Dzierzanowski *et al.*, 2011). Nevertheless, deciduous species have the advantage that captured particulates can be shed at leaf-fall and new shoots and buds provide new capture surfaces (Beckett *et al.*1998). Moreover, as the needles of conifers are retained for several years without being shed, they may reach a saturation point without recycling and diminish their ability to capture more particles (Beckett *et al.* 2000b; Dzierzanowski *et al.*, 2011). In addition, conifers are less tolerant to high traffic pollution and to winter salt, and hence their needles are potentially more susceptible to damage by the pollutants they capture the longer they are retained (Dzierzanowski *et al.*, 2011). However, studies have suggested that at least some particles captured on leaves are remobilised by rainfall and refresh the capture surfaces (Przybysz *et al.*, 2014; Wang *et al.*, 2015; Xu *et al.*, 2017); however, remobilisation behaviour of particulates following rainfall is not a well understood phenomenon and has not been extensively explored.

According to Janhäll (2015) most of the physics of PM deposition on vegetation can be explained by the state of an airstream passing over individual leaf surfaces rather than by the whole forest. Blanusa *et al.* (2015) conducted a leaf-level study on trees comparing the PM capture ability of leaves of Holm oak *Quercus ilex* L., a popular evergreen tree species in the Mediterranean urban environments, with leaves of three deciduous species (lime tree *Tilia cordata* Mill., London plane *Platanus* × *hispanica* Münch. and Turkey oak *Quercus cerris* L.) and the evergreen olive *Olea europaea* L., by using a NaCl aerosol and talcum powder (diameter in the 0.05 µm -15 µm range) in a wind tunnel, to identify potential alternative trees that can be used in Italian roadsides. Leaves of deciduous *P.* × *hispanica* and *T. cordata* showed the highest PM capture potential suggesting their suitability as PM filters. Leaf saturation isothermal remanent magnetization (SIRM) is strongly correlated with the presence of ferrimagnetic PM derived from anthropogenic sources such as industrial, traffic-based, and combustion sources (Maher *et al.*, 2008); Kardel *et al.* (2011) evaluated the inter-species variation in leaf SIRM on trees (two species of genus *Tilia* and common hornbeam *Carpinus betulus*) located in different environments in Belgium. Kardel *et al.* (2012) examined leaf SIRM of the same tree species used in Kardel *et al.* (2011) to assess the concentrations of the pollutants in different land use areas. Results of these studies indicated a considerable inter-species variation in their PM levels. Although this approach was recognised as providing data with high spatial density (Matzka and Maher, 1999; Gautam *et al.*, 2005), the resulting SIRM values (as a proxy of ferrimagetic PM) were not comparable with the values of other studies, given in mass or PNC for total PM, with variable chemical compositions.

Nowak *et al.* (2013 and 2006) reported that pollutant removal by trees can vary among cities based on their tree cover, leaf seasons, concentration of pollutants, and meteorological conditions. Liu *et al.* (2016) evaluated seasonal variation in PM capture and found higher deposition rates of finer particles in winter and higher deposition of coarse PM in spring due to changes in atmospheric conditions and concentration of PM. Prajapati and Tripathi (2008) also reported higher dust accumulation (total dust) on leaves in winter compared to summer or rainy seasons. Higher dust accumulation in winter has been explained by wet leaf surfaces and poor dust dispersion under foggy atmospheric conditions whilst lower accumulation in the rainy season has been explained by possible wash-off by rain.

1.3.2.2. The ability of PM accumulation by shorter vegetation compared to trees

Dzierzanowski *et al.* (2011) modified the 'PM wash-off and filter method' using chloroform as the solvent to collect PM trapped in epicuticular wax, and compared PM accumulation ($PM_{0.2}$-$PM_{2.5}$, $PM_{2.5}$ -PM_{10}, and PM_{10}-PM_{100}) on leaves of species of trees (*Fraxinus excelsior* L., *Platanus* × *hispanica* Mill. ex Muenchh., *Acer campestre* L. and *Tilia cordata* Mill.), shrubs (border forsythia *Forsythia* ×*intermedia* Zabel, Japanese spiraea *Spiraea japonica* L. and Common ninebark *Physocarpus opulifolius* (L.) Maxim.), and climbing common ivy *Hedera helix* L. grown in Poland. The results of this study showed a higher PM capture ability of the small-leaved shrub *Spiraea japonica* compared to other species. Although the modified wash-off and filter method could collect PM trapped in wax this approach ignored another fraction of PM (some non-polar organic particles) soluble in chloroform (Castelli *et al.*, 2002) in addition to the fraction dissolved in water. Sæbø *et al.* (2012) evaluated the PM accumulation ($PM_{0.2}$-$PM_{2.5}$, $PM_{2.5}$ -PM_{10}, and PM_{10}-PM_{100}) on leaves of 47 woody species (22 trees and 25 shrubs) in Norway and Poland using the modified wash-off and filter approach used in Dzierzanowski *et al.* (2011). The highest PM levels were found on leaves of a species of shrub Lace Shrub *Stephanandra incisa* and on leaf needles of Scots pine *Pinus sylvestris* in Norway and Poland respectively. The results of this study showed significant variation in PM accumulation among species; nevertheless, species with higher PM accumulation included both trees and shrubs, and variabilities were explained mostly with reference to their leaf characteristics rather than the type of vegetation. Evaluating seven species, common in both Norway and Poland, Sæbø *et al.* (2012) found higher PM accumulation levels on plants in Norway compared to Poland; the authors stated that these differences could partly be attributable to different PM deposition rates at different pollutant concentrations as elucidated by Nowak (2006). Reporting higher PM capture levels on the same species in both locations/countries compared to other species, they showed that the process of PM deposition is more species-specific than site-specific, and that the ability to capture PM can be generalised based on pollution concentration and the species of plant, irrespective of their different environments.

Popek *et al.* (2013) also evaluated PM accumulation (0.2 µm – 100 µm) on seven species of trees and six species of shrubs in central Poland using the wash-off and filter method; the highest PM accumulation was found on a species of shrub, Meyer lilac *Syringa meyeri* and, overall, most of the shrubs showed higher PM accumulation compared to trees. The lowest PM accumulation was found on a species of tree Southern catalpa *Catalpa bignonioides* and some trees and shrubs were found to be equally good in capturing PM, especially those which shared similar leaf characteristics. Mo *et al.* (2015) compared the PM accumulation levels on 10 species of shrubs and 25 species of trees in Beijing, China and did not find any obvious differences in their PM capture potential. Nevertheless, as there were slight differences observed in PM accumulation between shrubs and trees (even if they were not significant), planting both trees and shrubs was proposed as their variable heights can reduce PM on different spatial scales. Leonard *et al.* (2016) also evaluated PM (1.6 µm -100 µm) accumulation on sixteen species of plants in Greater Sydney, Australia using the wash-off and filter method and PM levels did not differ between trees and shrubs

Dochinger (1980) reported that PM levels on deciduous species without leaves (as in late autumn and winter) were not different from grasslands indicating low PM accumulation on grass. However, according to Smith and Jones (2000), grasses capture more particulates than shrubs, herbs, and trees due to grasses retaining dead leaves, and thus allowing capture of particulates over an extended period. Conversely, Fowler *et al.* (2004) reported that, in the West Midlands of England, the ability of woodlands to capture PM_{10} was three times higher than grasslands. Weber *et al.* (2014) studied deposition of PM_{10} on roadside herbaceous plants in Berlin using light microscopy and found that herbaceous plants are equally effective as woody species in immobilising urban traffic-generated coarse particulates. However, results of this study did not show any considerable capture difference compared to grass. Capture levels were found to vary between sites only based on the traffic density, leaf traits, and plant height. Hence, there is much debate about which types of vegetation actually capture more particulate pollutants.

In addition to the type of vegetation and species-specific characteristics, aerodynamics of trees can differentially affect PM concentration based on meteorological conditions and the morphology of the built environment, e.g. in a street canyon, plants with more gaps or spacing reduce PM levels in the air whilst dense vegetation increases the concentration of PM by obstructing the air flow (Abhijith *et al.*, 2017).

Ries and Eichhorn (2001), using a numerical model (MISCAM), Gromke and Ruck (2009) using a wind tunnel approach, and Langner *et al.* (2011) using low-volume samplers (IND, CEN 12341 reference sampler), demonstrated increased levels of PM concentration in street canyons due to dense tree canopies as they obstruct particle dispersion and air exchange, whereas some studies demonstrated that low-level hedgerows decrease the PM levels in street canyons (Gromke *et al.*, 2016; Li *et al.*, 2016). Pugh *et al.* (2012) estimated 60% reduction of PM_{10} in a street canyon by green walls using an improved atmospheric chemistry model (CiTTyCAT). Dense vegetation with low porosity which acts as a solid barrier allowing airflow over and above it, and vegetative barriers with high porosity that airflow can pass through, are also known to have a differential impact based on the nature of the built environment (Abhijith *et al.*, 2017; Gallagher *et al.*, 2012; Janhäll, 2015).

1.3.2.3. The importance of green walls and green roofs in the reduction of PM pollution

Impens and Delcarte (1979) reported that the use of street trees closer to roads have a greater impact in intercepting PM on their leaves than use of trees in less polluted areas, and the areas reported with higher concentrations of atmospheric PM were the places with a lack of greenery. However, irrespective of the ability of trees and ground vegetation to reduce PM pollution in open areas, there are several limitations in introducing vegetation into urban settings, these include: prevailing soil conditions (low soil fertility/contamination), available land area, compaction, availability of sunlight, the size of the trees compared to the buildings (potential shading effect), and sub-surface infrastructure (e.g. drainage systems) (Impens and Delcarte,1979; Johnston and Newton, 2004). Green walls and green roofs could overcome most of these limitations by transforming building walls to greenery; green walls may be particularly useful as they can potentially immobilize particles at street-level, unlike high rooftops. According to Macmillan (2004), roofs cover up to 35% of the land area in an urban setting, much of which could be converted to green roofs. Yang *et al.* (2008) estimated that 19.8 ha of green roof could remove 1,675 Kg of atmospheric pollutants including 14% PM_{10}. In Washington DC, if all the roofs were covered by vegetation, Deutsch *et al.* (2005) estimated that they could remove some 58 tonnes of atmospheric pollutants. Speak *et al.* (2012) assessed the potential of green roofs to capture PM pollutants in the UK atmosphere as a viable alternative to trees. They used SIRM to monitor atmospheric PM based on leaf magnetic particles on sedum *Sedum album,* red fescue *Festuca rubra,* creeping bentgrass *Agrostis stolonifera* and ribwort plantain *Plantago lanceolate* on green roofs. *A. stolonifera* and *F. rubra* were found as high impact plant species in the reduction of particulates. Assuming all the flat roofs in Manchester City Centre, UK were covered by vegetation, removal of 0.21 tons of PM_{10} per year was estimated.

According to Köhler (2006), the estimated vertical area which can be available for planting is approximately double the available ground area. Varshney and Mitra (1993) evaluated the particulate abatement capacity (PAC) of three hedge species, one climber, great bougainvillea *Bougainvillea spectabilis* and two shrubs golden dewdrop *Duranta plumieri* and Oleander *Nerium indicum* on near-road hedges in Delhi, India using dustfall measurements (dustfall jars and plastic films) and found a monthly mean dustfall of 70.3±7.8 t km^{-2} in front of the hedges and a monthly mean of 42.1±2.0 t km^{-2}

behind the hedges. The variability in PAC between species was in the order *D. plumieri* > *B. spectabilis* > *N. indicum*, and the authors concluded urban hedges were efficient PM barriers which could reduce road dust by 30-40% during most of the year. Tiwary *et al.* (2008) compared upwind and down-wind PM_{10} collection efficiencies of hawthorn *Crataegus* hedges using the 'wash-off and filter method' and demonstrated their potential to remove PM_{10} from the atmosphere. Most of these studies, which included PM size categorisation, found higher capture amounts of larger PM sizes suggesting a higher ability of vegetation to capture larger particles. However, in addition to differential deposition velocities and differential surface interaction of variable particle sizes, the higher mass of larger particles may result in higher weight when using gravimetric measurements.

According to pulmonary toxicity studies, the particle surface area and particulate count are more appropriate measures for smaller particles than particle mass (Sager and Castranova, 2009). The finer particles are numerically dominant in the atmosphere (i.e. ultrafine particles account for 80-90% of the total number), but are negligible in terms of mass and not accurately explained in mass-based approaches; PNC could give more insight in this respect (Hofman *et al.*, 2016). Ottele *et al.* (2010) used a Scanning Electron Microscope (SEM) and ImageJ image analysis software to quantify PNC ($\geq PM_{10}$, $PM_{2.5}$ – PM_{10} and $PM_{0.2}$ - PM_{10}) on leaves of *Hedera helix* picked from a noise barrier along a roadside and from a woodland in the Netherlands. Approximately 1.47×10^{10} m^{-2} particles were found on leaves taken from plants near the road compared to 8.72×10^{9} m^{-2} particles found on leaves taken from the woodland. Following the same PM quantification approach, Sternberg *et al.* (2011) evaluated $PM_{2.5}$ and PM_1 accumulation on leaves of *H. helix* (on direct green walls) grown along a high-traffic road, along a traffic lane in a residential area close to a construction site, and in a woodland in Oxford, UK. They also found effective PM capture levels by leaves of *H. helix,* approximately 2.9×10^{10} m^{-2} particles on leaves taken closer to the high-traffic road compared to 1.2×10^{4} m^{-2} particles reported from leaves taken from the woodland site. They also highlighted the added advantage of using green walls as they may protect the building wall by reducing the decay processes that impact stone walls (e.g. freeze-thaw cycles, UV degradation, air pollution). Dover and Phillips (2015) assessed the ability of green screens located along Bristol Street (a busy traffic-road) in Birmingham, UK, to remove PM_{10} and $PM_{2.5}$ pollutants and estimated a removal of 9,552 million particles by a 1 m^2 area of the green screen. Tong *et al.* (2016) used a CTAG model (Comprehensive Turbulent Aerosol Dynamics and Gas chemistry model) and a large eddy simulation (LES) on coniferous species to compare the effectiveness of roadside barriers in reduction of traffic-generated PM (12.6 nm - 289 nm). Findings of this research showed higher effectiveness of a 'combined tall vegetation-solid barrier scenario' and a wide vegetation barrier in the reduction of traffic-pollutants compared to green walls or solid barriers. They also noted an elevated PM concentration caused by upwind barriers. As these findings were only based on conifers, the use of other types of vegetation such as shrubs and grass to further evaluate these scenarios was recommended. Jayasooriya *et al.* (2017) estimated pollutant removal by integrated GI scenarios comprises trees, green walls and green roofs in Melbourne, Australia using i-Tree Eco software; hedges of *Laurus nobilis* (bay tree) were incorporated to simulate the effect of green walls, and *Eucalyptus macrocarpa* (mottlecah) roofs were incorporated to simulate the effect of green roofs. The study concluded a higher impact of trees to remove particles from the air and integrating green walls or roofs into forest tree models did not show any significant increase in PM reduction.

Studies evaluating the potential of green walls in the reduction of PM have mainly focused on climber vegetation (Cheetham *et al.*, 2012) or hedges with a single species of plant, apart from Varshney and Mitra (1993)'s work on total dust fall capture. However, Perini *et al.* (2017) evaluated PM deposition ($PM_{0.5}$ - PM_{10}) on leaves of two shrubs (*Cistus* 'Jessamy Beauty' and Jerusalem sage *Phlomis fruticose*), and two climbers (*H. helix* and Confederate jasmine *Trachelospermum jasminoides*) in a green facade located in Italy using the SEM/imageJ approach. The highest number of particles was found in the fine particle range (0.5–2.5 µm) and the highest PM densities were found on leaves of *T. jasminoides,* (approximately 4,300 particles mm^{-2}). However, this green wall was located above street level (approximately 5 ± 2 m) and so the values are not directly comparable with the results of street-level studies. As the PM composition and concentration in the atmosphere is different at different heights, measuring leaf PM accumulation at pedestrian-relevant heights was recommended when referring to human exposure levels (Maher *et al.*, 2008).

Research on the effectiveness of green walls or combined GI scenarios to capture and retain PM, with reference to different types of systems and variable species of plants have produced complex findings (Jayasooriya *et al.*, 2017; Ottele *et al.*, 2010; Perini *et al.*, 2017; Pugh *et al.*, 2012; Sternberg *et al.*, 2011; Tiwary *et al.*, 2008; Tong *et al.*, 2016). Despite the greater potential for delivering valuable ecosystem services by using a diverse collection of species, the value of living wall systems in the reduction of PM pollution has been overlooked. However, in a small-scale study evaluating $PM_{2.5}$ - PM_{10} capture by leaves of some shrubs and perennials using the 'wash-off and filter method', Shackleton *et al.* (undated) included a few species sampled from a living wall in Edgeware Road, London. Shrubs used in this study showed comparable capture efficiencies to trees. Prior to the research carried out for this thesis (Weerakkody *et al.,* 2017; Weerakkody *et al.,* 2018a; Weerakkody *et al.,* 2018b), there were no published studies specifically focused on living walls in this respect. Due to their vertical configuration and potentially different air movements, it is difficult to extrapolate the findings of tree literature directly to VGS. The studies focused on tree canopies showed that the dynamics in topography of the canopies directly influence surface resistance and turbulence of surrounding airflow patterns resulting in differential rates of PM deposition (Abhijith *et al.,* 2017; Davidson and Wu, 1990; Gallagher *et al.,* 2012; Janhall, 2015; Slinn, 1982). Therefore, in addition to the choice of species and type of vegetation used, dynamics in planting design might be important in the use of VGSs to immobilise PM. However, any such influence of planting design on the ability of PM capture and retention has not been explored in previous research.

1.3.3. Influence of leaf characteristics on PM accumulation

Once the particles are transported across the boundary layer, the ability of leaves to capture and retain these particles is driven by the interactions between particles and plant surfaces including geometrical properties such as leaf shape, size, orientation and surface morphology (Chen *et al.*, 2016; Freer-Smith *et al.*, 2005; Leonard *et al.*, 2016; Litschke and Kuttler, 2008; Petroff *et al.*, 2008a; Tomasević *et al.* 2005). According to Weber *et al.* (2014), the impact of leaf traits is more important than the type of vegetation in capturing particulates. Nowak (1994) suggested better PM capture performance of smaller leaves with rough leaf surfaces compared to large, smooth leaves. Of five species evaluated for PM deposition velocities, Freer-Smith *et al.* (2005) found higher deposition velocity/PM capture efficiency on species with smaller leaf needles (conifers) compared to broad-leaved species. However, higher PM accumulation on coniferous species has also been explained by invoking various other factors such as thick epicuticular wax layers, complex structure/leaf arrangement, evergreen habit, high total surface area, and aerodynamic properties (Beckett *et al.*, 2000a; Dzierzanowski *et al.*, 2011; Godzik *et al.* 1979; Wang *et al.* 2011). A study conducted by Popek *et al.* (2013) to evaluate PM capture levels on trees and shrubs, found the highest PM accumulation on leaves of a species of shrub sorbaria 'Sem' *Sorbaria sorbifolia* (25.68 $\mu g \cdot cm^{-2}$) and a tall tree ash *Fraxinus pennsylvanica* (20.58 $\mu g.cm^{-2}$), irrespective of their different leaf morphology and dimensions. The only common character that both species shared was smaller composite leaves, and hence the authors suggested an inverse relationship between leaf size and PM accumulation. According to Farmer (2002), smaller leaves potentially increase PM capture levels by increasing air turbulence around plants, in contrast, Sæbø *et al.* (2012) showed that there was no correlation between leaf PM accumulation and their size. Leonard *et al.* (2016) found higher PM accumulation on smaller leaves with shorter petioles (Australian rosemary *Westringia fruticose* and prickly paperbark *Melaleuca styphellioides*) compared to larger leaves with longer petioles. Although the literature suggested that smaller leaf sizes might be favourable in PM capture, there were no comprehensive evaluations conducted on the effect of individual leaf size on PM capture using a range of species, except for comparisons between conifers and broadleaves.

The shape of deposition surfaces is known to have an influence on surrounding airflow patterns as they create differential drag forces which affect the level of turbulence, resulting in different rates of PM deposition (Davidson and Wu, 1990; Gemba, 2007). Leonard *et al.* (2016) compared PM accumulation on leaves of different shapes (obovate, linear, elliptical, lanceolate, and needle-like/acicular) and different leaf arrangements (alternate, whorled, and opposite). Although leaf needles have often been cited as effective PM filters compared to broader-leaved species (Beckett *et al.*, 2000b; Fergusson *et al.*, 1980; Freer-Smith *et al.,* 2005), lanceolate leaves showed the highest PM levels, which were significantly higher compared to linear, elliptical and needle-like leaves. However, leaf arrangements did not show any significant impact in this respect. In contrast to these findings on linear leaves, Shackleton *et al.* (undated), reported linear ("grass-like") leaves as the most effective PM filters out of 16 species with variable leaf shapes. However, Leonard *et al.* (2016) noted that the differences in PM accumulation on leaves are attributable for the collective impact of leaf traits than individual characters. Except for these studies, previous research has focused more on canopy architecture or shape (Beckett *et al.*, 2000b; Nowak *et al.,* 2013; Ram *et al.*, 2012) than leaf level analysis; therefore, there is no clear picture about the effect of individual leaf shapes on PM accumulation.

Complex deposition surfaces are known to create an intricate turbulent airflow and hence, leaf micro-roughness and surface characteristics have a considerable impact on PM deposition (Beckett *et al.*, 1998). Specific leaf micro-morphological features including surface roughness, leaf hair, trichomes, stomata, glands, epicuticular wax, scales, furrows, and veins have a considerable impact on particulate accumulation on leaves (Pugh *et al.*, 2012; Ram *et al.*, 2012; Shackleton *et al*, undated; Terzaghi *et al.*, 2013). Leaf hairs are known to enhance PM acumulation in several ways: increasing surace area/capture area with their protrusions, by preventing captured PM resuspension to the atmosphere, and by creating a complex micro-topography on leaf surfaces (Prusty *et al.*, 2005; Qiu *et al.*, 2009; Weber *et al.*, 2014). According to Fernández *et al.* (2014), the hydrophobicity of leaf hair also has a positive impact on attracting and capturing charged particles which are mainly metal-based. Kardel *et al.* (2011) found higher leaf SIRM values (indicating higher PM_{10} levels) on hairy leaves of a *Tilia* sp. compared to a non-hairy *Tilia* sp.. Beckett *et al.* (2000b), Kardel *et al.* (2012), Räsänen *et al.* (2013) and Shackleton *et al.* (undated), also found a positive impact of hairy leaves on PM accumulation; in contrast to these findings, Perini *et al.* (2017) found a negative impact of leaf hair on PM accumulation by analysing the leaves in a VGS.

The epicuticular wax on leaf surfaces has frequently been cited as an important character in leaf PM capture and retention, as particles can be strongly bound to sticky waxy surfaces (Barima *et al.*, 2014; Hofman *et al.*, 2014; Mo *et al.* 2015; Räsänen *et al.*, 2013; Sgrigna *et al.* 2015). Popek *et al.* (2013) found a positive correlation between the amount of PM (2.5–10 µm) held on leaf surfaces and the quanitiy of leaf wax. In contrast, Dzierzanowski *et al.* (2011) did not find any correlation between PM accumulation on leaves and the amount of leaf wax; however, they suggested an importance of chemical structure and composition of leaf wax on PM accumulation rather than simply the wax quantity. Some studies found a negative influence of leaf waxes on PM capture, based on their structure and chemical composition (Faini *et al.*, 1999; Kardel *et al.*, 2012; Leonard *et al.* 2016), e.g., the self-cleansing ability of some wax tubules on leaves is known to reduce their ability to accumulate PM (Wang *et al.*, 2011). Waxy, hydrophobic, leaf surfaces, expressed by a large drop contact angle (DCP) or small leaf wettability is unable to accumulate as many magnetic particles as hydrophilic leaf surfaces with large wettability (Kardel *et al.*, 2011).

Burkhardt *et al.* (1995) reported high PM accumulation (based on particles with 0.5 µm diameter) levels on the stomatal regions of leaves in a wind tunnel study; leaves with a high stomatal density and higher transpiration rates were found to capture more PM by increased diffusive deposition (Beckett *et al.*, 1998) and by maintaining a moist layer on the surface (Tong, 1991). Particles impacting on smooth surfaces readily rebound to the atmosphere (Davidson and Wu, 1990) whilst rough leaf surfaces with dense grooves and ridges are better at both capturing and retaining PM (Kardel *et al.*, 2012; Ram *et al.,* 2012; Zhang *et al.*, 2017). Barima *et al.* (2014) found significantly higher PM accumulation on ridged leaf surfaces compared to smooth waxy surfaces. Kardel *et al.* (2011) found higher leaf SIRM values on rugose (ridged) leaves of *C. betulus* compared to smooth leaves of a *Tilia sp*.

According to Chamberlain (1975), sticky surfaces are better at capturing coarser PM, whilst the capture of finer PM is positively influenced by surface roughness. Sæbø *et al.* (2012) evaluated the correlation between leaf PM accumulation and most of the important micro-morphological features. Leaf PM accumulation showed a positive correlation with density of leaf hair and quantity of leaf wax; however, there was no correlation shown with leaf roughness. Liu *et al.* (2012) also evaluated the influence of leaf micromorphology (leaf-wax, trichomes, cuticle, and stomata) on PM accumulation and found a positive impact of stomatal density, trichomes and surface grooves and a negative impact of cuticular wax which reduced the PM accumulation on leaves. Ram *et al.* (2012) also studied these impacts using *F. benghalensis and P. longifolia* and found a positive impact of all examined features: stomatal frequency, number of hairs, length of trichomes, and leaf-roughness, on PM accumulation and on preventing them from being re-suspended. More particles were trapped on the adaxial surfaces of the leaves, suggesting the influence of sedimentation, and particles on the lower surfaces were found to be trapped in stomata.

Even with numerous studies conducted to evaluate the role of micromorphology in leaf PM accumulation, using several different types of vegetation and species, they produced mixed results, probably due to the synergistic effects of the specific set of leaf characteristics exhibited by the different species.

1.4. Rationale for the present study

Literature on the ability of trees and other types of vegetation to capture and retain particulates implies an important potential of living walls to remove particulate pollutants from the atmosphere. Traffic-based PM pollution has been particularly identified as a serious threat to human health, and as an environmental burden especially affecting urban dwellers. Therefore, evaluating the ability of living walls to capture and retain traffic-based PM will possibly unlock their potential to act as near-road PM filters. The optimal species composition and suitable planting designs for living walls to effectively capture and retain PM are not addressed by existing research. Since living wall plants are mostly evergreen, leaves are retained for a long period of time; therefore, evaluating the process of accumulated PM remobilisation due to rainfall is crucial to understand the long-term usefulness of these systems. Without such information, it is impossible to quantify the actual benefits of living walls in particulate pollution reduction. Hence, the present study will examine the effectiveness of living wall systems in the reduction of particulate matter pollution with special reference to traffic-generated particulates. The value of different living wall species in the reduction of PM pollution will, of necessity, be carried out in a local context allowing the effect of environmental conditions such as weather and different emission sources to be taken into consideration. However, the implications of this work will be valid at larger scales, e.g., the findings of these experiments can be applied in modelling approaches at scales ranging from the very local, through streetscape to settlement. The outcome of this research will potentially maximise the benefits of living wall systems which will help in providing a short-term solution to the problem of PM pollution to improve human wellbeing.

1.5. Aims and objectives

1.5.1. Aims of the research project

This research project aims to explore the potential of living wall systems to reduce particulate pollution generated from traffic-related sources. The main focus being to optimise the overall PM capture efficiency of living walls via exploring the optimum species characteristics and other influential factors required to trap traffic-generated particulates.

1.5.2. Objectives of the research

- Evaluate the inter-species variation in PM accumulation on leaves and identify optimal species composition for a living wall system to effectively remove atmospheric particulates in important PM size fractions.
- Identify important leaf morphological characters which have an impact on PM capture and retention on leaves
- Identify the elemental composition of PM including potentially hazardous elements to human health which can be immobilised by living wall plants
- Assess the impact of rainfall on remobilisation of PM captured on leaves
- Identify effective planting designs to enhance the PM capture potential of living wall plants

Chapter 2 : Generic Methods

This chapter contains generic information on materials and methods used in this research. Detailed information on the methods followed in each experiment are given in the respective chapters.

2.1 Site selection

The selection of living walls was made based on their relative location to potential pollution sources and accessibility for sampling. Existing living wall systems and an experimentally manipulable living wall system located in the cities of Stoke-on-Trent, Staffordshire and Birmingham, West Midlands, UK were used to evaluate their impact in the reduction of PM pollution (Fig. 2.1). A pilot study was conducted to develop a sampling protocol for this research using a few common living wall species present in an existing modular living wall system and a green screen (*Hedera helix* screen) located in Staffordshire University (detailed in Chapter 3). An existing modular living wall located in New Street Station, Birmingham, UK was selected to evaluate the role of living walls in the reduction of PM pollution caused by rail-traffic. An experimentally manipulable modular living wall, designed for the purpose of this study and installed along Leek road adjacent to Staffordshire University's Science Centre was employed to evaluate its impact on reduction of PM generated by motor-traffic and to study the optimal planting design to capture and retain PM. An existing living wall system in Staffordshire University was re-planted with scrambling species (*Rubus spp.*) with different morphologies to identify any differential impact of these scrambling species in the reduction of particulate pollution. PM remobilisation behaviour due to rainfall was studied using some species present in the experimentally manipulable living wall. In addition, the ivy used in green screens was also studied to evaluate its PM remobilisation behaviour as this has not been addressed in previous studies on green facades (Dover and Phillips, 2015; Ottelé *et al.*, 2010; Sternberg *et al.*, 2011); the implications of this work were relevant to living walls in general as *Hedera helix* is commonly used in all VGSs. Site descriptions and species descriptions of each of these systems are detailed in the relevant chapters.

2.2 Sample collection and storage

Leaf sampling was carried out in daylight only, and during dry weather conditions having at least three consecutive non-rainy days immediately preceding sampling. In all the experiments, every species was equally sampled, as same number of leaves from each species on each sampling occasion, to avoid any differential influence from weather changes between sample occasions. Sample numbers and the number of sampling occasions varied and were based on the number of species studied in each experiment, which are detailed in the relevant chapters. Leaves were randomly sampled using a random number table at accessible heights avoiding damaged or unhealthy leaves. Leaves were hand-picked and stored in plastic containers in such a way that they did not rub against one another or against the container and sealed to minimise particles falling-off the leaves or cross contamination (Fig. 2.2). Subsequently, samples were carefully transferred to the laboratory and analysed fresh or stored in a refrigerator (approximately 9 ºC) in the same containers until analysis within two days of sampling to avoid possible structural changes due to dehydration.

Figure 2-1 Images of a) a section of the living wall located adjacent to New Street train station Birmingham, b) the experimentally manipulable, roadside, living wall located at Staffordshire University adjacent to Leek Road, c) the existing living wall located within the campus of Staffordshire University, d) an existing living wall module at Staffordshire University re-planted with climbers e) the green screen located on the Staffordshire University campus adjacent to Leek Road.

Figure 2-2 An image showing how leaves were stored in a plastic container during sampling.

2.3 Evaluating inter-species variation of PM capture by plants

2.3.1 Quantifying the PM accumulation on leaves

2.3.1.1 Rationale for method selection

The literature revealed several different methods used to estimate PM deposition on vegetation and which mainly expressed PM accumulation in terms of mass concentration of the particles. However, as recent epidemiological research and pulmonary toxicity research suggest PNC is a more appropriate measure compared to their mass concentration (Kelly and Fussell, 2012; Sager and Castranova, 2009), this study used the SEM/imageJ approach (see below) (Ottele *et al.*, 2010; Sternberg *et al.,* 2010) to quantify and size-categorise the particulates captured on leaves. The added advantage of this method is that it provides additional information on PM size distributions, which helps identify the important leaf characteristics associated with PM capture. Environmental Scanning Electron Microscopes (ESEM) are particularly useful as they can operate with a low vacuum which allows the visualisation of non-conductive specimens (e.g. leaves, due to their high carbon content) without any conductive coating (such as volatilized gold); the signals from Back Scattered Electrons (BSE) provide high contrast images which differentiate leaf contaminants from the leaf surfaces (Ensikat, 2010). Leaves, in particular, have the advantage of having a cuticle that can minimise dehydration. However, imaging the small particles of interest in this study (i.e. 10 µm and below) needs high magnification. As the finest particles to be studied are in the nano-range the actual areas scanned are very small compared to the area of intact leaves. Therefore, the use of a large number of samples per leaf was essential to make estimations that could be considered robust. Further to this, as species with fragile leaves and thinner cuticles are susceptible to dehydration, compared to other leaves, they could not be kept in the vacuum chamber for a long period.

2.3.1.2. Environmental Scanning Electron Microscope/ImageJ approach

An ESEM, Model JSM-6610LV, was employed to image the particulates captured on leaves; the resolution of this microscope was 3.0 nm and 4.0 nm in high and low vacuums respectively. All the leaf specimens scanned (both natural and synthetic) throughout the study were uncoated and visualised only under a low vacuum (LV mode). The smallest particle size that could be imaged with enough resolution and less conductive charging using this technique was 0.1 µm (100 nm) in diameter using 1,000x magnification. Sampling of leaf sections followed the protocol developed using a pilot study detailed in Chapter 3. In order to visualise particulates using the ESEM, leaf sections were mounted on aluminium stubs using double-sided carbon adhesive tabs (Fig. 2.3a). Aluminium stubs holding the leaf sections were then inserted into a sample holder (Fig. 2.3b) and ESEM micrographs were taken using BSE at a range of magnifications (magnifications used are specified in the relevant chapters). Micrographs for each experiment were taken using the same working distance and same accelerating voltage as appropriate. Contrast and brightness levels were maintained as consistent as possible to avoid any difficulties in defining the threshold of the image analysis process. Each batch of micrographs (same species and magnification) were taken twice both with and without labels; the name of the species of plant, scale of the image, magnification, working distance, pressure inside the LV, the type of electron signals used and the spot-size of the electron beam were included in the labels. Another set of micrographs (at least 10 from each batch) were taken from the same scanning areas using Secondary Electron (SE) signals to help define thresholds in image analysis.

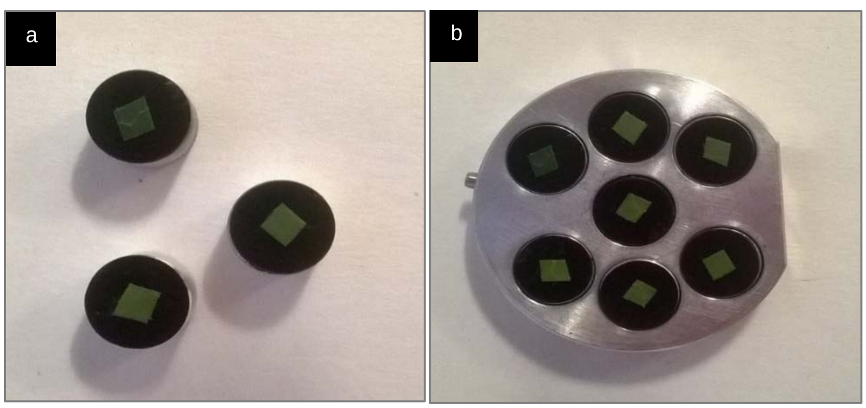


Figure 2-3 Images of a) leaf sections mounted on aluminium stubs using adhesive carbon tabs b) aluminium stubs carrying leaf sections held in an aluminium sample holder for particulate visualisation using the ESEM

ImageJ image analysis software version 1.5 (64 bit) was used to quantify and size-range PM imaged in micrographs (Collins, 2007) following the procedure given in Fig. 2.4. Micrographs without labels (Fig. 2.5) were processed to quantify the particles to avoid the interference of labels with PM counting. The most appropriate threshold available in the auto threshold tool was chosen (Ferreira and Rasband, 2012) by comparing the micrographs with their respective SE images at high resolution to ensure that the background (leaf surface) was subtracted and only the particles were counted. As there were a large number of micrographs in each batch of every experiment, they were analysed together using the Batch Processing tool. PM size ranges of interest are detailed in relevant chapters.

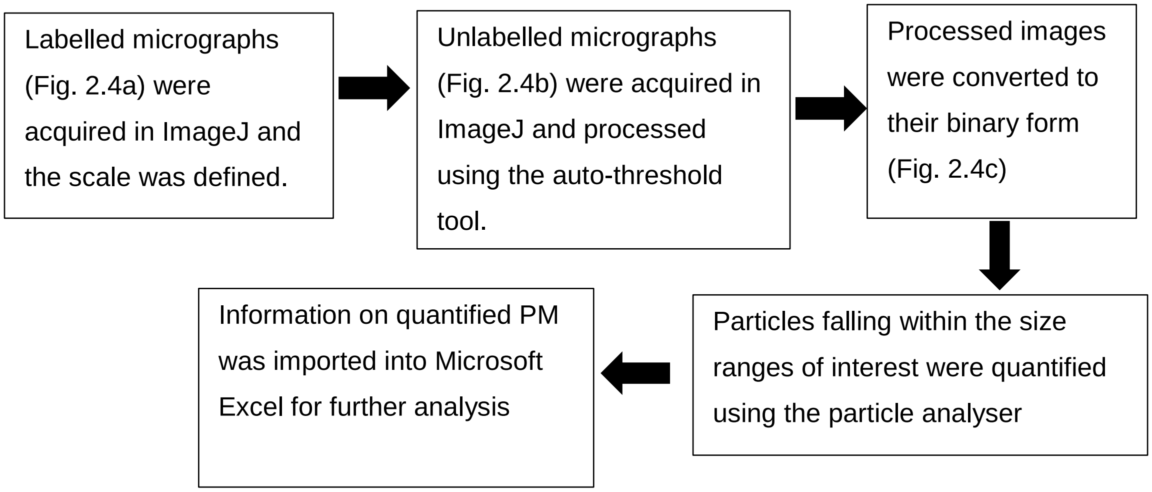


Figure 2-4 The detailed procedure followed in PM quantification using ImageJ software

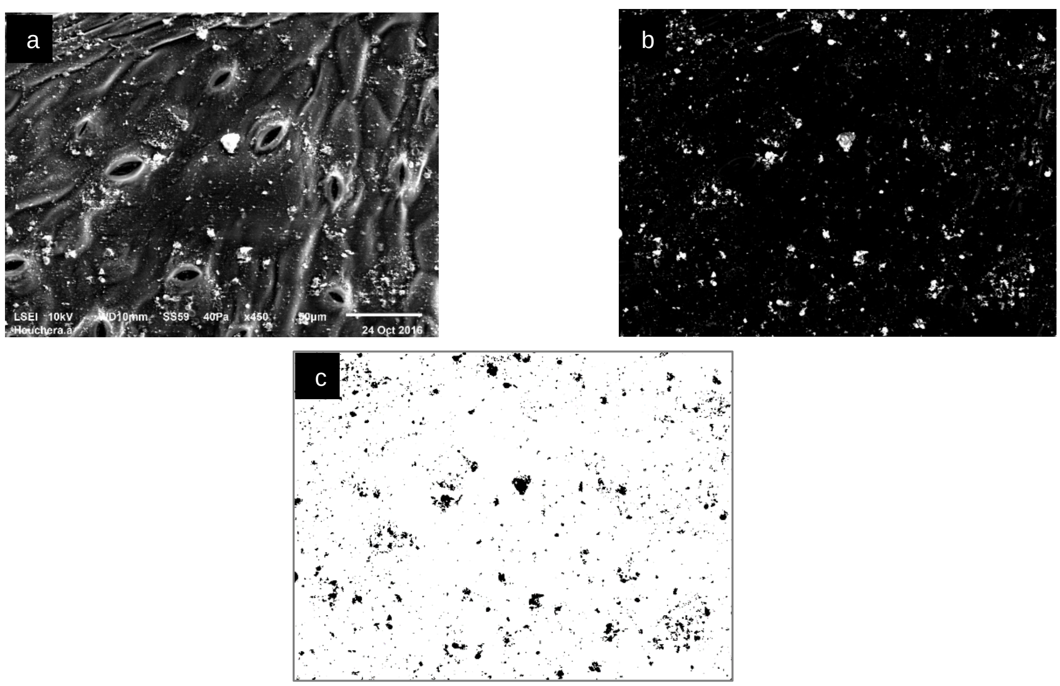


Figure 2-5 ESEM micrographs of a leaf section of *Heuchera amaricana* (x450) a) Secondary electron (SE) view and b) Back scattered electron (BSE) view and c) a processed image of a micrograph using ImageJ

2.3.2. Measuring the Leaf Area Index (LAI)

Different species of plants have differential leaf distribution in space; some species have many leaves closely arranged as canopies while others have a few sparsely arranged leaves. Therefore, the available leaf area to capture and retain particles is different between species and age/size of plant. The inclusion of total leaf area into analyses was thus essential to estimate their potential to capture and retain PM. The leaf area of plants can vary both spatially and temporally due to dynamics in their growth and development with age structure, seasonal variations, and site-specific conditions (Vose *et al.*, 1994). The LAI, "the ratio of leaf area to unit ground surface area (dimensionless)" (Zheng and Moskal, 2009) provides the information required to assess the available leaf area in different species of plant. Since the focus of this study was vertical greenery systems, LAI was measured relative to a unit vertical area of living walls. Ten individual leaves from each species were randomly sampled and photographed along with a ruler by the leaves. Subsequently, these photos were imported in ImageJ and the surface area of individual leaves was measured; the scale of each image was defined using the ruler. The mean leaf surface area of ten individual leaves of each species was then calculated. The number of leaves distributed on a unit vertical area of plants was counted using a 10 cm X 10 cm hand-made quadrat (Fig. 2.6). As living wall plants are mostly small shrubs or herbs, a 10 cm X 10 cm quadrat was considered appropriate. The mean number of leaves present within a quadrat was calculated using three random quadrats per species. The LAI of each species was then calculated using the following formula:

$$\text{LAI} = \frac{\text{Mean surface area of an individual leaf x Mean number of leaves per quadrat}}{\text{Total area of the quadrat}}$$

The ability of each species of plant to remove PM was estimated using PM accumulation and their LAI values. The calculations used are detailed in the relevant chapters as appropriate (Chapters 4 and 5).

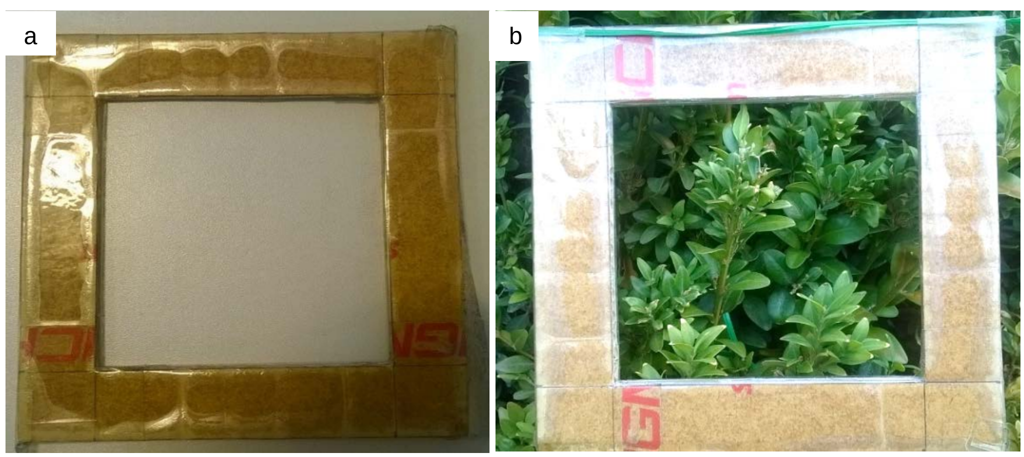


Figure 2-6 An image of the hand-made quadrat (10 cm X 10 cm) used to measure the LAI and b) a sample image of the quadrat being used in the field

2.3.4. Statistical analysis of data

R statistical software version 3.2.5 (R Development core team, 2016) was used for statistical analysis of data throughout this study. Statistical models used in different experiments were decided based on data distribution and variance. The Shapiro-Wilk test was used to test data normality. A Linear Model (LM) or a Generalised Linear Model (GLM) were fitted depending on data distribution (detailed in the relevant chapters). The R packages used in each analysis are given in the relevant chapters.

2.4 Elemental analysis of the particulates

The ESEM used in this study was equipped with Integrated Calibration and Application (INCA) microanalysis software to analyse the chemical composition of specimens using Energy Dispersive X-ray (EDX). The settings used in the analysis are detailed in the relevant chapters (Chapters 4 and 5). EDX analysis was identified as the most appropriate method to analyse the chemical composition of the PM captured on the leaves in-situ. EDX acts as both a qualitative and quantitative analyser to identify elements and quantify the amounts in terms of percentage weight of the element with reference to the total weight of the particle (Wt%) by detecting the signals of X-ray emitted from the specimen (Oxford Instruments, 2006).The limitation of this method was the inability to differentiate the organic component of particles from leaf material. Although the 'Point and ID' analyser of the software minimises this by scanning specific particles, there is still some interference from the background plant surface, particularly for very small particles which are difficult to capture individually. A few trials were conducted to explore the feasibility of using Fourier-Transform Infrared Spectroscopy (FTIR) and Raman spectroscopy to analyse the organic compounds of these particles by comparing the spectrums given for pre- and post-washed leaves. However, both these methods resulted in spectrums associated with leaf wax and pigments (Fig. 2.7) (Greene and Bain, 2005; Ribeiro da Luz, 2006) and were not successful in analysing the chemical composition of the particulates in-situ.

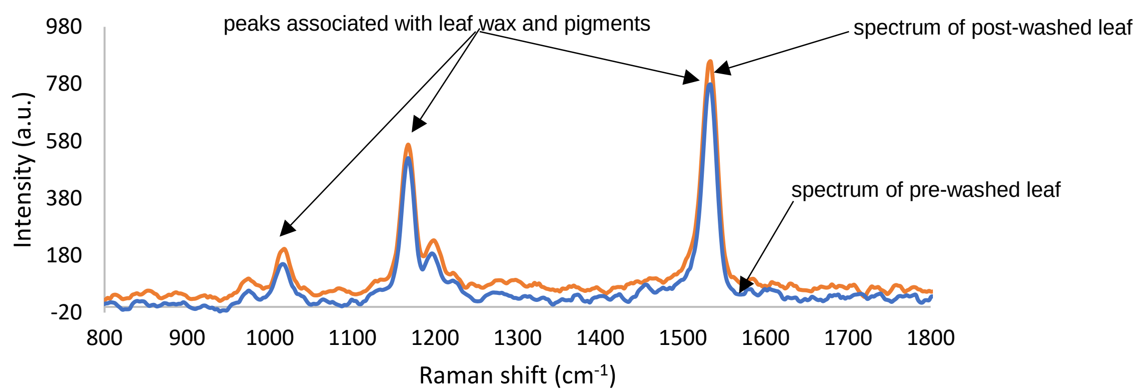


Figure 2-7 A Raman spectrum (514 nm excitation laser) of a pre- and post-washed leaf of *Heuchera americana* collected from *a* living wall located along Leek Road, Stoke-on-Trent, UK.

2.5. Identifying best species characteristics to capture and retain PM

2.5.1. Evaluating the correlation between leaf micromorphology and PM accumulation

Leaf micromorphological characters were imaged using the ESEM at a range of magnifications and different resolution and contrast levels as appropriate. The detailed experimental procedure is given in chapter 5. Some characters were not manually quantifiable due to their complex arrangement (e.g. surface ridges, grooves, elongated trichomes in *Rubus sp.*) and an alternative method was used to estimate their density using a Geographical Information System (GIS). In order to quantify micromorphological features using ArcGIS (ArcMap 10.4 © 2015 ESRI), leaf sections focused on each micromorphological feature on each species were scanned using the same resolution, contrast, and scale as appropriate. Each batch of micrographs were taken twice both with and without labels to obtain the information on scale and for image analysis respectively. The images were analysed in ArcMap10.4 software using the extensions of Spatial Analyst and ArcScan. The micrographs were acquired in software and the micromorphological characters on each micrograph and on each leaf were quantified following the procedure given in Fig. 2.8. The mean percentage area of each leaf surface covered by each micro-morphological character was estimated using 3 random micrographs from each adaxial and abaxial surface. Any significant correlation between specific leaf characteristics observed and their PM accumulation was identified using a Generalised Linear Mixed-effect Model (GLMM).

2.5.2. Evaluating the role of individual leaf traits on PM capture

Leaves of different species vary due to their different morphological traits, and the individual impact on PM capture and retention of these traits can be modified by synergistic or antagonistic effects. Assessing the impacts of each leaf trait on PM capture required additional experiments controlling for additional variables. Therefore, standardised experimental rigs were designed using natural and synthetic leaf models to understand the impact of individual leaf characters. Detailed experimental procedures are given in chapter 6.

2.6 Identifying the optimal conditions for a living wall system to act as an effective PM filter

Based on the outcome of all experiments discussed above, and on the findings of PM remobilisation by rainfall (methods are given in Chapter 7), best species compositions and optimal species characteristics for a living wall to act as an effective PM filter were identified. In addition, the impact of planting design on PM capture was evaluated (methods are given in Chapter 5) to identify a suitable planting design for a living wall in order to enhance its potential for PM capture.

Segmentation and Classification of the micrograph

Different micromorphological features were categorised into different classes using a range of training zones. Typically twenty images were used, although more were processed for complex images. Classified images, as different micromorphological features in different colours, were produced performing Interactive Supervised Classification.

Classified features were sampled allocating 500 random points using the Create Accuracy Assessment Point tool. The number of points that fell on areas of the intended character was quantified using structured query language (SQL).

Random sampling was repeated 5 times on each classified image and the mean number of random points that fell on areas of the intended character was calculated.

The area covered by the examined character was estimated as a percentage out of 500 points.

The mean percentage area covered by each character on each leaf surface, separately on adaxial and abaxial surfaces, was estimated using 3 random micrographs.

Figure 2-8 The detailed method used to quantify micromorphological characters on leaf surfaces using ArcMap10.4.

Chapter 3 : Pilot study - developing a protocol for leaf section selection

3.1 Introduction

Leaves have areas with different morphology (e.g. hairs/trichomes, ridges, groves, stomata, and pits), and hence potentially uneven PM distribution on their surfaces (Mitchell *et al.*, 2010). Therefore, sampling leaf sections from the areas with the most variable PM levels may result in considerable deviations from the mean PM levels on the leaves and hinder analysis. The general distribution of PM on leaves was thus evaluated to identify the areas with low variability in PM distribution for subsequent leaf section sampling.

3.2 Site description

An existing living wall and an existing green screen located on the Staffordshire University campus were used in this study. The former was installed in May 2011 for a biodiversity research project (Chiquet, 2014) and the latter installed as green fencing. Both systems were installed by Hedera Screens/Mobilane ®. The living wall was a north-facing rockwool-based modular system, 14 m x 2.4 m in size and located on a University building wall (Fig. 3.1a). The green screen used in this study was an artificially irrigated (self-sustained after established) hedera screen with *Hedera helix* L. (English ivy var. woerner) and located facing Leek Road, Stoke-on-Trent, 5 m from the roadside (Fig. 3.1b). Potential anthropogenic PM sources for both these systems were motor traffic and road dust. Four plant species with different morphotypes: hairy leaved *Geranium macrorhizzum* L*.* (Geranium macrorhizzum)*,* linear ("grass like") *Acorus gramineus* Sol. (grass-leaf sweet flag) and smooth leaved *Heuchera* 'Paris' *L.* (Alumroot 'Paris') and waxy leaved *Hedera helix* (Fig. 3.2), taken from the living wall and from the ivy green screen were used in this experiment.

Figure 3-1 Images of a) the living wall system and b) the green screen, used in this experiment, located at Staffordshire University, Stoke-on-Trent, UK.

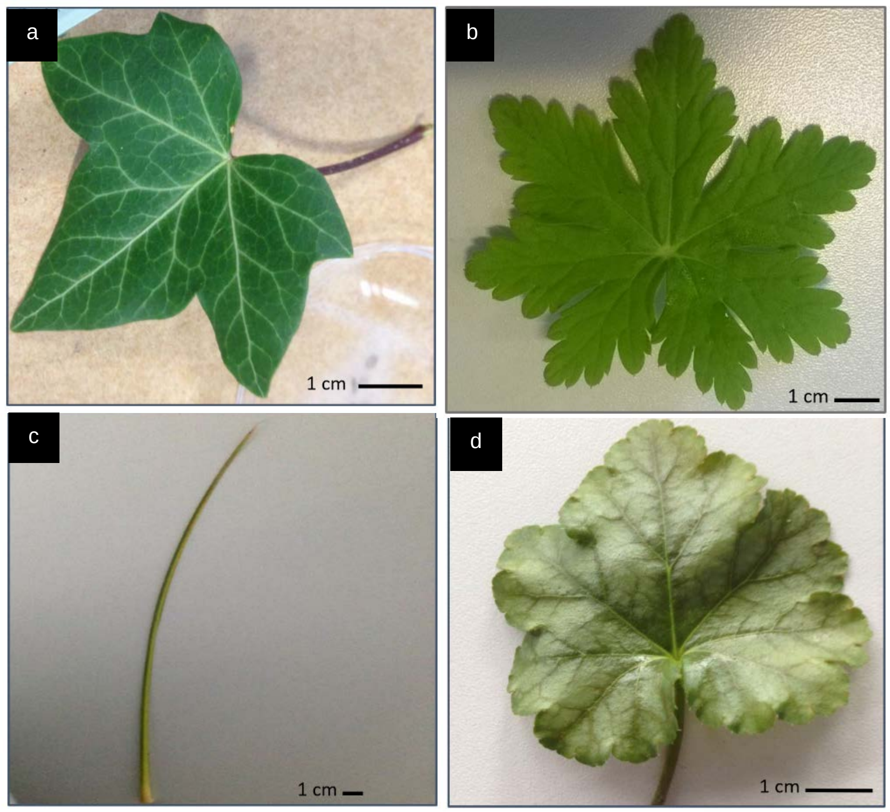


Figure 3-2 Images of leaves of a) *Hedera helix,* b) *Geranium macrorrhizum*, c) *Acorus gramineus* and d) *Heuchera* 'Paris'.

3.3 Methodology

3.3.1 Sampling

Twenty leaves were randomly sampled from each of four plant species on several occasions between May and August 2015. Sample storage and transfer to the laboratory followed the same approach given in Chapter 2 under generic methods.

3.3.2 Analysis of PM distribution on leaves

Ten leaves from each species were used to examine the PM distribution along the horizontal axis and ten for the vertical axis (Fig. 3.3). Leaf transects with 5 mm width were cropped along the horizontal and vertical axis of the leaves (Fig. 3.3) and mounted on a large aluminium sample holder using double-sided carbon adhesive tape (Fig. 3.4). The leaves of *A. gramineus* were too narrow for an x-x transect (approximately 4 mm to 5 mm) and they were only tested for PM distribution along the vertical axis. The transects that were longer than the diameter of the sample holder were cut into halves and mounted as 'next continuous sections'.

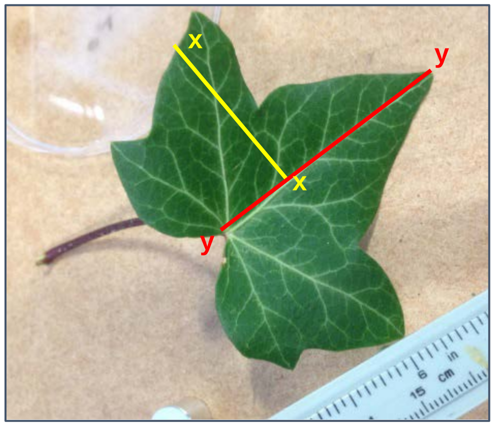


Figure 3-3 An image showing horizontal (x-x) and vertical (y-y) transects of a leaf as considered in this experiment.

Figure 3-4 An image showing sample leaf transects mounted on a large aluminium holder prior to scanning using the ESEM

Subsequently, leaf sections were scanned using the ESEM at 450x magnification. As considerably higher PM levels were observed on the adaxial surfaces of the leaves compared to abaxial surfaces, PM distribution on the adaxial surfaces was considered for the purpose of the pilot study. Micrographs were taken at 100 μm intervals along the lengths of horizontal and vertical transects of the leaves using the ESEM-embedded distance measuring tool. The amount of PM_{10} (all the measurable particles ≤ PM_{10} were considered here) on each micrograph was quantified using ImageJ software following the procedure given in Chapter 2. The smallest measurable particle size was 0.3 μm in diameter using this magnification. The number of PM_{10} accumulated along the horizontal and vertical axes were then plotted against the distance from leaf-edge and leaf tip respectively to identify any distribution pattern. Transects of different leaves varied in lengths both within and between species and hence, lengths were standardised to their percentage values to obtain meaningful graphs to develop a common protocol applicable for different species of plants.

3.4 Results

3.4.1. PM distribution along the horizontal transect of the leaves

The distribution of PM along the horizontal (x-x) axis was uneven and showed a similar pattern in all three species examined (Fig. 3.5). PM levels at leaf-edges (15% to 18% from the edge) and around the midrib (beyond 72% to 75% from the edge) were more uneven compared to the rest. There were elevated levels of PM found around the edges and at most of the points closer to the mid-rib.

3.4.2 PM distribution along the vertical transect of the leaves

PM accumulation along the vertical (y-y) axis of the leaves of all four species examined were also not uniform and of those, three species (*H. helix, Heuchera* 'Paris' *and G. macrorrhizum)* showed a similar pattern of PM distribution (Fig. 3.6). These species showed highly variable PM levels at the leaf-tip (up to 12% to 20% from the tip) and at the leaf-base (beyond 75% to 88% from the tip) with elevated levels at the leaf–tip. The PM distribution on *A. gramineus* was highly variable throughout the axis without any identifiable pattern.

3.5 Discussion

PM accumulation along the both horizontal and vertical axis was uneven in all tested species and hence, sampling of leaf sections should consider this variability. All three species tested for PM distribution along the horizontal axis of the leaf blade showed less variable distribution between 18% from the edge and 28% from the midrib. Increased PM deposition on edges of the leaves and on main leaf veins was also reported by Mitchell *et al.* (2010) and Tomasevic *et al.* (2005). The response of leaves to wind currents by swaying, fluttering or bending (Gillies *et al.*, 2002) creates differential turbulence in the surface boundary layer resulting in varying levels of PM impaction and interception on their surfaces (Davidson and Wu, 1990; Petroff *et al.*, 2009). The elevated PM levels at leaf-edges may be attributed to their high exposure to surrounding wind currents compared to leaf blades (the area between the edge and mid-rib of a leaf) which are only exposed to above or below wind currents. The differences in edge effect between species, ranging from 12% to 18% were probably caused by their variable shapes and sizes (Gemba, 2007).

Considering the PM distribution along the vertical axis, three out of the four species tested showed less variable PM distribution in the body of the leaf blade (between 20% from the tip and 25% from the leaf base). Elevated PM levels at leaf-tips probably result from a larger edge effect that may be created around a narrow shape. This can also be attributed to leaf orientation in vertical greening systems; the leaf-tips of most of the plants point towards the ground, with a slight angle accumulating some roll-off (due to gravity) or washing-off of particles at their tips. In contrast, *A. gramineus* has elongated linear leaves which readily bend-down with wind currents and are oriented forming a slight arch. In addition, due to their narrowed leaf shape, the edge effect can potentially apply all around the leaves without any specific pattern. These reasons probably explain their differential PM distribution compared to the other plants and their not having elevated PM levels at tips or edges.

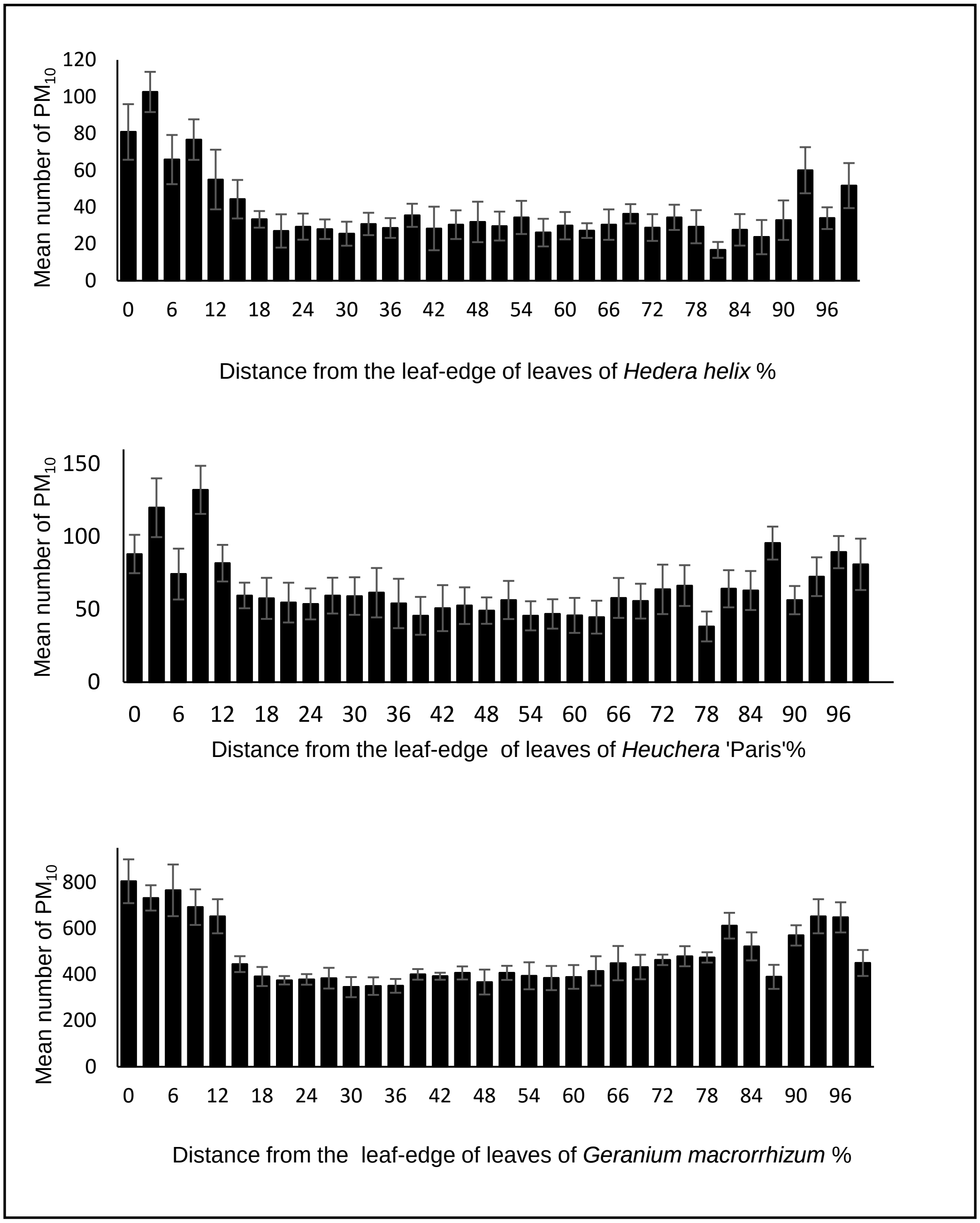


Figure 3-5 Mean number of PM_{10} ± 1SE ($PM_{0.3}$ - PM_{10}) accumulated along the horizontal axis of the leaves (starting from leaf-edge '0%' to midrib (100%))

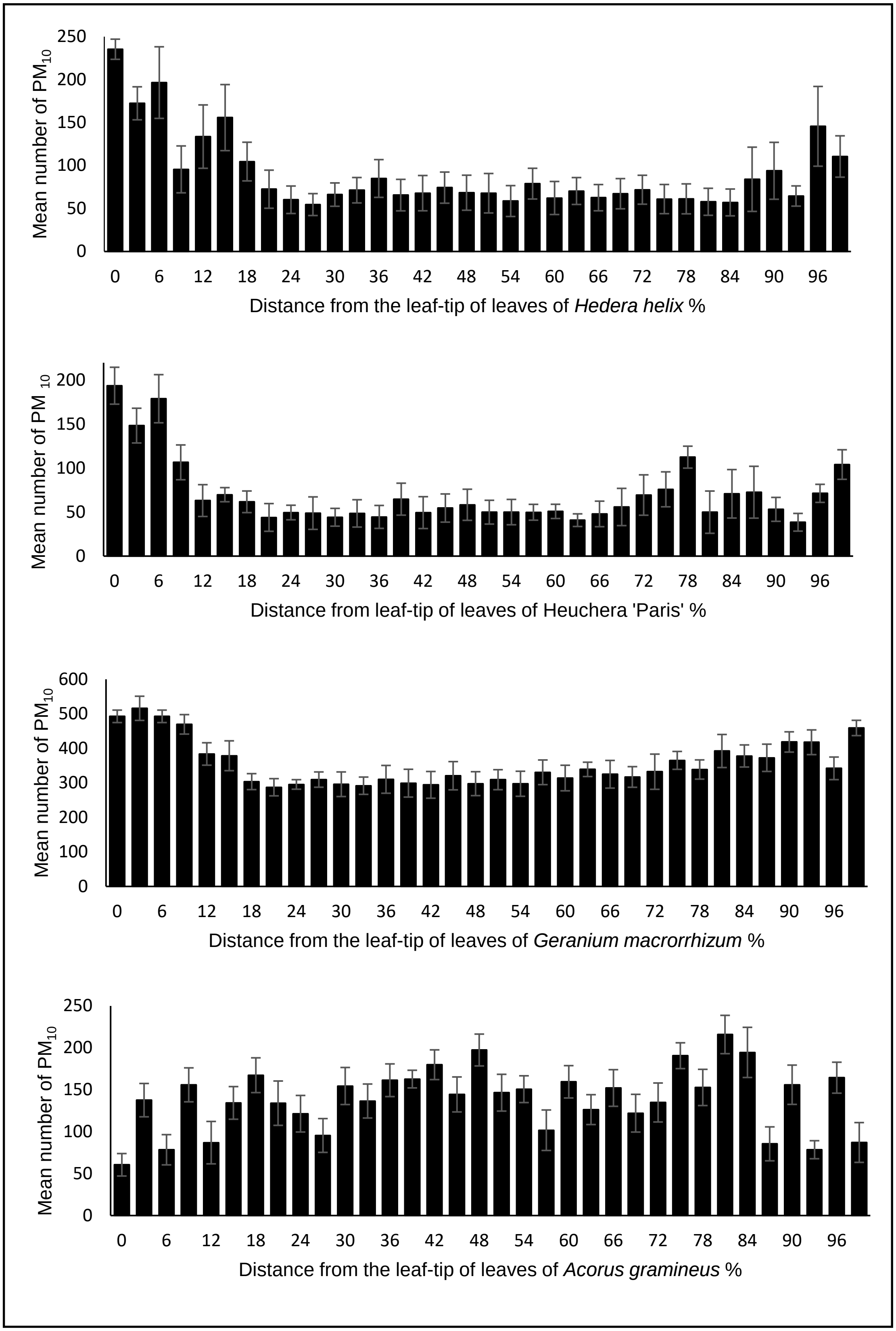


Figure 3-6 Mean number of PM_{10} ± 1SE ($PM_{0.3}$ - PM_{10}) accumulated along the vertical transects of the leaves (starting from leaf-tip to leaf-base)

3.6 The protocol for leaf section sampling

Based on the results of the majority of the species sampled, the area of the leaf blade with less variable PM distribution, avoiding following areas: 18% from the edge, 28% from the midrib, 20% from the tip and 25% from the leaf-base was selected as the most appropriate leaf area for leaf section sampling (shaded area in Fig. 3.7). This selected area for sample leaf sections will hereafter be referred to as the 'leaf blade' (Fig. 3.7).

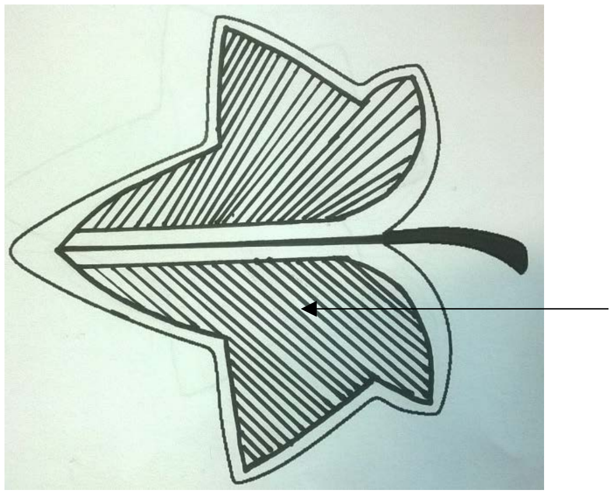


Figure 3-7 schematic diagram of a leaf showing the area selected for leaf section sampling (the shaded area).

This sampling protocol was applied in all the experiments that required leaf sections to be taken, throughout this research. Although linear leaves did not follow any particular pattern of PM distribution, the same approach was followed to avoid any biased selection. However, based on their narrow sizes they were only excised through their horizontal axis, and micrographs were only taken from the leaf-blade, avoiding the edge and midrib area (using the ESEM embedded measuring ruler). Leaves which were too small to cut into sections (less than 2.5 cm^2) could be mounted as whole leaves without cropping, micrographs were taken from the leaf blade following the same protocol using the ESEM ruler.

Chapter 4 : Particulate matter pollution capture by leaves of seventeen living wall species with special reference to rail-traffic at a metropolitan station

This chapter is presented as an article published in the *Journal of Urban Forestry and Urban Greening* (Weerakkody *et al.* 2017) with slight modification to align with the thesis structure.

4.1 Abstract

Atmospheric Particulate Matter (PM) constitutes a considerable fraction of urban air pollution, and urban greening is a potential method of mitigating this pollution. The value of living wall systems has received scant attention in this respect. This study examined the inter-species variation of particulate capture by leaves of seventeen plant species present in a living wall at New Street railway station, Birmingham, UK. The densities of different size fractions of particulate pollutants (PM_1, $PM_{2.5}$ and PM_{10}) on 20 leaves per species were quantified using an Environmental Scanning Electron Microscope (ESEM) and ImageJ image-analysis software. The overall ability of plant leaves to remove PM from air was quantified using PM density and LAI (Leaf Area Index); any inter-species variations were identified using one-way Anova followed by Tukey's pairwise comparison. This study demonstrates a considerable potential for living wall plants to remove particulate pollutants from the atmosphere. PM capture levels on leaves of different plant species were significantly different for all particle size fractions ($P < 0.001$). Smaller-leaved *Buxus sempervirens* L., *Hebe albicans* Cockayne, *Thymus vulgaris* L. and *Hebe x youngii* Metcalf showed significantly higher capture levels for all PM size fractions. PM densities on adaxial surfaces of the leaves were significantly higher compared to abaxial surfaces in the majority of the species studied (t-test, $P < 0.05$). According to EDX (Energy Dispersive X-ray) analysis, a wide spectrum of elements were captured by the leaves of the living wall plants, which were mainly typical railway exhaust particles and soil dust. Smaller leaves, and hairy and waxy leaf surfaces, appear to be leaf traits facilitating removal of PM from the air, and hence a collection of species which share these characters would probably optimize the benefit of living wall systems as atmospheric PM filters.

Keywords: Outdoor air pollution; Urban green infrastructure; Green walls; Railway pollution

4.2 Introduction

Outdoor air pollution caused an estimated 3.7 million premature deaths worldwide in 2012, mainly due to atmospheric Particulate Matter (PM) less than 10 µm in aerodynamic diameter (PM_{10}), ozone, nitrogen dioxide and sulphur dioxide (WHO, 2014). The European Environment Agency estimated that in the 2013-2015 period 50-62% and 82-85% of the urban population in Europe were exposed to levels of PM_{10} and $PM_{2.5}$ (respectively) which exceeded the recommended World Health Organisation (WHO) annual limits (PM_{10}: 20 µg.m^{-3} and $PM_{2.5}$: 10 µg.m^{-3}) (Guerreiro *et al.*, 2017). EEA (2016) also estimated that 467,000 premature deaths in Europe could be attributed to $PM_{2.5}$ (PM less than 2.5 µm in aerodynamic diameter) in 2013. Out of 40,000 annual deaths estimated to be caused by outdoor air pollution in the

UK, 29,000 were caused by PM pollution (Royal College of Physicians, 2016). Long-term exposure to airborne PM is directly associated with potentially fatal childhood diseases including post-neonatal infant mortality (Laden *et al.*, 2006), Sudden Infant Death Syndrome (SIDS) (Woodruff *et al.*, 2006) and various other diseases which affect all segments of the community such as cardiopulmonary diseases, lung cancer (Pope III *et al.*, 2011) atherosclerosis (Araujo, 2011) and asthma (Anderson *et al.*, 2013). Ultrafine particles ($PM_{0.1}$), PM less than 0.1 µm in aerodynamic diameter can cause serious damage by entering the liver, spleen, kidney and the brain (via the olfactory nerves) (Solomon *et al.,* 2012). They can also reach the lower respiratory system and change alveolar macrophage functions due to toxic chemicals carried by the particles (e.g. polycyclic aromatic hydrocarbons (PAHs) and heavy metals) (Riddle *et al.*, 2009). On entering the human bloodstream, they can create systemic inflammatory changes, which can lead to serious complications in blood coagulability (Seaton *et al.* 1995). The annual cost to society due to particulate pollution in the UK has been estimated at £16 billion (RCP, 2010).

Coarse particles can originate from natural sources and anthropogenic activities, while fine particles mainly originate from vehicle emissions (gasoline and diesel), combustion, and industrial processes (Chow *et al.*, 2006). Ultra-fine particles mostly originate from transport and photochemical reactions in the atmosphere (Chow *et al.*, 2006). These particles contain toxic compounds such as heavy metals, PAHs, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs), making them more hazardous and carcinogenic (Dzierzanowski *et al.,* 2011). The International Agency for Research on Cancer (IARC) classified diesel exhaust as a Group 1 (carcinogenic to humans) carcinogen (Silverman *et al.*, 2012). The railway network is one of the main sources of air pollution in the UK due to diesel and electric train emissions (Thornes *et al.*, 2016). In addition to particles generated via rail traffic exhaust, particles can also be generated due to wheel friction, friction with overhead cables and when applying brakes; the particles generated in these circumstances fall mainly within the ultrafine range (Thornes *et al.*, 2016).

Particulate levels in many large cities in the UK exceed both the WHO guidelines and EU safe limits; air pollution mitigation approaches such as emission reduction, enhancing atmospheric dispersion and building high emission sources away from currently polluted or highly populated areas (Pugh *et al.*, 2012) are unlikely to have any impact on city PM levels generated from transport. Increasing surface deposition has been identified as an effective short-term strategy to reduce atmospheric particulate pollutants (Pugh *et al.*, 2012), especially those locally produced within cities due to transportation systems. Since vegetation can act as a sink for particulates (Beckett *et al.*, 2000b; Fowler *et al.*, 2004; Freer-Smith *et al.*, 2004) it has the potential to have a high impact in this respect. Trees are often the main source of greening considered in urban landscapes, however, there are several limitations and barriers to achieving urban greening purely by using trees, including (but not limited to): prevailing soil conditions, space utilisation, sub-surface infrastructure, availability of sunlight and the size of the trees compared to the adjacent buildings (Johnston and Newton, 2004). Green walls (vertical greening) could overcome most of these limitations by transforming building walls to greenery while minimizing land-take and providing additional benefits including thermal insulation, noise reduction and conservation of urban biodiversity and rewilding of cityscapes (Alexandri *et al.*, 2007; Chiquet *et al.*, 2013; Dover, 2015; Jepson, 2016; Johnston and Newton, 2004). Previous studies on green walls have focused more on the value of climbing plants, such as ivy, in reducing PM pollution and little information is available on the value of living walls in this respect (Cheetham *et al.*, 2012) though see Perini *et al.* (2017) and Shackleton *et al.* (undated). Living walls are vertically growing hydroponic green wall systems which facilitate the growth of a variety of plant species with a potential for greater artistic expression than simply using climbing species (Dover, 2015). PM filtering behavior of living wall systems with reference to different PM size fractions of particulates and the optimal species composition for living wall systems to act as effective particulate matter traps are not well understood. This study explores the role of living wall systems in the reduction of PM pollution; in contrast to the work of Perini *et al.* (2017), particulate capture is investigated at street level adjacent to a pedestrian walkway.

In early research different technical approaches were taken to quantify particulate capture by vegetation, including comparison of dust-fall measurements between the canopy area and open space (Dochinger, 1980), atmospheric aerosol screening (Bache, 1979; Wiman, 1985) and deposition velocity models (Bache, 1979). The gravimetric method, which collects particulate matter in water by washing material off the leaves followed by filtering and weighing the residue has frequently been used (Beckett *et al.*, 2000b; Freer-Smith *et al.*, 2005; Ram *et al.*, 2012). However, there are several drawbacks to the latter technique: particulates held on the epicuticular wax or microstructures of leaves may not be washed-off and hence may not be weighed. In some research chloroform was used as the solvent to dissolve epicuticular wax and collect the PM trapped in the wax component (Dzierzanowski *et al.*, 2011; Sæbø *et al.*, 2012 and Song *et al.,* 2015). However, as chloroform is used as a solvent to dissolve non-polar molecules and the soluble fraction of PAHs (Castelli *et al.*, 2002) eluting with such solvents has the potential to dissolve some particulates comprised of non-polar materials. In addition, according to pulmonary toxicity studies, the particle surface area and particulate count are more appropriate measures for smaller particles than particle mass (Sager and Castranova, 2009). Ottele *et al.* (2010) quantified the number of particulates captured by leaves of *Hedera helix* by using a Scanning Electron Microscope (SEM) to image the particulates in situ and used an image analysis program to count and size-range the particles deposited on the leaves. However, SEM scanning areas are much smaller compared to leaf surface area; and hence, a representative number of micrographs should be taken to draw any conclusions on PM levels on leaves using this approach.

Removal of atmospheric particulates by vegetation is mainly driven by the interactions between the particles and plant surfaces including their morphological properties such as shape, size and orientation (Petroff *et al.*, 2008a). Particulate deposition on plants is thought to be influenced by particle diameter and the micro-roughness or micro-topography of the plant (Slinn, 1982). However, there is much debate on the impact of leaf size and morphology on particulate capture. Therefore, this study examined inter-species variation in PM removal by living wall species in order to understand the best species combinations to capture PM employing a SEM/image analysis approach. Particulate densities (the number of particulates deposited per unit area of leaf surface) on the adaxial (upper) and abaxial (under) surfaces of the leaves were also examined to understand any variation due to leaf size or morphology. Further to this, the elemental composition of the captured particulates was also studied to detect the elements which can be removed using living wall plants.

4.3 Material and methods

4.3.1 Site selection

Birmingham is a large city located in the West Midlands of England (Fig. 4.1) with a population of over 1.1 million (Birmingham City Council 2014). In Birmingham, PM accounted for 6.4% of the premature mortality rate in 2009 (Gowers *et al.*, 2014). Birmingham New Street railway station is one of the busiest railway stations in the UK, with up to 140,000 commuters and staff passing through daily (Thornes *et al.*, 2016). Approximately 1,000 trains/day (comprising equal numbers of diesel and electric powered trains) pass through this station (Thornes, 2016) and $PM_{2.5}$ levels of up to 58 $\mu g.m^{-3}$ for hourly intervals have been reported within the station (Zulkifli, 2015 cited in Thornes *et al.*, 2016). Given the amount of pollution generated in and around the station, a free-standing modular living wall located on it's north side, 5 m above the railway (which is sunk below street level) and 3.2 m from the closest platform (Fig. 4.1) ($52^0 28'41.2''$ N $10^0 53'48.7''$ W) was selected as the experimental site. The living wall was manufactured by ANS Global in 2012 and was subsequently managed by Network Rail; the structure is 77 m long and varies in height from 4.5 m near the station to 3.5 m at its furthest point (300 m^2 in total) and hosted twenty different species of plants (Table 4.1). A low (0.5 m) stone-clad planter with low-growing shrubs was present at the base of the wall (Fig. 4. 2). A mean wind speed of 4.1 m/s, a mean temperature of 15 ^{0}C, a mean daily rainfall of 101.2 mm and a mean humidity of 64% were reported from the study area during the period of sampling (Met Office - GOV.UK).

4.3.2 Sampling

Twenty leaves per species (n =20) of sixteen healthy plant species present in the living wall (Table 4.1) were randomly sampled (avoiding damaged leaves) at 2.0-2.5 m height above the footpath. In addition, 40 leaves of *Thymus vulgaris* L. were sampled (see section 2.3 for more details) at the same height (360 leaves in total). *Fragaria vesca* L. was only located in the uppermost rows of the living wall (was inaccessible), *Galanthus nivalis* L. and *Lysimachia nummularia* L. were not healthy within the sampling period; and hence these three species were not included in the study. Sampling dates and times were selected based on weather conditions and carried out on six occasions with similar weather conditions (Table 4. 2) between April and July 2016. Sampling was carried out only during dry weather conditions (having at least four consecutive non-rainy days immediately before sampling) and all the species were equally sampled on each sampling day (at least 3 leaves per species) to avoid differential influence from daily weather differences (Table 4. 2). Leaves were hand-picked and stored in plastic containers in such a way that they did not rub against one another or against the container (to avoid disturbing the particles) and sealed (Chapter 2).

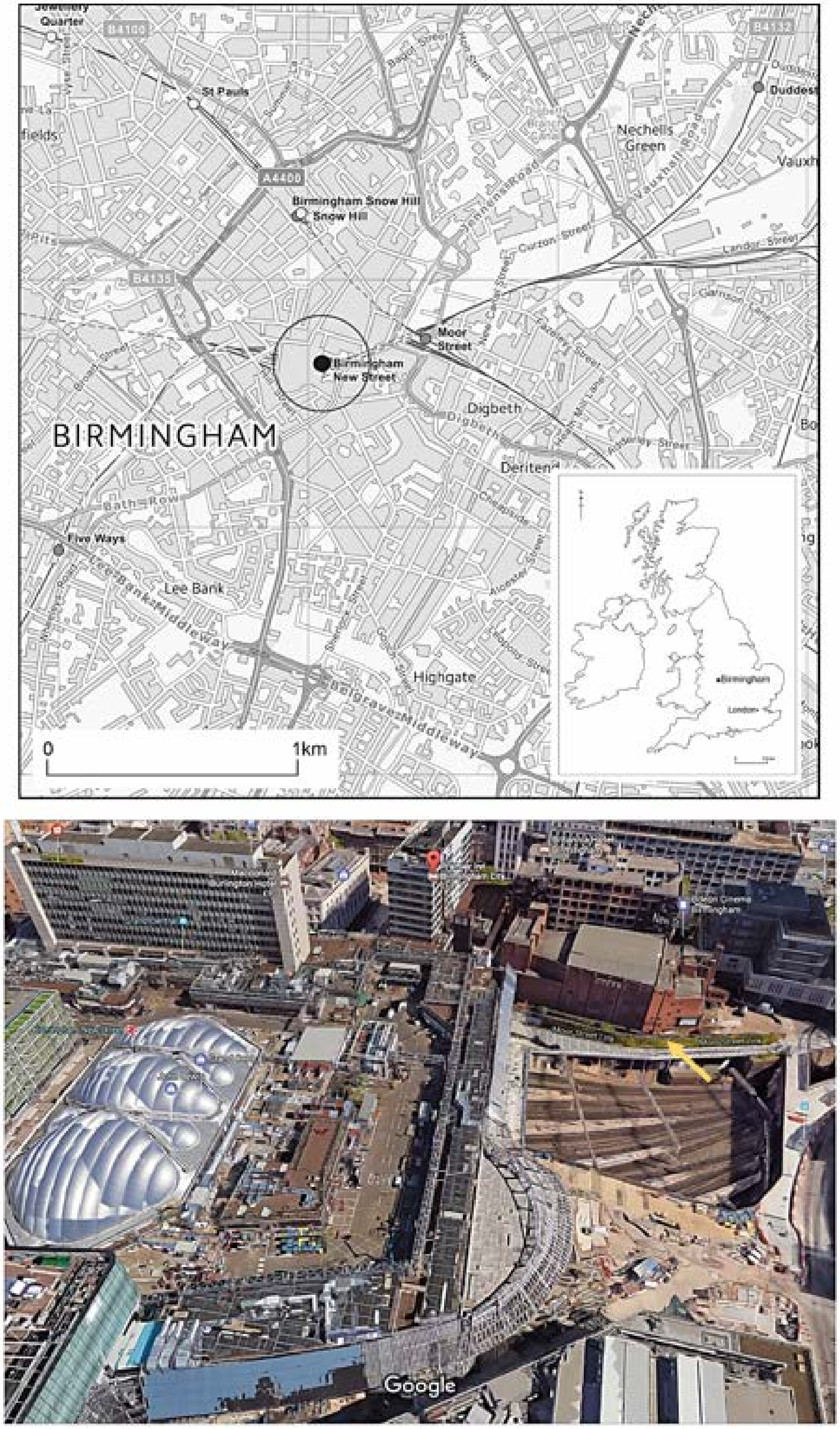


Figure 4-1 Map showing the location of New Street station in Birmingham, UK (upper image) *Contains OS data © Crown copyright and database right (2017).* The living wall located by the footpath adjacent to Birmingham New Street Train Station is marked by the arrow in the bottom image (Google Maps, 2017)

Figure 4-2 An image of a section of the living wall system located adjacent to the New Street station in Birmingham, UK in 2016.

4.3.3. Quantifying the PM densities on leaf surfaces

Samples were carefully stored in a refrigerator (approximately 9 ^{0}C) in the same storage boxes to prevent any dehydration and structural changes until sample preparation and analysis within two days of sampling. A pilot study (Chapter 3) conducted using common living wall plant species with a representative sample of different morphologies showed less variable particulate distributions on the leaf-blade compared to other leaf areas (tip, edges, base and mid rib) and the leaf-blade was thus selected as the most appropriate area to sample leaf sections for all the species. Samples were prepared for microscopic examination by removing six leaf sections 5.0 x 5.0 mm in size, from every leaf blade of all the species apart from small-leaved species (less than 250 mm^2). Three sections were used to examine the adaxial surface and three for the abaxial surface for each leaf. Leaf sections were mounted on aluminium stubs using double-sided carbon sticky tabs (Chapter 2). Environmental Scanning Electron Microscope (ESEM) (Model: JSM-6610LV) micrographs were taken at three random points per leaf section (providing nine micrographs per each side of every leaf) using Back Scattered Electrons (BSE) under a low vacuum (LV mode) at 450X and 1,000X (resulting micrographs were 60,681.5 μm^2 and 12,288.0 μm^2 in size) without any conductive coating.

Leaves can be imaged in the ESEM without any conductive coating due to their natural carbon content and their cuticle minimising dehydration (Ensikat *et al.*, 2010). Small-leaved species (less than 250 mm^2), which were too small to physically cut into sections were cut into halves and mounted without cropping, to scan adaxial and abaxial surfaces (using each half); nine micrographs were taken from the leaf blade of each half of the leaf adhering to the same protocol. However, leaves of *T. vulgaris* were too small to section into halves and, hence, adjacent leaves were used to image the abaxial surfaces using forty leaves in total. Micrographs were taken using the same working distance, while maintaining contrast and brightness levels as consistent as possible to avoid any difficulties in defining the threshold of the image analysis process.

Table 4.1 LAI ±SE and leaf characteristics of different species of plants present in the living wall at New Street Station, Birmingham, UK in 2016

Plant species	English name	LAI ±SE	Mean leaf size±SE (cm^2)	Description of the leaf characteristics and any specific micro-morphological features*
Armeria maritima L.	thrift	3.18 ±0.02	1.48± 0.06	Small, linear leaves forming tufts. Ridges, groves and a few sparsely arranged glandular hairs were present on both surfaces.
Blechnum spicant (L.) Sm.	hard-fern	0.59 ±0.04	6.67± 0.30	Medium, pinnate, leathery fronds. The adaxial surface was smooth with few folds and abaxial surfaces were smooth or some consist of sori**.
Buxus sempervirens L.	common box	1.66 ±0.03	0.9± 0.04	Small, oval, leathery leaves Smooth leaf surfaces with very few trichomes. The abaxial surface was slightly folded due to embossed stomata.
Euphorbia amygdaloides L.	purpurea	1.37 ±0.07	2.86±0.39	Medium, oblong, leaves forming rosettes. Leaf surfaces were leathery. Less prominent epicuticular wax plates were observed on the adaxial surface.
Galium odoratum (L.) Scop.	sweet woodruff	0.88 ±0.01	1.04±0.04	Small, lanceolate, glabrous leaves forming whorls. Leaf surfaces were smooth.
Geranium macrorrhizum L.	cranesbill 'Bevan's Variety'	2.85 ±0.09	31.69±2.24	Large, palmately lobed, broad leaves. Both leaf surfaces had densely arranged hair and glandular trichomes.
Hebe albicans Cockayne	white hebe	2.17 ±0.08	1.41±0.08	Small, oval leaves forming a broad mound. Epicuticular wax plates were slightly prominent on the adaxial surface.
Hebe salicifolia (G. Forst.) Pennell	koromiko	1.57 ±0.09	2.31±0.21	Small, narrowly lanceolate willow-like leaves. Leaf surfaces with few ridges, groves and few glands. Epicuticular wax was localized around the glands and less prominent.
Hebe x *youngii* Metcalf. (*Veronica elliptica* x *pimeleoides* Carl Teschner)	hebe youngii	2.85 ±0.05	0.74±0.03	Small, ovate leathery leaves. Glandular trichomes, ridges and groves were prominent on both leaf surfaces. Epiculticular wax plates were slightly prominent on the adaxial surface
Hedera helix L.	ivy (gold child)	2.69 ±0.07	9.64±0.65	Medium, palmately lobed leaves. The adaxial surface was covered with thick epicuticular wax layers.
Helleborus x *sternii* Turrill	blackthorn strain	0.72 ±0.01	17.9±0.38	Large, broad leaves with a serrate margin and a leathery surface. Localised deep ridges, groves and slightly prominent epicuticular wax layers were present on the adaxial surface.

Hyssopus officinalis L.	hyssop	0.93 ±0.02	0.46±0.03	Small, lanceolate leaves with a soft (gentle) texture. Leaf surfaces with few glandular hairs and localised, less prominent, epicuticular wax.
Luzula nivea (L.) DC.	snow rush	1.35 ±0.03	4.87±0.22	Medium, linear leaves ("grass-like") with hairy margins. Parallel ridges present in both leaf surfaces. Very few hairs were observed on the leaf blades.
Pachysandra terminalis Siebold & Zucc.	Japanese spurge	1.82 ±0.08	8.28±0.51	Medium, dentate, glabrous leaves. Smooth, leaf surfaces with very few wax glands.
Phyllitis scolopendrium L.	hart's tongue fern	1.01 ±0.07	18.74±0.87	Large, broad, leathery fronds forming a rosette. The adaxial surface had deep ridges and groves and abaxial surfaces were smooth or some consist of sori**.
Primula veris L.	common cowslip	1.57 ±0.04	16.56±0.62	Large, broad, obovate leaves forming a rosette. Leaf surfaces with sparsely arranged hair, ridges and groves.
Thymus vulgaris L.	common thyme	1.98 ±0.01	0.05±0.00	Small, ovate leaves forming whorls. Both leaf surfaces were waxy and had a complex microstructure with densely arranged short stubby trichomes, and glandular hairs.
Fragaria vesca L.	wild strawberry	not included in the study		
Galanthus nivalis L	common snowdrop	not included in the study		
Lysimachia nummularia L	golden creeping Jenny	not included in the study		

*Leaf shapes as in Hickey and King (2000) **Sori are groups of sporangia found in ferns (Bowler, 1899)

Table 4.2 Weather conditions at the study site during the time of sampling

Date	Time	Temperature	Wind speed	Humidity	Precipitation
19th April 2016	11.30 - 13.00	14 ^{0}C	3.04 ms^{-1}	51%	0
6th May 2016	11.30 - 13.00	18 ^{0}C	2.77 ms^{-1}	49%	0
14th May	11.30 - 13.00	15 ^{0}C	2.68 ms^{-1}	54%	0
9th June 2016	11.30 - 13.00	19 ^{0}C	2.64 ms^{-1}	56%	0
18th June 2016	11.30 - 13.00	15 ^{0}C	2.68 ms^{-1}	72%	0
4th July 2016	11.30 - 13.00	19 ^{0}C	3.13 ms^{-1}	56%	0

The amount of PM_{10} (excluding $PM_{2.5}$ and below), $PM_{2.5}$ (excluding PM_1 and below) and PM_1 (all the measurable PM less than 1 μm aerodynamic diameter) on each micrograph was quantified using ImageJ image analysis software (Collins, 2007; Ottele *et al.*, 2010; Sternberg *et al.,* 2010) and its auto threshold tool was used to minimise human error (Chapter 2). The smallest particle size that could be accurately counted (with enough resolution and less conductive charging) using this technique was 0.1 μm in diameter. Particles between 0.1 μm and 1 μm have similar aerodynamic behaviour resulting in similar deposition velocities (Slinn, 1982). Because smaller particles are linked to more severe health effects, PM_1 was quantified as a separate fraction in addition to the more commonly reported PM_{10} and $PM_{2.5}$ fractions. The most appropriate threshold was chosen for the image analysis process using 10 micrographs with reference to their respective secondary electron images (at high resolution) to ensure only the particles were filtered and the leaf surfaces were subtracted (Chapter 2). The mean PM density (per 1 mm^2) on each side of each leaf was calculated taking the mean of the PM density on each micrograph. The overall PM density on leaves per 1 mm^2 was calculated by combining the PM densities on both the adaxial and abaxial surfaces. The mean PM density on leaves of each species was calculated using the average PM density on each of the 20 leaves/species. A total of 360 random micrographs per species were used to estimate the mean PM densities on each species.

4.3.4 Leaf Area Index

Leaves of different plants have different surface areas and are distributed differently in space. The total particulate capture on leaves of different species may vary depending on the available leaf surface area to capture particles. As living walls are vertical, the LAI was measured relative to the unit vertical area of the living wall. The number of leaves distributed on a unit vertical area was calculated using a 100 mm x 100 mm quadrat (Chapter 2) for all seventeen species; the average number of leaves present within a quadrat was calculated using three random quadrats per species. The surface area of individual leaves was measured using ImageJ, the mean leaf surface area of leaves of each species was calculated using ten random leaves per each species. The LAI of each species was calculated using the following formula:

LAI = La x Nl / Qa

La = Mean surface area of an individual leaf, Nl = Average number of leaves per quadrat, Qa = Total area of the quadrat

4.3.5 Quantifying the overall PM removal capacity including LAI by different species of plants

The ability of each species of plant to remove PM using a 100 cm^2 area of living wall was calculated using the mean PM density on each species and the LAI:

PM removal by 100 cm^2 area of each species = Number of PM on 100 cm^2 area of the leaves x LAI = Mean PM density on the leaves x 10^4 x LAI

4.3.6 Observation of leaf characteristics

The surface morphology of leaves was examined using the ESEM at a range of magnifications as appropriate (100X, 250X, 350X, 450X and 900X) to understand any variability in PM density associated with specific leaf surface characteristics.

4.3.7 Statistical analyses

R statistical software version 3.2.5 (R Development Core Team, 2016) was used for all statistical tests in this study. Any significant variations in PM density on leaves of different plant species and any significant variations in PM removal ability (including LAI) of different plant species with reference to different particle size fractions were analysed using one-way ANOVA following confirmation of normality using the Shapiro-Wilk test. Significant differences in pairwise comparisons of species were identified and clustered using Tukey's HSD post-hoc test (package: Agricolae). As the adaxial and abaxial surfaces of the leaves of the same species may have different micro-morphology, they were separately analysed to explain any variation between plant species due to the differences in leaf properties (size, shape and texture). Any significant differences in PM density between adaxial and abaxial surfaces of the same species were identified using a Student's t-test.

4.3.8 Elemental analysis of particulates

Elemental composition of particulates captured on leaves was determined using Energy Dispersive X-ray analysis (EDX) using the INCA software coupled with the ESEM (Williamson *et al.*, 2004). Ten leaf sections per species (randomly selected, representing all sampling dates) were scanned using the SEM at 1,000X using an accelerating voltage of 15 kV and back scattered electrons. The scanning images were acquired in INCA software and the elemental composition of the particles was analysed using the Point and ID analyser. The Point and ID analyser works as both a qualitative and quantitative analytical tool to identify and quantify the elements in particles as their percentage weight (Wt%) (INCA energy operator manual, 2006). The mean quantity of each identified element in each species was calculated (mean Wt%) by taking the mean of ten random particles scanned for each species of plant.

4.4 Results

4.4.1 Overall PM capture and inter-species variation of PM removal by plants

4.4.1.1 PM density on leaves

Analysis of ESEM micrographs revealed differential PM densities on leaves of different plant species at all particulate size ranges (PM_1: $F = 39.97$, $p < 0.001$; $PM_{2.5}$: $F = 55.83$, $p < 0.001$; PM_{10}: $F = 44.08$, $p < 0.001$) (Table 4.3). The highest mean densities of PM_1 and $PM_{2.5}$ (45,000 ± 3,300 mm^{-2} and 16,500 ± 900 mm^{-2} respectively) were found on leaves of *B. sempervirens* and they were significantly higher than all the other species ($p < 0.05$) apart from *H. albicans* and *T. vulgaris* (Table 4.3). *T. vulgaris* had the highest mean density of PM_{10} on its leaves (4,040 ± 200 mm^{-2}; Table 4.3) which was significantly higher ($p < 0.05$) than most of the species apart from *B. sempervirens* and *H. albicans*. The lowest densities of PM_1, $PM_{2.5}$ and PM_{10} were found on leaves of *L. nivea*, *B. spicant* and *P. scolopendrium* respectively (Table 4.3).

Table 4.3 Mean PM density (±SE) per 1 mm^2 of a leaf (data for adaxial and abaxial surfaces are combined) of different species of plants on the living wall at New Street Station, Birmingham, UK in 2016

	Mean PM density ± SE (mm^{-2}) and Tukey's groups of significance					
Species	**PM_1 ± SE x 10^3**	**Group**	**$PM_{2.5}$ ± SE x 10^3**	**Group**	**PM_{10} ± SE x 10^3**	**Group**
B. sempervirens	45.03 ± 3.3	a	16.46 ±0.9	a	3.04 ±0.2	ab
H. albicans	40.41 ±6.2	a	13.01 ±1.4	ab	2.77 ±0.2	abc
T. vulgaris	27.86 ±2.6	a	11.41 ±0.7	abc	4.04 ±0.2	a
H. salicifolia	14.76 ±2.4	b	8.32 ±1.1	bcd	1.87 ±0.2	cd
G. macrorrhizum	12.28 ±2.2	bc	2.79 ±0.3	efgh	0.90 ±0.1	efg
H. youngii	11.65 ±0.8	bc	7.26 ±0.5	cd	2.53 ±0.2	bc
H. helix	9.99 ±1.4	bcd	4.90 ±0.5	de	1.45 ±0.2	de
E. amygdaloides	9.88 ±1.5	bcd	4.23 ±0.6	ef	1.68 ±0.1	efg
A. maritima	9.24 ±1.1	bcd	3.51 ±0.3	efg	1.28 ±0.1	de
P. terminalis	8.37 ±1.0	bcd	2.51 ±0.2	fghi	0.83 ±0.1	efg
G. odoratum	6.87 ±1.3	cde	2.79 ±0.4	efgh	1.09 ±0.1	def
P. veris	6.58 ±0.8	cde	2.17 ±0.2	ghi	0.72 ±0.07	efg
H. officinalis	5.82 ±0.8	def	2.69 ±0.3	fgh	0.90 ±0.09	efg
H. sternii	4.35 ±0.7	efg	2.49 ±0.4	fghi	0.68 ±0.1	fg
B. spicant	3.08 ±0.3	efg	0.74 ±0.06	j	0.26 ±0.02	hi
P. scolopendrium	3.01 ±0.5	fg	1.47 ±0.1	ij	0.21 ±0.03	i
L. nivea	2.78 ±0.4	g	1.74 ±0.3	hi	0.54 ±0.09	gh

4.4.1. 2 PM capture incorporating LAI measures

The PM numbers given in this section incorporate the LAI; as this generates exceptionally large numbers we have given the data in terms of millions of particles, hence they should be multiplied by 10^6. On average 250 ±17 of PM_1, 99 ± 5 of $PM_{2.5}$ and 27 ± 1 of PM_{10} were estimated to have been removed by a 100 cm^2 of the living wall (assuming an equal area of each plant species). There was a significant variation in the ability of different species of plants to remove PM in all size fractions (PM_1: $F = 77.1$, $p <0.001$; $PM_{2.5}$: $F = 122.9$, $p <0.001$; PM_{10}: $F = 88.73$, $p <0.001$) (Fig. 4.3) these variations can be attributed to the varied PM capture rate per unit leaf area (Table 4.3) and their LAI values (Table 4.1). PM_1 removal by *H. albicans* (876 ± 130) was significantly higher than for all the other species ($p <0.05$) apart from *B. sempervirens* ($p =1.00$) and *T. vulgaris* ($p =0.81$) (Fig. 4.3a), and removal of $PM_{2.5}$ (282 ± 30) was also significantly higher than for most other species ($p <0.05$) apart from *B. sempervirens* ($p =1.00$), *T. vulgaris* ($p =0.85$) and *H. youngii* ($p =0.26$) (Fig. 4.3b). The best performing species in PM_{10} capture (79.9 ± 5.2) was *T. vulgaris* (Fig. 4.3c) and this was significantly higher than for most of the species ($p <0.05$) apart from *H. youngii* ($p =0.99$), *H. albicans* ($p =0.78$) and *B. sempervirens* ($p =0.13$). If higher PM-capturing species are arranged in descending order (only those species in Tukey's HSD post-hoc test groups with higher PM levels are considered i.e. – those given a, b and c in Fig. 4.3):

PM_1: *H. albicans > B. sempervirens > T. vulgaris> G. macrorrhizum > H. youngii*;
$PM_{2.5}$: *H. albicans > B. sempervirens > T. vulgaris> H. youngii> H. helix> H. salicifolia> A. maritima;*
PM_{10}: *T. vulgaris> H. youngii> H. albicans > B. sempervirens> A. maritima> H. helix> H. salicifolia*.
Considering all three particle size fractions, *B. sempervirens*, *H. albicans, T. vulgaris and H. youngii* were the species with highest PM removal capacity.

The worst performing species in PM capture, in all particle size fractions, was *B. spicant* (PM_1: 18.2 ± 2.1, $PM_{2.5}$: 4.41± 0.35 and PM_{10}: 1.33 ± 0.16) (Fig. 4.3). PM_1 capture by *P. scolopendrium* and *L. nivea* was significantly lower than most of the other species but not significantly different from the lowest (*B. spicant*). $PM_{2.5}$ captured by *B. spicant* was significantly lower than all the other species of plants and PM_{10} captured by *B. spicant* was significantly lower than all the other species apart from *P. scolopendrium*.

a)

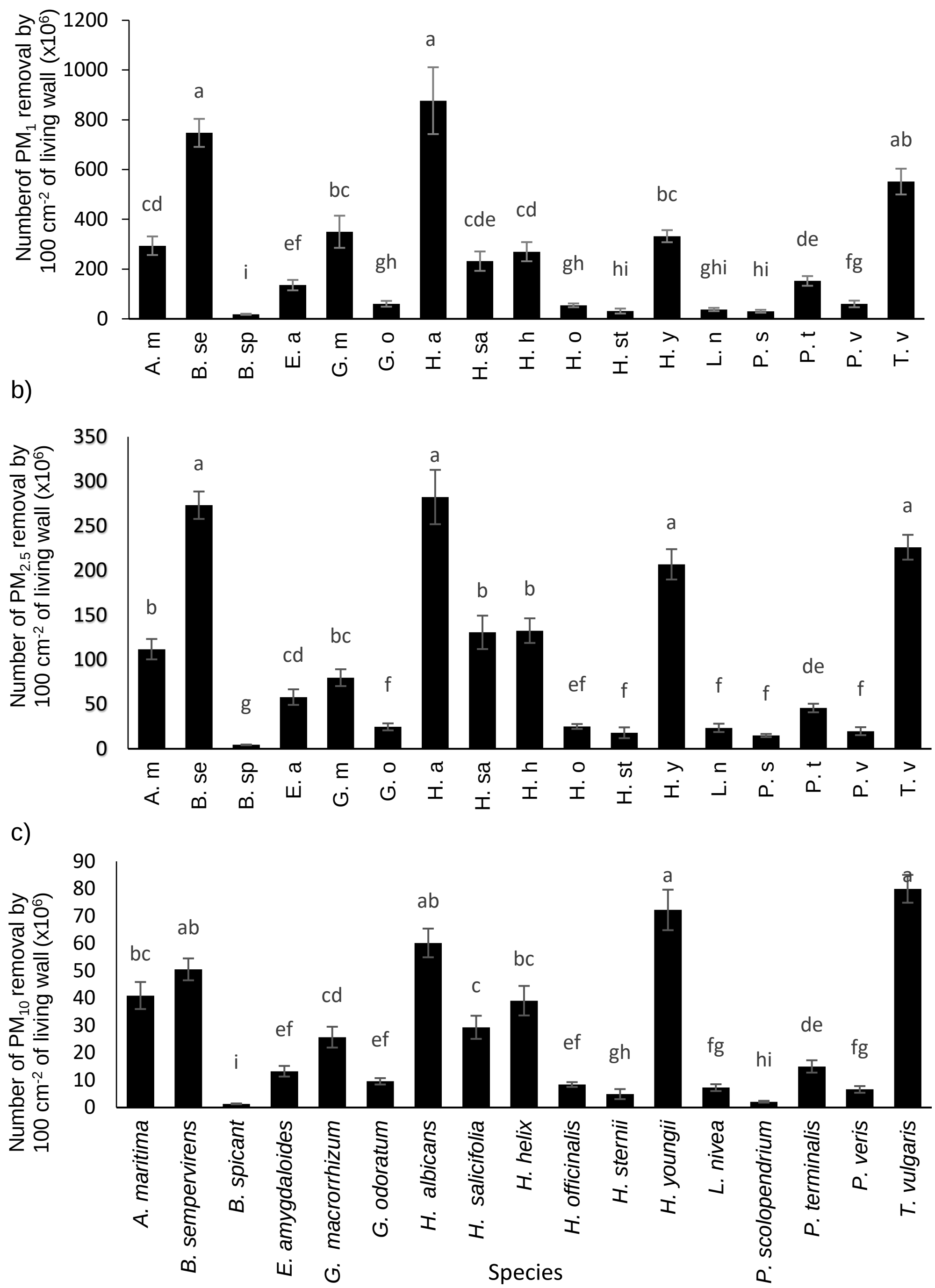


Figure 4-3 Estimated mean ±1SE ($x10^6$) PM removal by leaves of different species of plants on a living wall at New Street Station, Birmingham, UK in 2016; taking into account the leaf area index. a) PM_1, b) $PM_{2.5}$, c) PM_{10}. (species sharing the same letter are not significantly different using Tukey's HSD post hoc test with 95% confidence level, p>0.05). Note the different values on the Y-axis.

4.4.2 PM density on adaxial and abaxial surfaces of the leaves

The comparison of PM densities on adaxial and abaxial surfaces of leaves (Fig. 4.4) showed that PM density on adaxial surfaces were almost always higher than on the abaxial surfaces; the exceptions to this were PM densities on *E. amygdaloides* and PM_{10} densities on *H. albicans* which were a little higher on the abaxial surfaces, though not significantly so ($p > 0.05$). There were significant variations in PM densities (excluding LAI) on both leaf surfaces, among different species of plants (Table 4.4). The highest mean densities of PM_1 and $PM_{2.5}$ (34,633 mm^{-2} and 12,839 mm^{-2} respectively) on the adaxial surfaces of leaves were found in *B. sempervirens* and the highest average density of PM_{10} was found on leaves of *T. vulgaris* (2,991 mm^{-2}). On the abaxial surfaces of the leaves, *H. albicans* showed the highest average PM density in all particle size fractions (PM_1: 18,464 mm^{-2}, $PM_{2.5}$: 6,290 mm^{-2} and PM_{10}: 1,547 mm^{-2}). Similar to the results for total PM removal ability (Fig. 4.3), *B. spicant, P. scolopendrium, L. nivea, H. sternii* and *P. veris* showed relatively low PM densities on both adaxial and abaxial surfaces in all PM size fractions (Fig. 4.4), these levels were significantly lower than most of the other species (Table 4.4).

Table 4.4 Variations in PM density on adaxial and abaxial surfaces of leaves of different species of plants on a living wall at New Street Station, Birmingham, UK in 2016

	Adaxial surface			Abaxial surface		
Species	**PM_1**	**$PM_{2.5}$**	**PM_{10}**	**PM_1**	**$PM_{2.5}$**	**PM_{10}**
	$F = 37.5$, $p < 0.001$	$F = 34.6$, $p < 0.001$	$F = 33.8$, $p < 0.001$	$F = 25.5$, $p < 0.001$	$F = 64.82$, $p < 0.001$	$F = 29.7$, $p < 0.001$
A. maritima	cde	efg	cde	cde	cd	bcd
B. sempervirens	a	a	a	ab	ab	bc
B. spicant	fg	i	g	g	h	f
E. amygdaloides	cdef	efg	efg	bcde	bc	bcd
G. macrorrhizum	cd	efg	cde	def	ef	e
G. odoratum	def	efgh	cdef	efg	de	cde
H. salicifolia	c	bcd	cd	bcd	b	ab
H. albicans	b	b	bc	a	a	a
H. helix	cde	cd	bcd	efg	de	de
H. officinalis	ef	efgh	cdef	fg	def	de
H. sternii	fg	fghi	fg	g	de	de
H. youngii	cd	bc	ab	cde	bc	bc
L. nivea	g	hi	fg	g	ef	e
P. scolopendrium	g	ghi	g	g	gh	f
P. terminalis	cde	def	cdef	fg	h	de
P. veris	efg	fgh	def	defg	fg	e
T. vulgaris	b	ab	a	bc	b	Ab

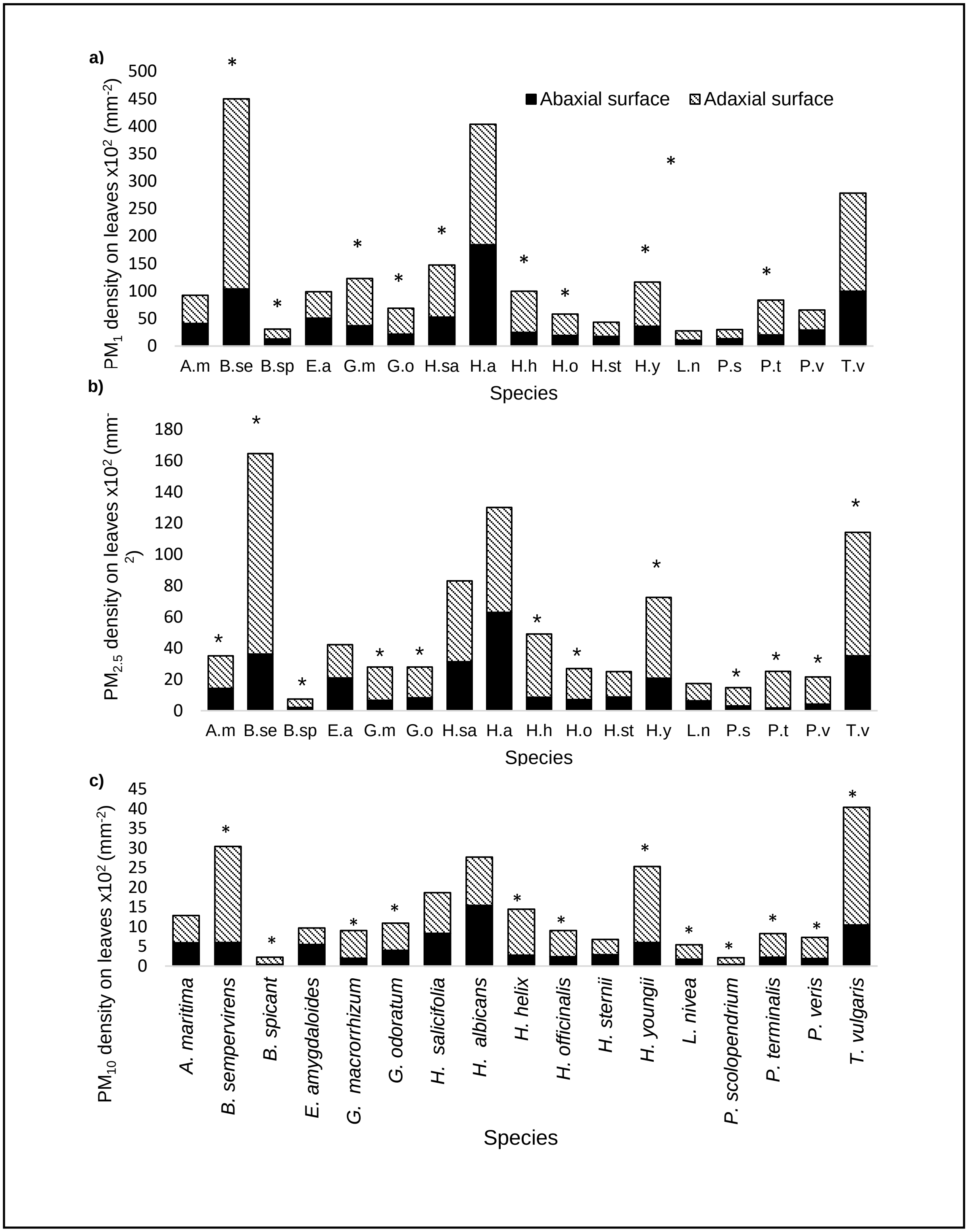


Figure 4-4 Mean PM densities ±1SE (x10^2) on adaxial and abaxial surfaces of the leaves of different species of plants on a living wall at New Street Station, Birmingham, UK in 2016. a) PM_1, b) $PM_{2.5}$, c) PM_{10}.
* PM densities between adaxial and abaxial surfaces are significantly different ($p < 0.05$). Error bars not shown for clarity; contractions of species names given on the x-axis of panel c) are used in panels a) and b).

4.4.3 Observations of leaf characteristics

Average leaf size, shape and any specific micro-morphological characters observed (Appendix 1) are given in Table 4.1. Out of all the species studied, micromorphology of *T. vulgaris* was noticeably more complex in both leaf-surfaces due to their densely arranged short stubby trichomes and essential oil secretory glands/glandular hairs (Fig. 4.5). Epicuticular wax layers on both surfaces of *T. vulgaris* and on the adaxial surface of *H. helix* were prominent compared to other species. In addition, slightly prominent wax plates or wax layers were observed on adaxial surfaces of *H. youngi*, *H. albicans* and *H. sternii*. Both leaf surfaces of *G. macrorrhizum* were hairy with densely arranged hairs and glandular trichomes, and there were a few other species with sparsely arranged hairs (Table 4.1).

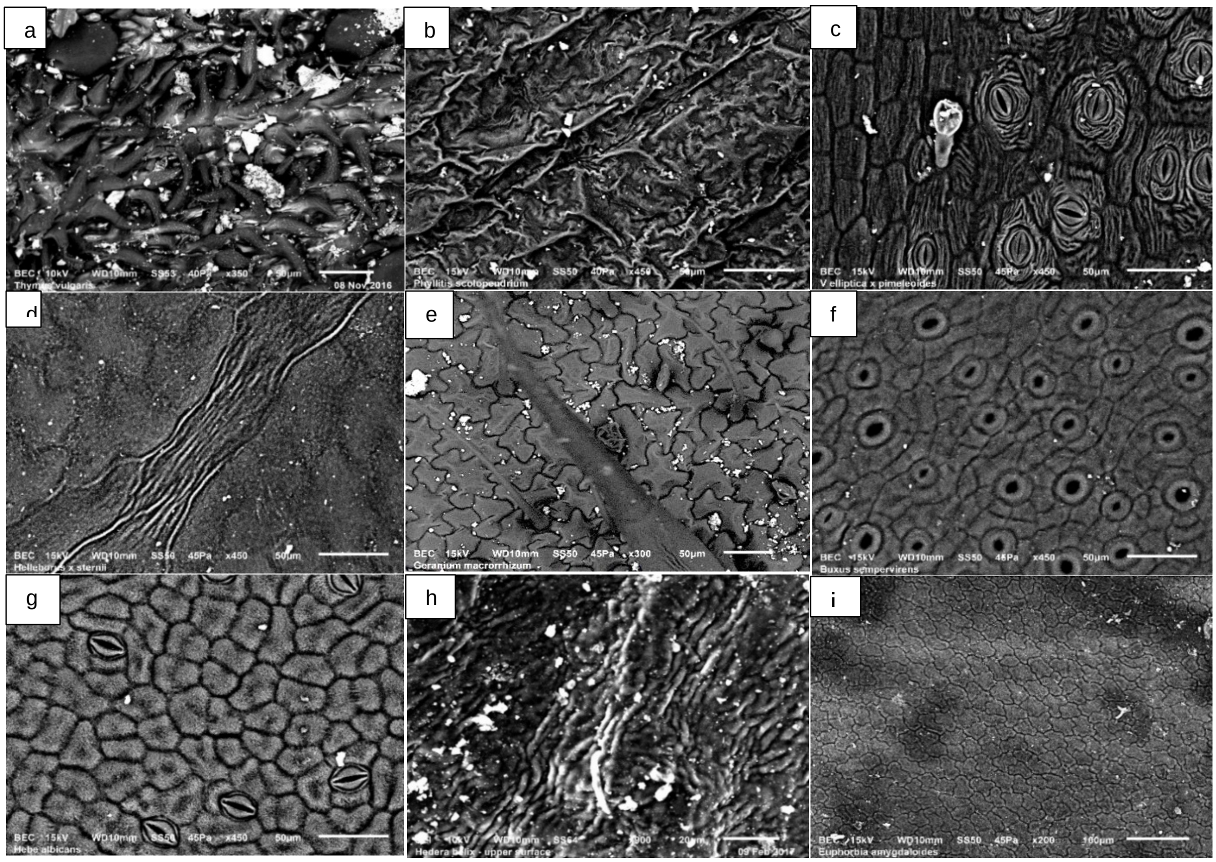


Figure 4-5 Sample Scanning Electron Microscope images of leaf micromorphology on the a) adaxial surface of *T. vulgaris* (x350), b) adaxial surface of *P. scolopendrium* (x450), c) abaxial surface of *H. youngii* (x450), d) adaxial surface of *H. sternii* (x450), e) adaxial surface of *G. macrorrhizum* (x450), f) abaxial surface of *B. sempervirens* (x450), g) adaxial surface of *H.albicans* (x450), h) adaxial surface of *H. helix* (x900), and i) adaxial surface of *E. amygdaloide.*

4.4.4 Elemental analysis

A wide range of elements were found in the PM captured on all species of plants in various quantities (Fig. 4.6 and Appendix 2). The mean weight percentage (Wt%) of elements found in PM captured on leaves of different species of plants are given in Table 4.5 (as C and O are mostly derived from plant material, they were excluded from the analysis). The most abundant element, Fe, was found in the PM captured by all species of plants. Ca, K, Mg and Si were also found in all plant species in variable quantities with levels being Ca> Si> K> Mg on average. The heavy metals Ti, Cr, Cu, Mn, Sb, Co and Zn were also found in these particles; however, they were found in trace quantities. The amount of Ti was relatively high compared to other heavy metals.

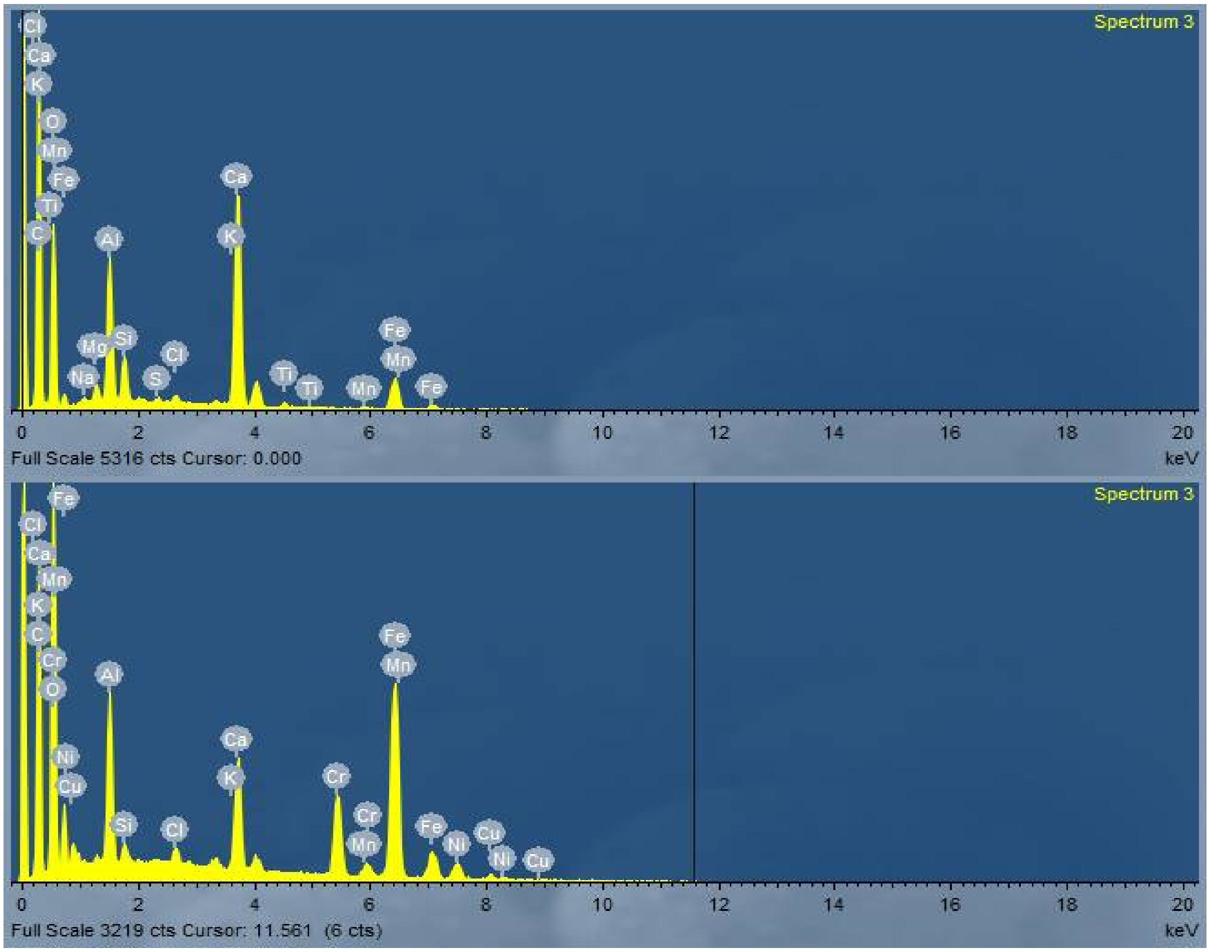


Figure 4-6 Sample EDX spectra of elemental compositions of PM captured on leaves of *B. sempervirens* (top) and *B. spicant* (bottom) grown on a living wall at New Street Station, Birmingham, UK in 2016.

4.5 Discussion

4.5.1 PM removal capacity of living walls

Plants growing in a living wall near a train station were shown to be capable of capturing a considerable amount of particulate pollution. It is likely that variable PM capture levels between different PM size fractions was due to their different aerodynamic behaviour and hence variable deposition velocities (Slinn, 1982). Dry deposition of PM on leaves occurs via different processes (e.g. sedimentation under gravity, impaction, interception), resulting in different deposition velocities. PM trapping on leaves could also be temporary since there is a possibility of remobilisation, e.g. wash-off by rain or re-suspension by wind (Currie and Bass, 2008; McPherson *et al.*, 1994; Terzaghi *et al.*, 2013). Gregory (1973) found different remobilisation rates of different particle masses, this may also influence the quantities of PM on

Table 4.5 The Mean weight percentage (Wt%) of elements* found in the PM captured on leaves of different species of plants on a living wall at New Street Station, Birmingham, UK in 2016.

Species	Mean weight percentage of elements in particulates (Wt%)																			
	Al	**Ba**	**Ca**	**Cl**	**Co**	**Cr**	**Cu**	**Fe**	**K**	**Mg**	**Mn**	**N**	**Na**	**Ni**	**P**	**S**	**Sb**	**Si**	**Ti**	**Zn**
A. maritima	0.08	0.32	4.09	0.33	-	-	0.10	4.52	0.18	0.05	-	-	-	-	0.15	2.11	-	0.23	0.77	0.16
B. sempervirens	1.03	-	7.38	0.88	-	-	-	0.65	0.46	0.23	3.03	-	0.06	-	4.84	0.50	-	0.33	30.44	-
B. spicant	1.80	1.38	2.23	1.32	-	2.01	0.15	6.17	1.19	0.21	0.03	-	0.54	0.67	0.07	1.33	-	3.60	-	-
E. amygdaloides	0.24	-	2.97	0.18	-	-	-	0.21	0.39	0.15	-	-	0.02	-	0.57	0.03	-	0.56	0.02	-
G. macrorrhizum	-	0.09	4.00	1.49	-	-	0.09	5.77	0.14	0.38	-	-	0.88	-	2.12	0.20	-	0.05	-	-
G. odoratum	0.07	0.21	2.60	0.36	0.05	-	0.07	17.02	1.06	0.37	-	-	0.04	-	0.49	0.92	-	2.23	0.10	0.04
H. albicans	0.22	0.65	1.69	2.79	-	-	0.53	3.29	0.21	0.34	-	-	1.60	-	0.14	1.09	-	2.10	-	-
H. salicifolia	1.87	0.04	0.86	0.01	-	-	0.09	4.21	1.53	0.17	-	-	0.28	-	0.03	0.51	-	3.99	0.03	-
H. helix	0.04	0.09	4.14	0.41	-	-	0.09	6.14	1.29	0.49	-	-	0.10	-	0.20	0.15	-	0.96	-	-
H. officinalis	0.66	0.09	2.15	0.77	0.05	-	0.61	6.95	0.49	0.44	-	-	0.01	-	0.20	0.95	-	1.84	0.21	0.42
H. sternii	0.49	-	2.20	0.25	-	-	0.01	4.56	0.22	0.31	0.01	-	0.05	-	0.17	0.18	-	3.26	0.01	-
H. youngii	1.06	0.23	0.14	-	-	-	0.01	46.21	0.57	0.08	0.39	2.14	-	-	-	-	-	1.50	0.02	-
L. nivea	0.67	0.68	0.46	2.54	-	-	0.30	19.00	0.32	0.24	0.02	-	1.52	-	0.07	0.25	-	1.91	0.03	0.23
P. scolopendrium	1.19	0.09	1.11	1.24	-	-	0.54	14.84	0.23	0.51	0.05	-	0.32	-	0.10	0.13	-	3.15	0.16	0.08
P. terminalis	0.17	-	2.00	0.24	-	0.13	-	4.05	0.39	0.22	0.09	0.08	0.02	-	0.07	0.11	-	1.28	-	-
P. veris	0.38	0.11	8.04	0.58	-	-	0.30	3.64	0.60	0.98	-	-	0.01	-	2.11	0.26	0.11	2.06	-	0.05
T. vulgaris	0.72	-	1.34	0.23	-	-	-	14.46	0.63	0.14	0.09	-	0.04	-	0.23	0.02	-	3.52	-	-

*as C and O are mostly derived from plant material, they were excluded from the analysis and hence weight percentages do not add-up to 100%

leaves. Here, it was found that the smaller the diameter of particles the higher the quantities captured, reflecting the findings of Freer-Smith *et al.* (2005) on conifers and Ottele *et al.* (2010) on *Hedera helix*. If we consider all seventeen plant species, the average number of PM_1 was 2.5 times higher than $PM_{2.5}$ and 9 times higher than PM_{10}, suggesting that the living wall is more effective in removing smaller sized-particles or that the leaves are better at retaining the smaller sized particles (Przybysz *et al.*, 2014). In contrast, Dzierzanowski *et al.* (2011) found relatively larger numbers of bigger particles on plant leaves compared to smaller particles using a gravimetric method (weight/area). This disparity might be attributed to different techniques applied in PM quantification. The higher mass of coarse particles compared to fine particles may result in higher weight/area in the gravimetric method, whereas the SEM/image analysis approach quantifies the number of different PM size fractions captured on leaves. The proportion of different PM size fractions in the atmosphere in different locations may also be a reason for this disparity.

4.5.2 Inter-species variation in PM capture

There was a considerable variation in PM removal capacity by different species of plants. Higher capture levels of all PM size fractions by *H. albicans*, *B. sempervirens*, *T. vulgaris* and *H. youngii* show their greater potential to remove particulates from the atmosphere. Reflecting the findings of Freer-Smith *et al.*, (2004) on woodland species, all these best PM-capturing species are smaller-leaved (Table 4.1). PM densities on leaves of these species (ignoring the impact of LAI) were also high suggesting an important role of leaf size in removing PM from air. The LAI of these species further enhances their PM removal capacity. Regardless of having smaller leaves, relatively low PM densities found in *H. officinalis* and *G. odoratum* (Table 4.3) may be due to their soft nature (less rigidity). Soft leaves with low rigidity may have a reduced ability to withstand the air-flow (containing PM) and hence have less turbulence around the leaf boundaries resulting in low levels of PM deposition. Also, their simple leaf arrangement, with larger gaps between their leaves than other small-leaved species, might create less turbulence around the foliage leading to lower levels of impaction and interception. The relatively low LAI of these species further reduces their PM removal capacity resulting in significantly lower capture levels in all particle size fractions (Fig. 4.3).

PM capture levels on the poorly performing *B. spicant* (Fig. 4.3) was 50 and 65 times lower (for PM_1 and $PM_{2.5}$ respectively) than the highest capture levels on *H. albicans*, and sixty-fold lower than the PM_{10} capture levels on the best performing species, *T. vulgaris*. The second lowest performing plant, *P. scolopendrium*, also showed very poor PM capture levels on its leaves and these quantities were significantly lower than majority of other species (Fig. 4.3). Interestingly, regardless of their different leaf morphology (Table 4.1), both of these lower performing species are ferns which are commonly grown on living walls. In contrast to their important role in removing some VOCs (eg. formaldehyde) (Kim *et al.,* 2008), ferns may not be considered as good PM filters.

Low PM densities on leaves of wide leaved species (*P. scolopendrium*, *H. sternii* and *P. veris*) with the exception of *G. macrorrhizum* reflect the findings of Beckett *et al.* (2000) and Hwang *et al.* (2011) that those species with broad leaves have low PM capture potential. Hairy leaves with a complex micromorphology have frequently been cited as being effective in capturing more particulates than smooth leaved plants by trapping them on the leaf-hairs/trichromes (Beckett *et al.*, 2000b; Leonard *et al.*, 2016; Ram *et al.*, 2012; Sæbø *et al.*, 2012). The high PM_1 densities on *G. macrorrhizum* may have resulted, in part, due to dense surface hair of their leaves (Table 4.1). In contrast, the densities of $PM_{2.5}$ and PM_{10} on *G. macrorrhizum* leaves were relatively low; however, the PM removal ability in those particle size fractions were higher once its relatively high LAI was taken into account. In addition to their smaller leaves, high PM densities on *T. vulgaris* may also be attributed to their larger number of trichomes and glandular hairs. As there were no other hairy-leaved species present in this living wall system, the impact of leaf hair/trichomes on trapping and retaining particles with reference to different PM size fractions requires more research using more hairy-leaved species.

Sæbø *et al.* (2012) found a positive correlation between PM accumulation on leaves and the leaf-wax content using trees and shrubs. In contrast, Dzierzanowski *et al.* (2011) concluded that PM accumulation on leaves is not related to wax content but to the chemical composition and structure of epicuticular wax; nevertheless, both waxy-leaved species used in this study, *T. vulgaris* and *H. helix*, showed relatively high PM densities in all size fractions suggesting an important role of leaf surface wax in removing particles. However, high PM capture levels on *T. vulgaris* could be attributed to any of those leaf properties (trichomes, essential oil glands, epicuticular wax and smaller size) or their collective impact combined with complex morphology and LAI (Fig. 4.5). In contrast to the findings of Shackleton *et al.* (undated) on the higher PM removal capacity of “grass like” (linear) species, *L. nivea* showed a very low potential in removing PM from the air. Currie and Bass (2008) found grass performed less well in PM removal compared to trees and shrubs and Dochinger (1980) mentioned that PM capture levels of deciduous species without leaves (as in late autumn and winter) are equivalent to the PM removal capacity of grasslands indicating their poor capture levels. Leonard *et al.* (2016) also found relatively low capture levels on linear shaped leaves. Effectiveness of “grass like” species in removing PM requires more research using multiple species.

4.5.3 PM densities on adaxial and abaxial surfaces of the leaves

Similar to the findings of Ottele *et al.* (2010) and Ram *et al.* (2012), PM capture levels on adaxial surfaces of the leaves were generally higher compared to abaxial surfaces. This could be due to the orientation of the leaves in space where the adaxial surface gets more exposure to particulates through sedimentation under gravity. Compared to the density of PM_1, a greater number of species show significant differences in their $PM_{2.5}$ and PM_{10} levels between the adaxial and abaxial surfaces of the leaves, probably due to the higher influence of sedimentation on larger particulates.

4.5.4 Elemental composition

The PM captured by all plant species showed a wide range of important elements including heavy metals, regardless of their PM capture efficiency. Rail traffic was the closest pollution source to the living wall with the nearest potential road pollution source some 47 m away. Carbon and Oxygen were present, mainly due to the organic materials of leaves. However, since the hourly level of black carbon has been reported as being up to 29 $\mu g.m^{-3}$ in Birmingham New Street Station (Zulkifli, 2015 cited in Thornes *et al.*, 2016) a considerable portion of carbon can probably be attributed to diesel exhaust from trains. In addition to diesel exhaust, hydrocarbons from lubrication oils, wooden sleepers, and wheel flanges could also be sources of these C and O levels (Burkhardt *et al.*, 2008). Higher quantities of Fe in PM are mainly due to engine wear (Lombaert *et al.*, 2004) and abrasion of wheels and brake pads (Burkhardt *et al.*, 2008; Thorpe and Harrison, 2008). Removing these Fe-rich metal particles from the air may be particularly beneficial due to their potential of causing oxidative brain damage which potentially leads to neurodegenerative conditions such as Alzheimer’s disease and Parkinson’s disease (Allsop *et al.*, 2008; Maher *et al.* 2013). Phosphorus, S and Si were found on almost all of the species and are typical diesel exhaust particles and Cu, Ca, and Mg probably originate from exhaust particles (Abbasi *et al.*, 2013). In addition to Al and Si from road dust (Chow *et al.*, 2006), Ca, Si, Na, and Al were probably emitted by the friction between wheels and railway lines, from being the main elements of ballast and concrete sleepers. Ca, Mg, Cu and Zn could have also resulted from lubrication oil additives in rail traffic (Lombaert *et al.*, 2004). The trace quantities of Ti, Mn, Ba, Cu, Sb and K found in the particulates can be mainly attributed to wear of brake pads (Thorpe and Harrison, 2008). Trace amounts of Ni and Cr could be associated with particles generated through wheel abrasion (Burkhardt *et al.*, 2008).

4.5.5 Implications for the use of Living walls in PM reduction

This study showed a considerable potential for living wall plants to remove PM pollutants from the atmosphere and the efficiency of smaller particle removal was notable. However, high variability in PM removal capacity of different species of plants highlights the importance of careful species selection for living wall systems when using them as PM filters. Smaller leaved species with complex morphology were found to be the best performing species in this respect. Beckett *et al.* (2000b) noted that evergreen species retain their leaves for several years and reach a saturation point, whereas Dzierzanowski *et al.* (2011) argued that PM captured on leaves can be washed-off with rain allowing them to act as a sink for PM throughout the year. As living wall species are mostly evergreen, PM remobilisation behaviour of the species is an important factor to be explored and requires more research. However, plants in most living wall systems are easily removed and can be replaced after a few seasons if required.

4.6 Conclusion

The living wall located by New Street Station showed a promising potential for capture of atmospheric PM pollutants. The effectiveness of capturing smaller particles appeared to be substantially higher compared to larger particles. Inter-species variation of PM capture by living wall plants was significant and smaller leaved species with a high LAI were found to have a higher PM removal potential compared to species with wider leaves. Results suggested that hairy-leaved species could be better in capturing the particularly hazardous PM_1 fraction, and the epicuticular wax and surface morphology of leaves may be important traits helping in the trapping all PM size fractions. However, further research using a greater number of species is required to draw more accurate conclusions in this respect. All the plant species studied were shown to have removed a wide range of elements from the atmosphere including potentially hazardous heavy metals.

Chapter 5 : The potential of living wall species to immobilise PM associated with road-traffic

This chapter compiles three different experiments conducted in Stoke-on-Trent, UK, focusing on different aspects relevant to the use of living walls as PM traps. The experiment described in Section 5.1 explored the potential of twenty living-wall plants to capture and retain traffic-based PM, covering the aspects of inter-species variation, the impact of leaf morphology on PM capture and the elemental composition of the captured PM. This experiment is presented, with slight modification, as an article published in the *Journal of Science of the Total Environment* (Weerakkody *et al.* 2018b). The experiment described in Section 5.2, explored the impact of planting design on the pollutant capture potential of living walls. The experiment described in Section 5.3 explored the potential use of scrambling species in living walls to filter-out PM pollutants and, the effect of inter-species variation in PM capture within a single genus.

5.1 Quantification of the traffic-generated particulate matter capture by plant species in a living wall and evaluation of the important leaf characteristics.

5.1.1 Abstract

Traffic-generated particulate matter (PM) is a significant fraction of urban PM pollution and little is known about the use of living walls as a short-term strategy to reduce this pollution. The present study evaluated the potential of twenty living wall plants to reduce traffic-based PM using a living wall system located along a busy road in Stoke-on-Trent, UK. An Environmental Scanning Electron Microscope (ESEM) and ImageJ software were employed to quantify PM accumulation on leaves (PM_1, $PM_{2.5}$ and PM_{10}) and their elemental composition was determined using Energy Dispersive X-ray (EDX). Inter-species variation in leaf-PM accumulation was evaluated using a Generalised Linear Mixed-effect Model (GLMM) using time as a factor; any differential PM accumulation due to specific leaf characteristics (stomatal density, hair/trichomes, ridges and grooves) was identified. The study showed a promising potential for living wall plants to remove atmospheric PM; an estimated average number of 122.08 ± 6.9 x 10^7 PM_1, 8.24 ± 0.72 x 10^7 $PM_{2.5}$ and 4.45 ± 0.33 x 10^7 PM_{10} were captured on 100 cm^2 of the living wall used in this study. Different species captured significantly different quantities of all particle sizes; the highest amount of all particle sizes were found on the leaf-needles of *Juniperus chinensis* L., followed by smaller-leaved species. In the absence of an apparent pattern in correlation between PM accumulation and leaf surface characteristics, the study highlighted the importance of individual leaf size in PM capture irrespective of their variable micro-morphology. The elemental composition of the captured particles showed a strong correlation with traffic-based PM and a wide range of important heavy metals. We conclude that the use of living walls that consist largely of smaller-leaved species and conifers can potentially have a significant

impact in ameliorating air quality by removing traffic-generated PM pollution to improve the wellbeing of urban dwellers.

Key words: Green infrastructure; Air pollution; Traffic-based pollution; Leaf characteristics

5.1.2 Introduction

Epidemiological studies have revealed short and long-term impacts of PM and found increased levels of hospital admissions, morbidity, and mortality around the world (Pascal *et al.*, 2014; Pope III *et al.*, 2011; Seaton *et al.*, 1995). Traffic-generated particulate matter (PM) is known to be responsible for a significant portion of urban particulate pollution (Pant and Harrison, 2013; Ranft *et al.*, 2009). Globally, 25% of $PM_{2.5}$ (PM with less than 2.5 µm aerodynamic diameter) and PM_{10} (PM with less than 10 µm aerodynamic diameter) were estimated as being generated from road traffic (Karagulian *et al.*, 2015) and in Europe road traffic is known to be responsible for more than 50% of PM_{10} (Künzli *et al.*, 2000). Some studies found direct links between traffic-based PM and detrimental health effects (Brook *et al.*, 2010; Hyun Cho *et al.*, 2005; Ranft *et al.*, 2009). Long-term exposure to these particles is known to cause premature death and various illnesses such as cardio-pulmonary diseases, lung cancer, allergies, brain damage and neuro-degenerative diseases (Brook *et al.*, 2010; Pascal *et al.*, 2014; Pope III *et al.*, 2011; Seaton *et al.*, 1995; Maher *et al.*, 2016), and have even been linked to cognitive impairments in elderly people (Ranft *et al.*, 2009). Pascal *et al.* (2014) also found a considerable rate of cardiovascular, cardiac, and ischemic mortality following short-term exposure to PM. Vehicular emission of $PM_{0.1}$ in the ultrafine range was recently highlighted as being important due to its higher toxicity and larger numbers compared to other size fractions (Lin *et al.*, 2005). PM such as elemental carbon, hydrocarbons, metals, oxides of nitrogen, sulphates, ammonia (Fauser, 1999; Sharma *et al.*, 2005) from vehicle exhaust, and various non-exhaust emissions from brake wear, tyre wear, clutch wear, abrasives, and road dust, are responsible for this pollution (Mulawa *et al.*, 1997; Slezakova *et al.*, 2007; Thorpe and Harrison, 2008; Timmers and Achten, 2016; Wåhlin *et al.*, 2006).

Despite several initiatives taken to mitigate this pollution, such as reducing emission levels and separating high emission zones from populous areas (Pugh *et al.*, 2012a), particulate levels in air still remain problematic for human health and the environment (e.g. reducing visibility, affecting urban thermal environment) (Han *et al.,* 2012; Yan *et al.*, 2016). Since plants are known to filter these pollutants, the ability of vegetative barriers to reduce near-road PM levels has recently been studied under different environmental and weather conditions using various techniques (Abhijith *et al.*, 2017; Blanusa *et al.*, 2015; Gautam *et al.*, 2005; Maher *et al.*, 2008; Mori *et al.*, 2015; Šíp and Beneš, 2016; Tong *et al.*, 2015; Weber *et al.*, 2014; Weerakkody *et al.*, 2017). The potential of vertical greenery systems is frequently cited as a possible short-term solution due to their smaller land utilization, quick installation, reduced dependency on existing soil conditions, and additional ecosystem services (Cheetham *et al.*, 2012; Dover, 2015; Johnston and Newton, 2004). Previous research has mainly focused on the use of climbing species to capture PM with most not specifically relating to road traffic (Ottelé *et al.*, 2010; Sternberg *et* *al.*, 2010; Tiwary *et al.*, 2008; Xia *et al.*, 2011) but see Dover and Phillips (2015). Except for the work of Perini *et al.*, (2017), who studied two shrub species and two climbing species in a green façade located above street level, a small-scale study on shrubs (which included few living wall species) by Shackleton *et al.* (undated), and work by Weerakkody *et al.* (2017), relating to PM generated by rail traffic, the value of living wall plants in the reduction of motor traffic-based PM has not been evaluated.

Living walls are vertical greenery systems holding multiple species of plants usually grown at ground level, and contrasts with facade systems using climbers; living walls require piped irrigation systems which typically also deliver nutrients (Dover, 2015). The present study evaluated the potential of different living wall plants to capture and retain near-road PM which are mainly derived from traffic pollution. Inter-species variation in capture and retention of PM under the same environmental and weather conditions were evaluated to identify the best species to use as near-road PM filters. Leaf micro-morphological variations suspected to control their ability to capture and retain PM (Dzierzanowski *et al.*, 2011; Kardel *et al.*, 2012; Leonard *et al.*, 2016; Sæbø *et al.*, 2012; Räsänen *et al.*, 2013; Wang *et al.*, 2011; Weerakkody *et al.*, 2018a) were also investigated. The elemental composition of captured particles was analysed to identify their links to traffic-generated PM and potential health effects.

5.1.3 Material and methods

5.1.3.1 Site description

An experimentally manipulable, freestanding, modular living wall system was employed in this research. The wall was donated, designed, and installed by Nemec Cascade Garden Ltd., Czech Republic, in June 2016. The wall was erected 6 m from the edge of Leek Road, a busy 'A' road adjacent to Staffordshire University, Stoke-on-Trent, UK (Fig. 5.1). The road has an average daily traffic flow of 20,251 vehicles (Department for Transport, 2017). Stoke-on-Trent is a city located in central England with a population density of 6,640 people/km^2. The local mortality burden attributed to anthropogenic $PM_{2.5}$ in Stoke-on-Trent was estimated at 5.6% in 2009 (Gowers *et al.*, 2014).

This living wall was a first-generation prototype, having two 3.98 m x 2.09 m planting areas, one on each side of the wall. The planting areas were separated from the ground by a 0.5 m tall concrete platform encased in metal. This was an artificially irrigated system equipped with water and thermal sensors for easy maintenance. As traffic pollution is of concern, only the planting area of the front side (facing the road) is described here. Ten plants per species of twenty different species (Table 5.1), both evergreen and deciduous species, with various leaf morphotypes (size, shape and micromorphology) were planted as ten rows and twenty columns using a Latin Square design.

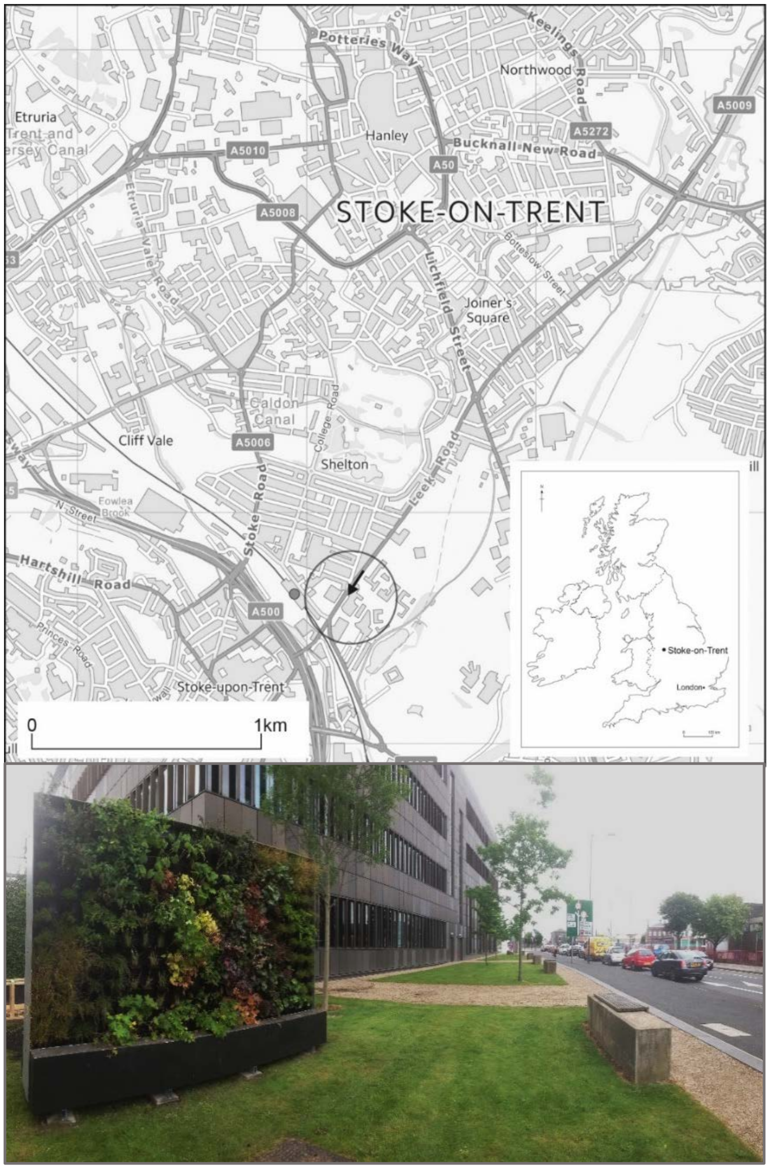


Figure 5-1 The location of the study site in Stoke-on-Trent, UK (top) *Contains OS data ©Crown copyright and database right (2017),* and the location and orientation of the living wall relative to the road (bottom).

5.1.3.2 Leaf sampling

Samples were only taken from the middle six rows of the wall, avoiding the upper most two rows (based on accessibility) and the bottom two rows to avoid ground-level contamination. Prior to sampling, plants were washed using a watering hose connected to a pressure head to remove existing particles from the leaves. Plants were left undisturbed for ten weeks (23rd of June, 2016 to 1st September, 2016) allowing continuous exposure to weather changes to minimize the effect of any variability in baseline PM levels. Sampling was carried out on six days during September and October 2016 under dry weather conditions. Thirty-six leaves from *Thymus vulgaris* and 18 leaves from each of the other species were taken in total.

Three leaves were randomly taken from each species on every sample day (i.e. 6 days x 3 leaves/day = 18 leaves) with the exception of *T. vulgaris* which had 6 leaves/day removed. All species were sampled on every sample day. Sampled leaves were arranged in clean plastic containers using blue tac on their petioles, avoiding cross contamination between leaves or with container surfaces (Weerakkody *et al.*, 2017, Chapter 4). Subsequently, containers were sealed and transferred to the laboratory for analysis. Samples were analysed fresh or stored at 9 °C in a refrigerator in the same storage containers until analysed (within 2 days of sampling) (Weerakkody *et al.*, 2017, Chapter 4).

5.1.3.3 Imaging leaves using a Scanning Electron Microscope (SEM)

Sample preparation and imaging followed the same approach as in Weerakkody *et al.* (2017) (Chapter 4). An Environmental Scanning Electron Microscope (ESEM) (Model: JSM-6610LV) was employed to image the particulates captured on leaves and the leaf micromorphology. Six leaf sections (5.0 mm x 5.0 mm), three from each adaxial and abaxial surface were excised from each of the leaves larger than 2.5 cm^2 in area. Smaller leaves (apart from *T. vulgaris*) were cut into halves, one half to visualize the adaxial surface, and one for the abaxial surface and mounted without further cropping. Leaves of *T. vulgaris* and leaf needles (*J. chinensis*) were scanned without cropping. *T. vulgaris* was too small to cut into halves and hence the adaxial and abaxial surfaces were scanned using adjacent leaves. Leaf sections were only excised from the leaf-blades (Weerakkody *et al.*, 2017, Chapter 4); for the leaves mounted without cropping, three scanning areas were randomly selected from the leaf-blade following the same approach. As leaves of *J. chinensis* were needle-like, rather than having leaf blades, a different sampling approach was used: areas to be scanned were selected from the middle of the needles avoiding their tips and base. Nine micrographs from each leaf surface (both adaxial and abaxial surfaces) were taken at 450x and 1,000x using back scattered electron signals under a low vacuum to quantify the amounts of PM captured on the leaves.

Duplicates of each micrograph were taken with and without labels (including species name, scale, magnification and accelerating voltage) to use in the image analysis process for scale definition. As needles of *J. chinensis* were cylindrical, it was not possible to distinguish specific surfaces and hence nine micrographs were taken from a random part of the leaf surface. The same working distance, brightness, and contrast levels were used for all scans to avoid any complications in the image analysis process.

Following visualization of particulates, a further set of micrographs were taken to image micro-morphological features from ten random leaves previously scanned to quantify particles. As surface structures such as leaf hairs and trichomes are substantially larger than particulates, leaf surfaces were scanned at 90 x, 100 x and 250 x; in order to image leaf stomata, grooves and ridges, leaves were scanned at 300 x and 450 x. As these characters were unevenly distributed on leaves, focus settings were changed accordingly and three random micrographs per character, from each adaxial and abaxial surface of the leaves, were taken.

Table 5.1 Plant species used in the experimental living wall with a brief species description [a]

Species name	**Species abbreviations**	**Common name**	**Species description and leaf morphology**
Acorus gramineus Sol.	A. g	Grass-leaf sweet flag	Semi-evergreen rhizomatous species. Liinear (“grass-like”), slightly rough leaves forming tufts.
Berberis buxifolia Lam.	B.b	Box-leaved barberry	Evergreen shrubs. Small, obovate, smooth and leathery leaves
Bergenia cordifolia (Haw.) Sternb.	B.c	Elephant's ears	Evergreen species. Large, ovate, glossy and smooth leaves.
Berberis × *media* Groot. ex Boom	B.m	Barberry 'Red Jewel'	Evergreen shrubs. Small, obovate, smooth and leathery leaves.
Carex caryophyllea Lat.	C.c	Spring sedge	Evergreen rhizomatous species. Linear, “grass like”, rough leaves.
Deschampsia cespitosa (L.) Beauv	D.c	Tufted hair grass	Evergreen bushy grass with rough, linear leaves forming dense tufts.
Geranium macrorrhizum L.	G.m	Cranesbill ‘Bevan's variety	Semi-evergreen species. Large, palmately-lobed, hairy leaves.
Geranium renardii Trautv.	G.r	Renard geranium	Deciduous herbaceous plant with lobed and furrowed leaves
Heuchera americana L.	H.a	American alum root	Evergreen species. Large, glossy and palmately-lobed leaves.
Heuchera villosa Michx. var. *macrorhiza*	H.v	Hairy alumroot	Evergreen species. Large, hairy and palmately-lobed leaves.
Helleborus × *sternii* Turrill	H.s	Backthorn strain	Evergreen species. Large, obovate, leathery leaves with serrated margins.
Veronica vernicosa Hook.F. (*Hebe vernicosa*)	V.v	Varnished hebe	Evergreen shrub. Small, elliptic, glossy and leathery leaves.
Juniperus chinensis L.	J.c	Chinese juniper	Evergreen conifer. Small, branched leaf needles with thick epicuticular wax.
Lonicera kamtschatica Pojark.	L.k	Blue honeysuckle	Deciduous shrub. Large, elliptic leaves with slightly hairy/velvety texture.
Persicaria amplexicaulis (D. Don) Ronse Decraene	P.a	Red bistort 'Firetail'	Deciduous herbaceous species. Large, cordate and slightly wrinkled leaves.
Phyllitis scolopendrium L.	P.s	Hart's tongue fern	Evergreen fern. Large, elongated leathery fronds with irregular margins.
Spiraea betulifolia Pall.	S.be	Tor	Deciduous shrub. Medium-sized, smooth and oval leaves.
Spirea japonica L.	S.j	Japanese meadowsweet	Deciduous species. Small and ovate leaves with toothed margins.
Stachys byzantina K. Koch × *Stachys debilis* Kunth	S.by	Lamb's ear 'Silver carpet'	Evergreen species. Densely hairy, velvety and elliptically shaped leaves.
Thymus vulgaris L.	T.v	Common thyme	Evergreen shrubs. Small, ovate leaves with slightly velvety surface texture.

[a] Leaf shape descriptions based on Hickey and King (2000) and Beentje (2012)

5.1.3.4. Analysing inter-species variation of PM capture

PM quantification followed the same approach described in Weerakkody *et al.* (2017) (Chapter 4). The micrographs taken for visualizing particulates were imported in imageJ image analysis software; those with labels were first used to define the scale, and those without labels were used to quantify PM_1, $PM_{2.5}$, and PM_{10} on each micrograph using the auto-threshold tool. As leaves have different surface areas, the amount of PM on leaves was expressed in terms of PM density, which is the number of particulates captured on a unit surface area (1 mm^2). The PM density on each leaf was estimated separately for adaxial and abaxial surfaces by taking the mean PM densities on the respective micrographs. The mean PM density on each leaf was calculated by totalling the mean PM densities on adaxial and abaxial surfaces. The total PM density on leaf needles of *J. chinensis* was estimated as twice the PM density on the imaged surface to make it comparable with the other species, although we recognised this may underestimate total PM density on cylindrical leaf needles.

Inter-species variation in total PM density was identified using a mixed-Anova in a Generalized Linear Mixed-effect Model (GLMM) using R statistical software version 3.2.5 (R Development core team, 2016); as leaves were sampled on six different occasions, sample date (time) was included as a random factor. This analysis was then separately repeated for adaxial and abaxial surfaces to identify any variability between the species. Any significant difference in PM density between adaxial and abaxial surfaces of the same species of plant was identified using the Student's t-test (R 3.2.5); as this was not applicable for needle-like leaves, *J. chinensis* was not included in this analysis.

The Leaf Area Index (LAI) was measured to estimate the total leaf area of the plants using the following formula (Weerakkody *et al.,* 2017, Chapter 4):

LAI = Mean surface area of a leaf x Mean number of leaves per quadrat/ total surface area of the quadrat

Ten random leaves per species were used to calculate the mean leaf size and three random 10 cm x 10 cm quadrats per species were taken to estimate the mean number of leaves in a unit area of living wall surface (Chapter 2). Individual leaf sizes were measured using ImageJ software, the area of leaf needles of *J. chinensis* was estimated as for cylindrical shapes (Zhang *et al.*, 2015). The total PM capture by a 100 cm^2 area of each species was then estimated using their mean PM densities on the leaves and LAI values (i.e. Mean PM density on the leaves x LAI of the species x 100 cm^2) (Weerakkody *et al.*, 2017, Chapter 4). Inter-species variation in total PM capture levels (incorporating LAI) were identified using a mixed-Anova in GLMM (R 3.2.5) including time as a factor.

5.1.3.5 Evaluating the correlation between leaf micromorphology and PM accumulation

The number of leaf hairs/trichomes and stomata on micrographs were quantified and expressed as number of characters per unit area (1 mm^2). Ridges and grooves on leaves were varied in shape, size and distribution and they were thus estimated as the percentage area covered by the character with reference to 1 mm^2 area of leaf surface using ArcGIS (ArcMap 10.4 © 2015 ESRI) (Chapter 2: Fig. 2.8). All epidermal protrusions of the leaves were classified as hairs/trichomes. The micrographs were loaded into ArcGIS as .jpg (image) files and an interactive supervised classification was performed by assigning twenty training zones on each character (grooves, ridges and back ground) to produce a classified image. Five hundred random points were allocated using the create accuracy assessments point tool and the number of those that fell on areas of each character was recorded. This random sampling procedure was repeated five times on each classified image and the mean number of random points that fell on each character (ridges and grooves) on each image was expressed as a percentage out of 500 total points.

The mean density of leaf hairs/trichomes, stomata, grooves and ridges were then calculated taking the mean densities of each of these characters in their respective micrographs. Ten of the leaves of each species used to assess PM levels (Section 5.1.3.3) were taken at random and used in this experiment. Any correlation between these leaf characters and their PM densities were then identified using GLMM; as these characters came from twenty species, plant species was included as a random factor in this model. As observed characters were not evenly distributed on the adaxial and abaxial surfaces, they were analysed separately.

5.1.3.6 Analysing the elemental composition of PM captured on leaves

Elemental analysis of captured PM was conducted whilst scanning the leaves using the ESEM for their PM densities described in Section 5.1.3.3 and 5.1.3.4. Once the particles were clearly focused at 450x and 1000x magnifications at 15 Kv accelerating voltage, the areas being scanned were acquired in Integrated Calibration and Application (INCA) microanalysis software coupled with the ESEM. The elemental composition of ten random particles from each PM size fraction per species (30 particles per plant species in total) were analysed using EDX (Weerakkody *et al.*, 2017, Chapter 4). The Point and ID analyser was used on selected particles to identify the elements and quantify their amount as a percentage of the total weight of the particle (Wt%) minimizing interference from the background leaf material.

5.1.4. Results

5.1.4.1 Inter-species variation in PM capture

Considering all the species of plants, on average, 122.08 ± 6.9 x 10^7 of PM_1, 8.24 ± 0.72 x 10^7 of $PM_{2.5}$ and 4.45 ± 0.33 x 10^7 of PM_{10} were estimated to have been captured on 100 cm^2 of the living wall. However, there was a significant inter-species variation in both PM densities ($p < 0.0001$ for all PM sizes and F = 34.69, 41.33 and 59.31 for PM_1, $PM_{2.5}$ and PM_{10} respectively) (Table 5.2) and in their overall ability to capture and retain PM when LAI was incorporated (Table 5.3 and Fig. 5.2) ($p < 0.0001$ for all PM sizes and F = 62.83, 59.31 and 32.11 for PM_1, $PM_{2.5}$ and PM_{10} respectively). The separate analysis conducted on adaxial and abaxial surfaces of the leaves also revealed a significant variation in PM densities between species (on adaxial surfaces: $p < 0.0001$ for all PM sizes and F = 16.64, 24.9 and 20.94 for PM_1, $PM_{2.5}$ and PM_{10} respectively, and on abaxial surfaces: $p < 0.0001$ for all PM sizes and F = 16.75, 41.73 and 12.82 for PM_1, $PM_{2.5}$ and PM_{10} respectively) (Fig. 5.3). *J. chinensis* showed the highest PM densities on its leaves (Table 5.2) and the highest overall PM capture levels (incorporating LAI) for all particle size fractions (Fig. 5.2). Both the PM density on leaves and the overall PM capture of *J. chinensis* were significantly higher than all the other species of plants for all PM sizes ($p < 0.0001$). The second highest densities of $PM_{2.5}$ and PM_{10} were found on leaves of *V. vernicosa* and the second highest density of PM_1 was found on leaves of *B. buxifolia,* which was not significantly different from the third highest density of PM_1 found on leaves of *V. vernicosa* (Table 5.2). Once the LAI was taken into account (Fig. 5.2), the second highest ability to remove PM was shown by *V. vernicosa* for all particle sizes. Leaves of *P. amplexicaulis* showed the lowest density of PM_1 and $PM_{2.5}$ and *B. cordifolia* showed the lowest density of PM_{10} (Table 5.2). Once the LAI was taken into account, the lowest ability to remove PM_1 and PM_{10} was shown by *L. kamtschatica* whilst *P. amplexicaulis* remained the lowest for capturing $PM_{2.5}$ (Fig. 5.2).

Table 5.2 Mean PM densities ± 1SE on leaves of plant species in the experimental living wall system on Leek Road, Stoke-on-Trent [b]

Species	Mean PM_1 density on leaves (x 10^3) ± SE	Group assigned by Tukey's HSD test	Mean $PM_{2.5}$ density on leaves (x 10^3) ± SE	Group assigned by Tukey's HSD test	Mean PM_{10} density on leaves (x 10^3) ± SE	Group assigned by Tukey's HSD test
A. gramineus	46.54 ± 2.96	def	1.99 ± 0.21	ef	1.44 ± 0.19	de
B. buxifolia	101.08 ± 7.34	b	6.25 ± 0.43	bc	2.44 ± 0.21	cd
B. cordifolia	23.14 ± 2.42	fg	1.04 ± 0.09	f	0.45 ± 0.07	e
B. ×media	76.86 ± 6.10	bc	5.34 ± 0.46	bc	3.10 ± 0.20	bc
C. caryophyllea	66.23 ± 5.75	cd	1.90 ± 0.19	ef	1.91 ± 0.19	cde
D. cespitosa	46.81 ± 3.79	def	2.08 ± 0.22	ef	1.79 ± 0.21	cde
G. macrorrhizum	51.81 ± 4.82	cde	2.04 ± 0.24	ef	0.70 ± 0.05	e
G. renardii	48.45 ± 5.21	def	2.02 ± 0.33	ef	1.38 ± 0.20	de
H. americana	39.39 ± 2.84	efg	1.66 ± 0.12	ef	1.51 ± 0.27	de
H. villosa	40.48 ± 2.61	defg	1.88 ± 0.30	ef	0.64 ± 0.07	e
H. × sternii	36.05 ± 5.62	efg	1.55 ± 0.10	ef	0.82 ± 0.09	e
V. vernicosa	96.35 ± 8.26	b	7.29 ± 1.12	b	4.07 ± 0.61	b
J. chinensis	143.90 ± 13.23	a	14.94 ± 1.60	a	6.14 ± 0.84	a
L. kamtschatica	18.77 ± 2.58	g	1.62 ± 0.17	ef	0.63 ± 0.03	e
P. amplexicaulis	16.49 ± 2.29	g	0.60 ± 0.04	f	0.60 ± 0.05	e
P. scolopendrium	35.04 ± 3.72	efg	1.38 ± 0.09	ef	1.65 ± 0.13	cde
S. betulifolia	33.80 ± 2.77	efg	2.51 ± 0.52	def	2.61 ± 0.50	bcd
S. japonica	52.77 ± 4.04	cde	5.03 ± 0.39	bcd	1.72 ± 0.18	cde
S. byzantina	51.59 ± 6.73	cde	2.62 ± 0.23	def	1.90 ± 0.24	cde
T. vulgaris	66.29 ± 6.05	cd	3.84 ± 0.16	cde	3.04 ± 0.25	bc

[b] PM density on leaves of species assigned with the same letter in each PM size range, were not significantly different within their respective PM size category (Tukey's HSD post hoc test, $p > 0.05$).

Table 5.3 Mean leaf size ±1SE, LAI ±1SE and mean quantities ±1SE of micro-morphological characters of the leaves of plant species used in the experimental living wall located near Leek Road, Stoke-on-Trent

Species	Leaf area±SE (cm^2)	LAI±SE	Micromorphology on adaxial surface of the leaves ±SE				Micromorphology on abaxial surface of the leaves ±SE			
			Stomatal density (mm^{-2})	Density of Hair(mm^{-2})	Grooves%	Ridges %	Stomatal density (mm^{-2})	Density of Hair (mm^{-2})	Grooves%	Ridges%
A. gramineus	5.5 ± 0.16	1.1 ± 0.01	78.8 ± 12.46	0	40.9 ± 1.0	43.7 ± 1.7	95.1 ± 4.76	0	35.8 ± 2.4	41.2 ± 1.8
B. buxifolia	1.5 ± 0.08	2.3 ± 0.03	0	0	7.6 ± 1.7	11.8 ± 2.5	207.3 ±13.17	0	10.9 ± 0.9	14.4 ± 1.0
B. cordifolia	69.8 ± 0.94	1.9 ± 0.12	0	0	0.3 ± 0.1	12.1 ± 0.8	53.7 ± 6.81	0	7.7 ± 1.0	11.3 ± 1.2
B. ×media	1.3 ± 0.08	2.2 ± 0.04	0	0	14.0 ± 0.5	24.9 ± 2.0	191.5 ± 35.72	0	22.7 ± 1.8	19.9 ± 2.5
C. caryophyllea	6.8 ± 0.13	2.0 ± 0.07	24.3 ± 6.70	23.0 ± 8.56	46.7 ± 2.8	50.9 ± 0.9	180.5 ± 48.76	4.2 ± 1.86	48.1 ± 1.7	48.6 ± 1.0
D. cespitosa	6.6 ± 0.12	2.3 ± 0.08	54.5 ± 7.62	75.9 ± 25.23	37.9 ± 1.1	40.5 ± 2.0	112.9 ± 5.37	34.0 ± 11.33	30.8 ± 2.4	36.2 ± 1.8
G. macrorrhizum	30.4 ± 2.03	3.3 ± 0.13	0	137.3 ± 23.39	31.4 ± 3.7	26.5 ± 2.5	238.7 ± 9.51	142.3 ± 20.48	26.2 ± 2.4	25.2 ± 2.6
G. renardii	7.1 ± 0.13	0.8 ± 0.06	0	4.6 ± 0.91	38.4 ± 2.9	40.1 ± 2.8	297.1 ± 73.69	56.7 ± 13.39	25.7 ± 2.8	25.8 ± 2.3
H. americana	34.7 ± 1.87	2.4 ± 0.10	0	5.2 ± 0.55	14.8 ± 2.0	9.9 ± 1.2	132.3 ± 6.25	27.2 ± 3.42	11.7 ± 1.7	14.4 ± 2.3
H. villosa	59.9 ± 1.67	1.5 ± 0.17	0	58.1 ± 12.52	21.8 ± 2.5	15.3 ± 2.0	147.6 ± 15.21	56.3 ± 10.51	16.7 ± 1.4	25.1 ± 2.7
H. × sternii	20.0 ± 0.57	0.8 ± 0.11	0	0	29.6 ± 1.9	37.4 ± 2.4	121.6 ± 7.64	0	5.9 ± 1.0	5.3 ± 0.7
J. chinensis	0.1 ± 0.001	3.3 ± 0.04	68.9 ± 15.58	0	27.3 ± 2.0	30.9 ± 2.6	73.3 ± 16.27	0	27.3 ± 2.0	30.9 ± 2.6
L. kamtschatica	12.8 ± 0.94	1.0 ± 0.04	0	37.2 ± 2.27	31.1 ± 3.0	26.6 ± 1.9	186.0 ± 14.97	25.2 ± 5.81	22.8 ± 2.1	28.6 ± 2.6
P. amplexicaulis	69.9 ± 0.25	1.6 ± 0.12	0	0	45.5 ± 1.7	46.6 ± 2.2	117.7 ± 7.32	10.1 ± 1.39	25.1 ± 2.3	27.8 ± 2.9
P. scolopendrium	19.4 ± 1.44	1.1 ± 0.06	0	5.3 ± 2.01	23.9 ± 2.2	24.5 ± 2.3	19.6 ± 3.04	0	27.6 ± 2.2	26.9 ± 2.0
S. betulifolia	3.8 ± 0.10	2.8 ± 0.19	0	0	26.2 ± 2.8	19.6 ± 1.8	286.4 ± 31.82	0	18.4 ± 1.2	22.5 ± 2.3
S. japonica	1.9 ± 0.11	2.2 ± 0.02	0	0	30.4 ± 2.1	27.9 ± 1.8	218.2±21.13	0	28.1 ± 2.6	31.7 ± 2.3
S. byzantina	14.1 ± 1.10	2.1 ± 0.06	0	393.9 ± 35.01	0	0	178.7 ± 41.28	323.6 ± 37.97	12.1 ± 1.3	16.2 ± 1.7
T. vulgaris	0.1 ± 001	1.8 ± 0.05	0	814.6 ± 107.2	28.1 ± 2.4	31.4 ± 1.7	457.3 ± 29.94	667.2 ± 52.05	27.8 ± 2.0	29.3 ± 1.7
V. vernicosa	0.1 ± 0.002	2.5 ± 0.02	89.0 ± 6.81	4.2 ±1.58	11.1 ± 1.6	12.6 ± 1.4	227.1 ± 14.14	0	14.5 ± 1.6	11.1 ± 1.6

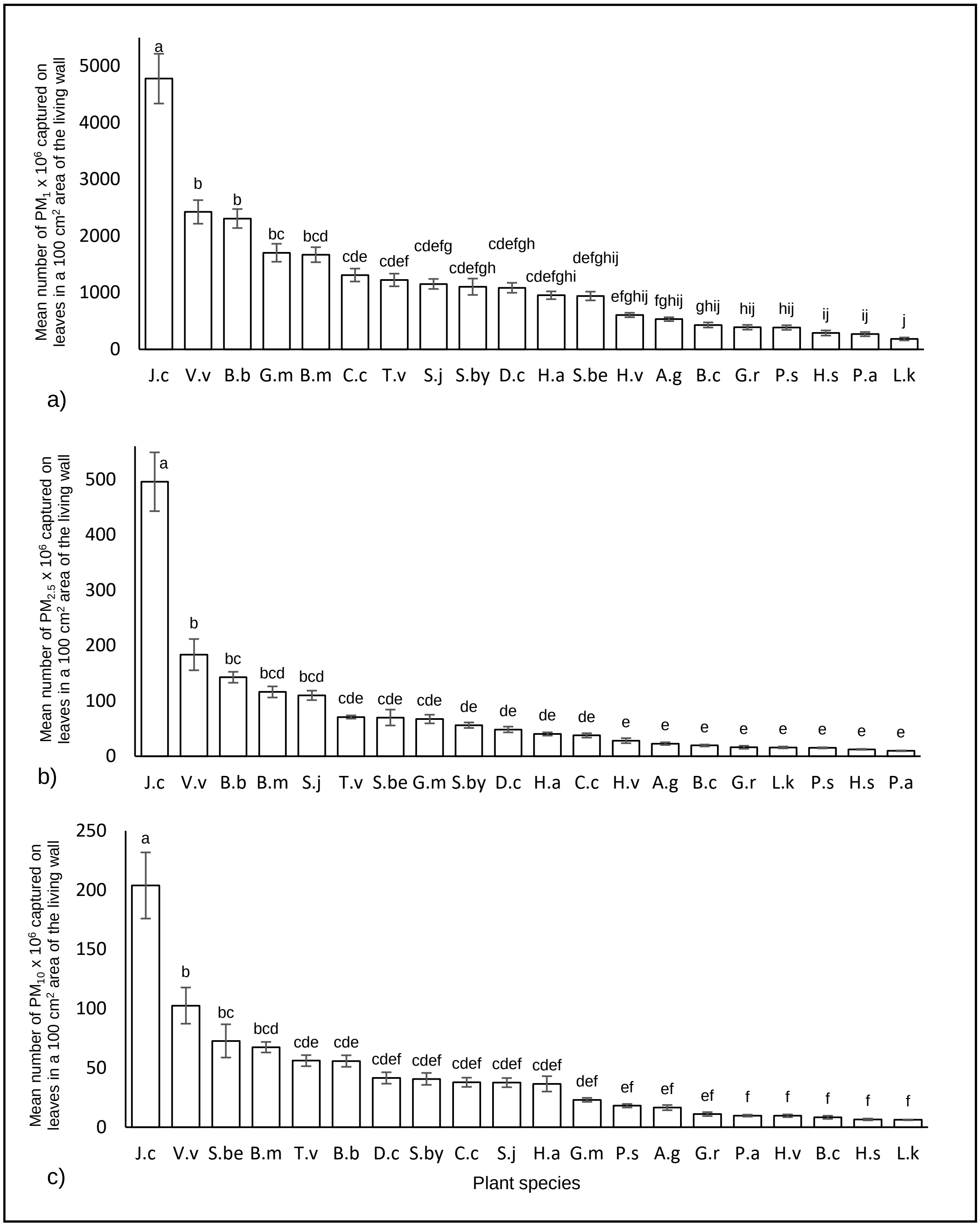


Figure 5-2 Estimated Mean number ±1 SE (x 10^6) of PM captured on leaves in a 100 cm^2 area of living wall (incorporating the LAI) by different species of plants on the experimental living wall on Leek Road, Stoke-on-Trent: a) PM_1 b) $PM_{2.5}$ and c) PM_{10}.

Data are arranged in descending order of PM accumulation for each size range, for an explanation of the species codes see further Table 5.1. Note different scales on the y-axes. *Species marked with same letters were not significantly different within each particle size category (Tukey's HSD post hoc test, p> 0.05).

The mean PM densities on the adaxial surfaces of the leaves were higher than those on the abaxial surfaces in all the species of plants apart from a slightly higher density of PM_1 on the abaxial surface of *B. cordifolia* which was not significantly different ($p = 0.48$) (Fig. 5.3). The mean PM density on the cylindrical surface of *J. chinensis* was significantly higher compared to both leaf surfaces of all the other species (Fig. 5.3). On the adaxial surfaces of the leaves, the second highest mean densities of all PM sizes were found on *V. vernicosa* and on the abaxial surfaces, the second highest density of PM_{10} was also found on *V. vernicosa.* On the abaxial surfaces, the second highest mean density of $PM_{2.5}$ was found on *S. japonica* and PM_1 on *B. buxifolia*. Leaves of *P. amplexicaulis* showed the lowest PM_1 and $PM_{2.5}$ densities on the adaxial surfaces of the leaves whilst *B. cordifolia* showed the lowest mean density of PM_{10}. On the abaxial surfaces, the lowest mean densities of all PM sizes were found on *P. amplexicaulis*.

5.1.4.2 Correlation between leaf micromorphology and PM densities

The densities of micro-morphological characters varied on leaves of different species of plants and between adaxial and abaxial surfaces of the same species (Table 5.3, Fig. 5.4 and Appendix 1). The density of PM_1 did not show any significant relationship between any of the leaf characters examined (hair/trichomes, stomata, grooves and ridges) on any of the leaf surfaces (Appendix 3). Of the characters examined, the stomatal density on the adaxial surfaces of the leaves showed a positive relationship with densities of $PM_{2.5}$ ($t = 38.99$ and $p = 0.034$) and PM_{10} ($t = 62.72$ and $p < 0.001$). Leaf hair/trichomes showed a positive relationship with the density of PM_{10} ($t = 3.27$ and $p = 0.0013$). PM densities on adaxial surfaces did not show any significant relationship with ridges or grooves (Appendix 3). Whereas on the abaxial surfaces, out of all the characters, only ridges showed a positive relationship with the density of $PM_{2.5}$ ($t = 2.99$ and $p = 0.0031$). The density of PM_{10} on abaxial surfaces was only correlated with stomatal density ($t = 2.39$ and $p = 0.018$) but not for any of the other characters.

5.1.4.3. Elemental composition of the captured particles

Twenty-eight different elements Ag, Al, Ba, C, O, N, Br, Ca, Cl, Cr, Co, Cu, F, Fe, K, Mg, Mn, Na, P, S, Sb, Si, Sn, Ti, Tl, V, Zn and Zr were found, in various quantities, in PM accumulated on leaves (Fig. 5.5 and Appendix 2). The amounts of C and O were considerably larger compared to other elements and they were thus excluded from the graphs to improve the clarity. Out of all the elements Co, Br and Tl were found only in PM_1 and N, Sb and V were only found in PM_{10}. Leaves of all the species carried PM with Ca, K, Mg and S in all particle sizes. In addition, Fe and Cl were present in PM_1 captured on leaves of all the species whilst Si was found in PM_{10} captured on all the species. In terms of quantity, the highest Wt% was shown by Fe (13.16%) in all particle size fractions (apart from C: 48.43% and O: 32.62%).

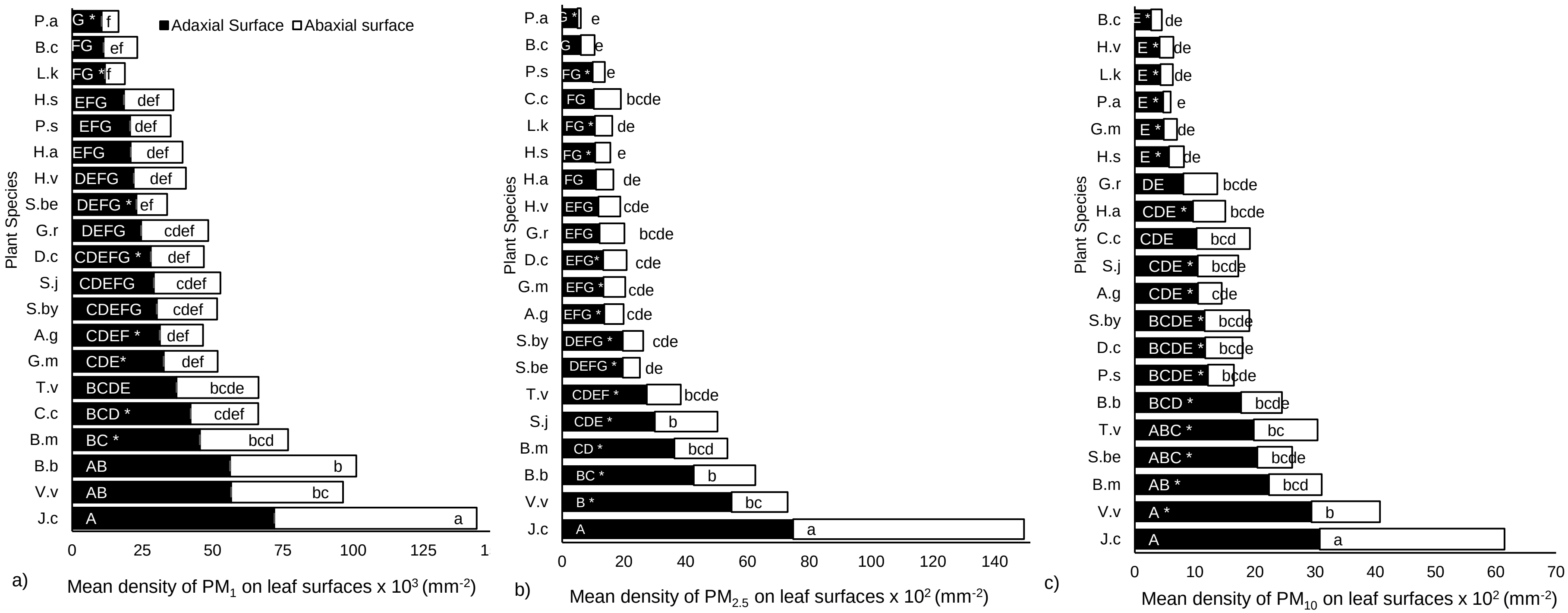


Figure 5-3 Mean PM density ±1 SE on 1 mm^2 of the adaxial and abaxial surfaces of the leaves of plant species on the experimental living wall located on Leek Road, Stoke-on-Trent (PM densities are given in different scales): a) PM_1 b) $PM_{2.5}$ and c) PM_{10}.

Plant species marked with the same letters are not significantly different from each other (p > 0.05). ***Upper case letters*** *- Variation between adaxial surfaces of the leaves;* ***lower case letters*** *- variations between abaxial surfaces of the leaves. Note different scales on the y-axes.* ***** *indicates that the PM densities on adaxial* and abaxial surfaces were significantly different. For explanation of the species codes see further Table 5.1.

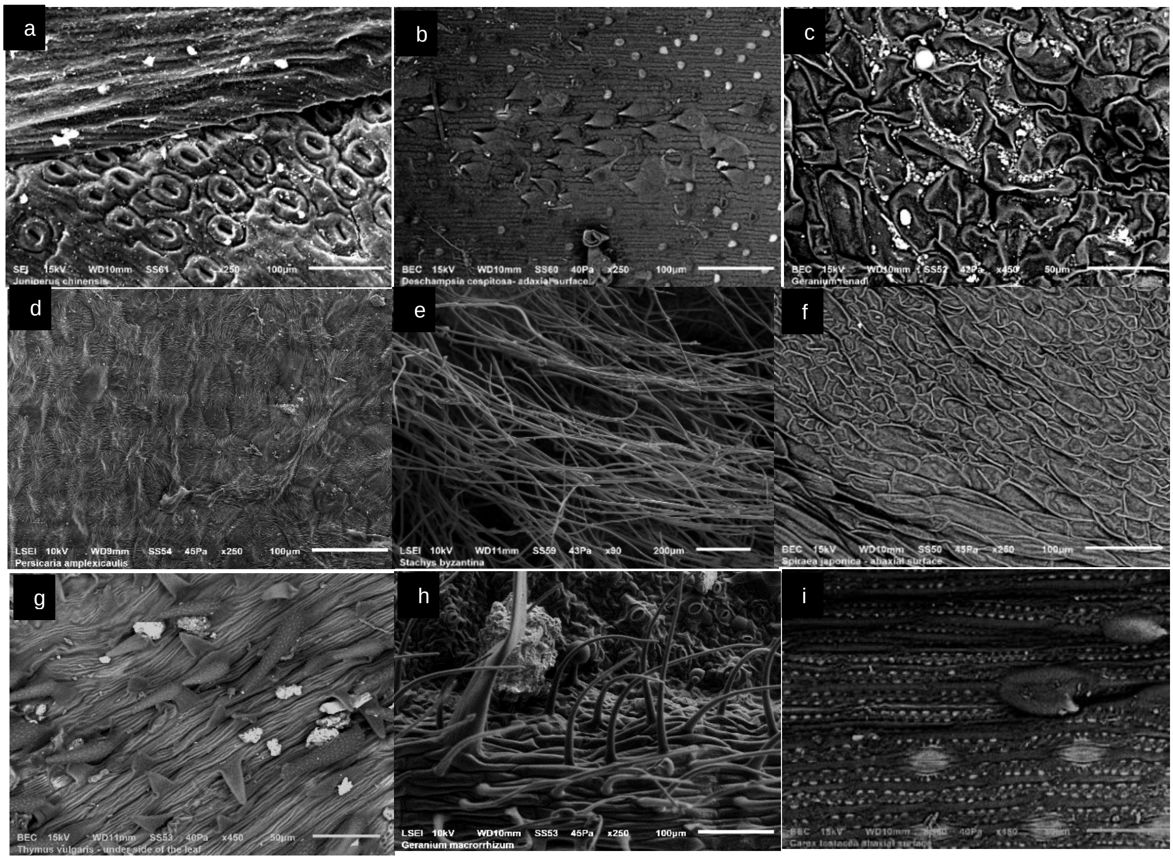


Figure 5-4 ESEM micrographs showing micro-morphological characteristics on leaves of some plant species in the experimental living wall located on Leek Road, Stoke-on-Trent: a) leaf needles of *J. chinensis* (x250), adaxial surfaces of b) *D. cespitosa* (x100) c) *G. renardii* (x450) d) *P. amplexicaulis* (x250) e) *S. byzantine* (x90) and abaxial surfaces of f) *S.japonica* (x250) g) *T. vulgaris* (x450) *h) G. macrorrhizum* (x250) i) *C. caryophyllea* (x450).

5.1.5. Discussion

5.1.5.1 Inter-species variation PM capture by leaves

All the plant species on the living wall located in Staffordshire University along Leek Road showed considerable levels of PM capture and retention ability. In agreement with the results of Freer-Smith *et al.* (2005), Ottele *et al.* (2010), and Weerakkody *et al.* (2017), the total amount of particles captured on leaves increased with decreasing particle diameter. At the species level, although the majority of the species followed the same pattern, the amount of PM_{10} on *C. caryophyllea*, *P. amplexicaulis, P. scolopendrium* and *S. betulifolia* were slightly higher or equal to their $PM_{2.5}$ levels (Fig. 5.2). These varied deposition levels of particles with different aerodynamic diameter probably reflects the different aerodynamics properties of the particles and their interactions with different leaf characteristics (see Davidson and Wu, 1990; Petroff *et al.*, 2008a; Slinn, 1982; Weerakkody *et al.*, 2017). There was a considerable variation in PM densities (i.e. excluding LAI) and in overall PM accumulation (i.e. including LAI) on different species, as has been found previously for various types of green infrastructure (Blanusa *et al.*, 2015; Beckett *et al.*, 2000b; Freer-Smith *et al.*, 2004; Song *et al.*, 2015). The inter-species variation in total PM density and in density on the adaxial and abaxial

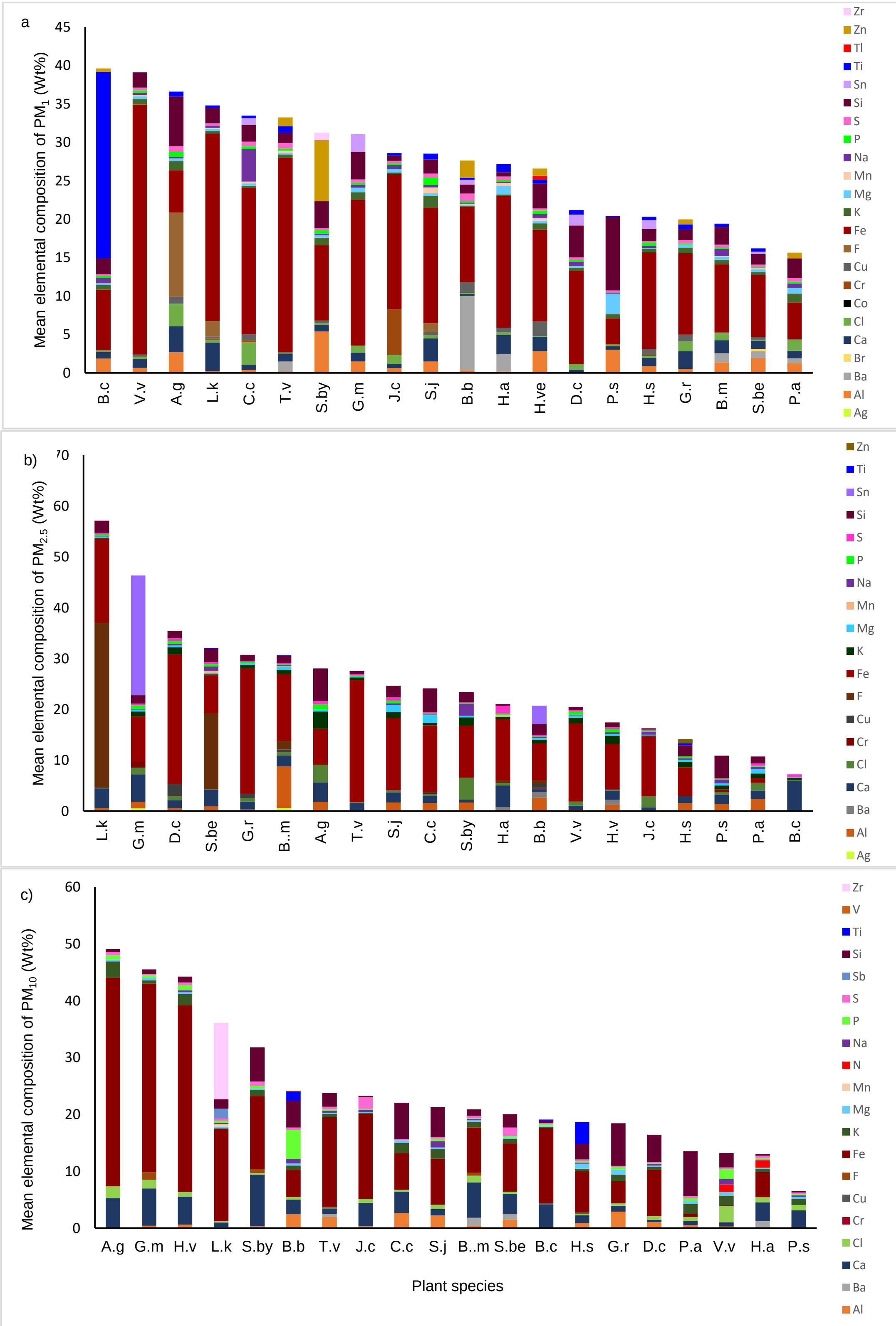


Figure 5-5 Quantity of different elements as their percentage weight (Wt%) in captured PM on leaves of plant species on the living wall located on Leek Road, Stoke-on-Trent: a) PM_1 b) $PM_{2.5}$ c) PM_{10}.
For explanation of the species codes see further Table 5.1. Note different scales on the y-axes.

surfaces of leaves reflect the influence of species-specific leaf characteristics on PM accumulation (Weerakkody *et al.,* 2017).

Evergreen conifers have frequently been recognized for more efficient PM capture compared to other species in tree- and canopy-based studies (Beckett *et al.*, 2000b; Chen *et al.*, 2017; Freer-Smith *et al.*, 2005; Tallis *et al.*, 2011); that the highest PM density found in this study was on the leaf needles of *J. chinensis* confirms the value of species with needle/needle-like leaves in living walls and probably in other vertical greenery systems. The relatively high LAI (Table 5.3) of conifers, as demonstrated here and Gower *et al.* (1995), further increases their PM removal potential (Fig. 5.2) suggesting their greater suitability as PM filters. Larger densities of all PM sizes were evident on leaves of all the smaller-leaved species - *V. vernicosa*, *B. buxifolia*, *B. x media*, *T. vulgaris* and *S. japonica* - and confirmed their high PM filtering potential as suggested in Weerakkody *et al.* (2017) (Chapter 4) with reference to PM generated from rail-traffic (Table 5.3). LAI values of these species were also high and enhanced their higher ability to remove PM.

Supporting the findings of Beckett *et al.* (2000), Hwang *et al.* (2011) and Weerakkody *et al.* (2017), most of the wide-leaved species - *P. amplexicaulis*, *B. cordifolia*, *L. kamtschatica*, *P. scolopendrium*, *H. sternii*, *H. americana* and *H. villosa* - showed comparatively lower densities in PM capture; however, Sæbø *et al.* (2012) did not find a correlation between leaf size and PM accumulation. This study indicates that individual leaf size is an important variable causing inter-species differences in PM capture and retention as higher PM accumulation occurred on smaller leaves and lower PM accumulation on wide leaves. Freer-Smith *et al.* (2005) and Leonard *et al.* (2016) made similar conclusions about the higher PM acquisition ability of smaller leaves. The differences in opinion on the impact of leaf size evident in the literature can possibly be attributed to the influence of other variables such as leaf shape and surface micromorphology exhibited by different species of plants (Weerakkody *et al.,* 2017).

Leaf hair/trichomes were identified as potentially helping in the accumulation of particulates on leaves by several authors (Beckett *et al.*, 2000b; Räsänen *et al.*, 2013; Weber *et al.*, 2014; Weerakkody *et al.*, 2017; Weerakkody *et al.,* 2018a) presumably through reducing re-suspension and increasing the effective surface area (Qiu *et al.*, 2009). Suggesting a similar effect, relatively high PM densities were found on hairy *S. byzantina* and *G. macrorrhizum* (Fig. 5.4) compared to other wide-leaved species. However, in contrast to Sæbø *et al.* (2012), there was no significant correlation between the density of leaf hair/trichomes with the density of PM captured - apart from with PM_{10} on adaxial surfaces. Perini *et al.* (2017) also made similar conclusions pointing out that there was no impact of leaf hairs on PM accumulation. Species-specific differences in the nature of hairs (size, length, texture, hydrophobicity and secretory/non-secretory) might have influenced their ability to help in trapping PM. Other variables such as leaf size, shape, and other micro-morphological features might also have enhanced or limited these abilities (e.g. PM accumulation on hairy leaves of *H. villosa* might have been limited by a larger leaf size) (Weerakkody *et al.*, 2018a). Rough leaf surfaces with dense ridges and grooves have frequently been cited as features likely to improve PM accumulation/retention (Barima *et al.*, 2014; Kardel *et al.*, 2012; Weerakkody *et al.*, 2017; Zhang *et* *al.*, 2017). However, in this study, the density of ridges or grooves did not show any significant correlation with PM densities apart from with $PM_{2.5}$ on abaxial surfaces. The diverse morphology of these characters (Fig. 5.4) (e.g. curved ridges forming broader deep grooves in *G. renardii*, densely arranged ridges forming narrower grooves in *P. amplexicaulis*, and parallel ridges/grooves in *D. cespitosa*) might be responsible for these results with some variants of these surface characteristics being better than others in PM capture/retention. Nevertheless, irrespective of the influence of other variables, stomatal density of the leaves was found to have a positive impact on PM accumulation by showing significant correlations with both adaxial and abaxial surface PM_{10} densities and $PM_{2.5}$ density on adaxial surfaces. Species with high stomatal conductance are known to increase PM deposition by increased diffusive deposition and by increasing deposition of hygroscopic PM (Beckett *et al*., 1998; Tong, 1991). Complex microtopography on leaf surfaces can increase PM deposition by slowdown the laminar boundary layer and by increasing surface roughness (Slinn, 1982); increased stomatal density may have a direct positive influence in this respect. However, differences in chemical affinities of PM to the epicuticular wax layers surrounding the stomatal regions, and phoretic effects closer to stomatal openings cause variable levels of PM accumulation in stomatal regions (Willmer, 1983).

Leonard *et al.* (2016) and Weerakkody *et al.* (2017) found relatively low PM accumulation on linear leaves. Of the four species with linear leaves used in this study, *J. chinensis* showed the highest PM accumulation whereas *D. cespitosa*, *C. caryophyllea* and *A. gramineus* showed moderate PM densities, which were higher than most of the wide-leaved species and lower than most of the smaller-leaved species. Differences in their leaf size, micro-morphology, surface texture and flexibility probably explain this variability. The smaller, rigid, leaf needles of *J. chinensis* and the grass and 'grass-like' leaves of *D. cespitosa*, *C. caryophyllea*, and *A. gramineus* probably create variable drag forces with wind currents (Gemba, 2007) resulting in differential turbulence around their leaves (Davidson and Wu, 1990). The readily-bending nature of elongated flexible grass-like leaves probably creates less turbulence compared to needles, resulting in increased PM accumulation on the latter.

The higher PM accumulation on adaxial leaf surfaces, compared to abaxial surfaces, reflects previous findings of Ottelé *et al.* (2010), Ram *et al.* (2012) and Weerakkody *et al.* (2017). The leaf orientation on vertical greenery systems is such that PM exposure on abaxial surfaces, which were facing the wall, can be much lower compared to adaxial surfaces facing the road. Therefore, lower densities of all PM sizes on abaxial surfaces could mainly be attributed to their reduced exposure level. Nevertheless, significant differences in PM accumulation on adaxial and abaxial leaves became less prevalent with decreasing particle diameter such that 17 out of 19 species had significantly higher levels of PM_{10} on their adaxial surfaces compared with 14 out of 19 species for $PM_{2.5}$ and 8 out of 19 species for PM_1 (Fig. 5.3). Particles deposited via sedimentation under gravity are most likely to be affected by leaf orientation, and this effect is greatest for particles of larger diameter (Slinn, 1982); these factors may explain the large differences in PM_{10} deposition.

5.1.5.2 Elemental composition

There was a close similarity of the elemental composition of PM found on leaves taken from the Leek Road living wall when compared with the elemental composition of known traffic-generated particles (Dong *et al.*, 2017; Lawrence *et al.*, 2013; Lin *et al.*, 2005; Sharma *et al.*, 2005). This similarity indicates that, as expected, a considerable proportion of PM filtered by the plants on the living wall were likely to be generated from road traffic. Nevertheless, we did not detect Pb, Pt or Ni in these PM, which were typically present in samples reported in the literature (Becker *et al.,* 2000; Lawrence *et al.*, 2013) ; the absence of Pb and Ni is probably due to changes in the composition of fuel additives (Lawrence *et al.*, 2013) e.g. Pb was banned in petrol in the UK in 2000 (Farmer *et al.*, 2002).

In addition to the influence of surrounding plant material, higher levels of organic and elemental C in vehicle exhaust (Kleeman *et al.*, 2000; Sharma *et al.*, 2005; Zhang *et al.*, 2017) might explain elevated C masses in PM in all size ranges. High C and O levels can also indicate presence of human carcinogenic polycyclic aromatic hydrocarbons (PAHs), one of the main toxic components of PM released from road traffic (mainly from fuel exhaust) (Velali *et al.*, 2016; Zhang *et al.*, 2017) and tyre wear (Boonyatumanond *et al.*, 2007). The high abundance of Fe is typical in traffic-generated PM as it can be derived from both exhaust and non-exhaust emissions. Both petrol and diesel exhaust PM are known to contain a considerable amount of Fe (Lin *et al.*, 2005; Manoli *et al.*, 2002; Wang *et al.*, 2003) and it is a predominant component in PM release from brake wear, tyre wear, road dust, and abrasives (e.g. brake linings) (Birmili *et al.* 2006; Cadle *et al.*, 1997; Cheng *et al.*, 2010; Pio *et al.*, 2013; Thorpe and Harrison, 2008). Trace quanitites of Cu, Ca, Ba, Mn, Ag, Co, Cr, Sb, Sr, Ti, V, and Zn can also be present in vehicle exhaust as functional groups of organic components (Chan, 2002; Lin *et al.*, 2005; Sharma *et al.*, 2005). In addition, Al, Ba, Ca, Co, Cr, Cu, K, Mg, Mn, Na, Sb, Sr and Zn can be attributed to non-exhaust particles released from brake and tyre wear (Pio *et al.*, 2013; Thorpe and Harrison, 2008; Weckwerth, 2001). Fine particles are mostly anthropogenic in origin and carry a variety of heavy metals which are toxic to humans and some are even carcinogens (e.g. Cr, Co, Cu, Mn) (Manalis *et al.*, 2005). Iron- rich particulates, falling in the range of $PM_{1,}$ are particularly hazardous due to their ability to cause oxidative brain damage potentially leading to important disease conditions such as Alzheimer’s and Parkinson’s (Maher *et al.*, 2013). In additon to PM originating from road traffic, PM_{10} containing Al, Ca, Na, Si, Cl, F, and N can originate from soil dust (Maher *et al.*, 2013). The presence of Tl (Thallium) has not been cited as a traffic-derived element in previous research and cement dust can be a potential source of PM_1 containing Tl (Brockhaus *et al.*, 1981).

5.1.5.3 Best species composition to use in living walls to filter near-road PM

Based on the outcome of this study, the use of coniferous species with leaf needles and smaller-leaved species with large LAIs can enhance the efficiency of PM filtering by living walls. Although stomatal density showed a higher impact on accumulating PM compared to other micro-morphological characters stomatal density can vary within a species depending on the environmental conditions (Beerling and Chaloner, 1993), and thus can be less useful to use as a direct indicator in species selection without a closer examination of the impact of local climate and growing conditions.

Based on specific observations in the present study and previous findings, species with hairy or rough leaves can still be useful in PM capture; avoiding species with larger leaves and smaller LAI would potentially enhance living wall performance.

5.1.6. Conclusion

Leaves of plant species on a living wall located along Leek Road, Stoke-on-Trent accumulated substantial amounts of PM_1, $PM_{2.5}$, and PM_{10}, demonstrating their potential to reduce PM pollutants in the atmosphere. Different plant species showed a varied potential to capture and retain particles; the highest PM levels found in leaf-needles of *J. chinenisis* were considerably higher in all particle size fractions. PM accumulation on all smaller-leaved species used in this study were considerably higher compared to most of the larger leaved species irrespective of their micro-morphological features. Accumulation of PM_1 did not show any significant correlation with the leaf micro-morphological features (leaf hair/trichomes, stomatal density, surface ridges and grooves) examined. Although there was no apparent pattern valid for all particle sizes, $PM_{2.5}$ and PM_{10} showed significant correlations with some of the micro-morphological features on at least one of the leaf surfaces (adaxial or abaxial). Of these characteristics, stomatal density showed more influence on $PM_{2.5}$ and PM_{10} accumulation compared to other characters. Irrespective of the absence of statistical correlations when all the species were considered, higher levels of PM on leaves of *S*. byzantina and *G. macrorrhizum* suggested some positive influence of leaf hairs in PM accumulation. Morphological variation within each character probably had an impact on their influence in PM accumulation (i.e. length and shape of hair, depth and shape of ridges or grooves). The elemental composition of accumulated particles showed a strong similarity with the chemical composition of traffic-generated particles.

5.2 Exploring a suitable planting design for a living wall for trapping PM effectively.

In addition to the choice of species and type of vegetation, planting design was predicted to be important in the use of vegetation to increase PM dry deposition (Freer-Smith *et al.*, 2003; Madders and Lawrence, 1981). Dynamics in topography and porosity of vegetation directly influence surface resistance and surrounding airflow patterns, hence producing different levels of turbulence resulting in different PM deposition rates (Abhijith *et al.*, 2017; Davidson and Wu, 1990; Gallagher *et al.*, 2012; Janhall, 2015; Slinn, 1982). In order to use living walls as PM traps, understanding effective design features is crucial. This can be studied by creating topographical variation in wall vegetation using a single plant species (to standardise the impact of species characteristics). This impact was evaluated using *Buxus sempervirens* L., one of the species recorded with higher PM accumulation levels in Weerakkody *et al.* (2017) (Chapter 4).

5.2.2. Methodology

The experimentally manipulable living wall system used in the previous experiment (Section 5.1) was also used in this experiment. Plants of *B. sempervirens* were purchased from the Hedge Nursery, UK in two commercially available heights 10-20 cm and 30-40 cm (referred to as 'short' and 'tall' plants respectively). The number of planting designs were limited to two due to the relatively compact dimensions of the experimental wall (3.98 m x 2.09 m). Equal halves of the road-facing side of the living wall (through its mid-vertical axis) were planted using different planting designs simultaneously, to standardise the influence of weather on both the designs. One half of the wall was planted as a random design to create a variable (heterogeneous) topography by having plants of both heights in a random order. The other half was planted as a cluster design having two distinct groups of plants, each group with plants all of the same height to create two areas with homogeneous topography (one height at the top and the other at the bottom of the wall) (Fig. 5.6). This experiment was replicated four times, and in each of the replicates, the allotted side for each planting design and the arrangement of clusters (top or bottom) were changed in a random order (Fig. 5.6). The working hypotheses for this study were:

1. If increased turbulence caused by topographical heterogeneity increases PM deposition, plants allocated to the 'random' design should collect more PM than plants allocated to the more homogeneous 'cluster' design.

2. Increased PM levels are predicted to occur on leaves of plants at the interface (edge) between tall and short plants in the cluster design due to increased turbulence.

Once the plants were installed for each experimental trial, they were washed using a watering hose with a pressure head to remove existing particles, and 20 leaves from the short plants and 20 leaves from the tall plants were randomly collected from each design (a total of 80 leaves) to quantify the baseline PM levels. Sample storage and transfer followed the same protocol used in Section 5.1. Subsequently, baseline PM (PM_1, $PM_{2.5}$ and PM_{10}) levels on the leaves were quantified using the ESEM/imageJ approach used for *B. sempervirens* (for smaller-leaves) in Weerakkody *et al.* (2017) (detailed in Chapters 2 and 4). Any significant variation in baseline PM levels, between tall and short plants and between two planting designs were identified using student's t-test following application of the Shapiro-Wilk test to confirm normality.

Figure 5-6 Images showing two random arrangements of two planting designs of *Buxus sempervirens* used in the living wall located along Leek Road, Stoke-on-Trent.

Subsequently, plants were left untouched and exposed to pollution (mainly generated from traffic on Leek Road, see Section 5.1.3) for five consecutive rain-free days. Before the first replicate, in order to identify any potential edge-effect at the interface between short and tall plant clusters in cluster design, leaves were randomly collected from the middle and edges of the clusters (20 leaves from each category). PM densities on these leaves were quantified using the ESEM/ImageJ approach. Any edge-effect at the interface between short and tall plant clusters was identified by comparing PM densities on the leaves taken from the middle and edges of the clusters, using Student's t-test. This was done to adjust for any influence from a potential edge effect when sampling from the cluster design.

Forty leaves from each planting design, i.e. 20 leaves from each height per design, were sampled to evaluate any differential impact of topography. Sample storage and transfer followed the same approach given in the previous experiment (Section 5.1) and Weerakkody *et al.* (2017) (Chapter 4). The density of PM_1, $PM_{2.5}$, and PM_{10} on leaves were estimated using the ESEM/ImageJ approach (Weerakkody *et al.*, 2017 and Section 5.1). PM accumulation on leaves in each experimental trial were quantified following exactly the same exposure time and the same experimental procedure. At the beginning of each experiment (after plants were re-allocated for the experiment) plants were washed with a watering hose to remove existing PM trapped in the previous trial and any variability in baseline PM levels (even after wash-off) were identified using student's t-test after normality testing.

At the completion of all four experimental trials, any significant differences in densities of PM_1, $PM_{2.5}$, and PM_{10} on the leaves taken from different designs were identified using GLMM. As these leaves were sampled in different experimental trials, time was included as a random factor in the model to avoid any variable influence of weather. Any variability in PM accumulation on the leaves of plants with different heights, within the same design, was also identified using GLMM using time as a factor.

5.2.3 Results

PM densities on the leaves taken from the middle of the clusters and from the edges were not significantly different from each other for any particle size fraction (Fig. 5.7) and hence, the sampling location within the cluster was random in all the sampling attempts (i.e. no guard rows were used when selecting leaves for sampling).

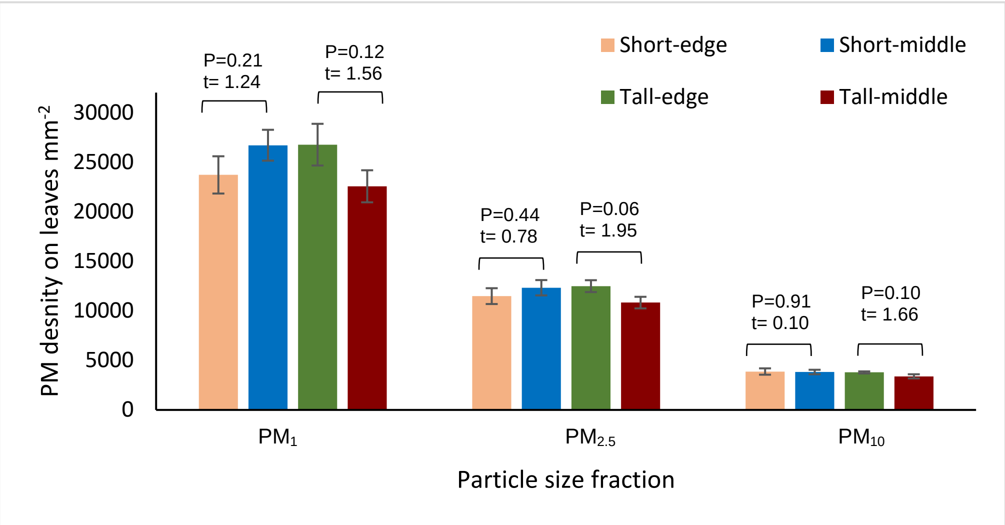


Figure 5-7 The mean PM density ± SE on leaves taken from edges and middle of the plant clusters (plants of the same height were grouped together to form a homogeneous topography) of *B. sempervirens* in the living wall located along Leek Road, Stoke-on-Trent.

Short-edge and short-middle: leaves taken from edges and middle of the short plant clusters; tall-edge and tall-middle: leaves taken from edges and middle of the short plant clusters.

There were no significant differences in baseline PM densities after washing the leaves off (Appendix 4), and hence they were not included in the analyses following the 5-day exposure periods. The GLMM revealed significantly higher densities of PM_1, $PM_{2.5}$, and PM_{10} on the leaves sampled from the random planting design compared to the leaves taken from the cluster design, for both short (Fig. 5.8) and tall (Fig. 5.9) plants. PM densities on the leaves of plants with different heights, within the same design, were not significantly different for any PM size fraction (Fig. 5.10 and 5.11).

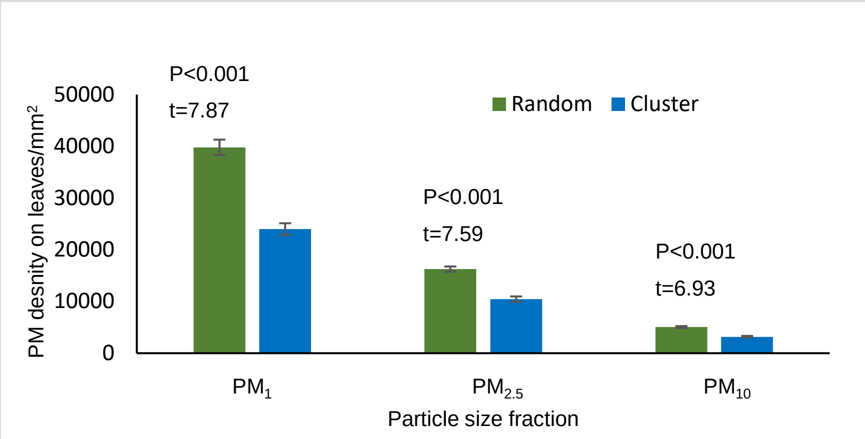


Figure 5-8 The mean PM density ± SE on leaves of short *B. sempervirens* plants used in the living wall located along Leek Road, Stoke-on-Trent in planting designs where plants of the same height were grouped together to form a homogeneous topography 'Cluster' or interspersed with tall plants to form a heterogeneous topography 'Random'.

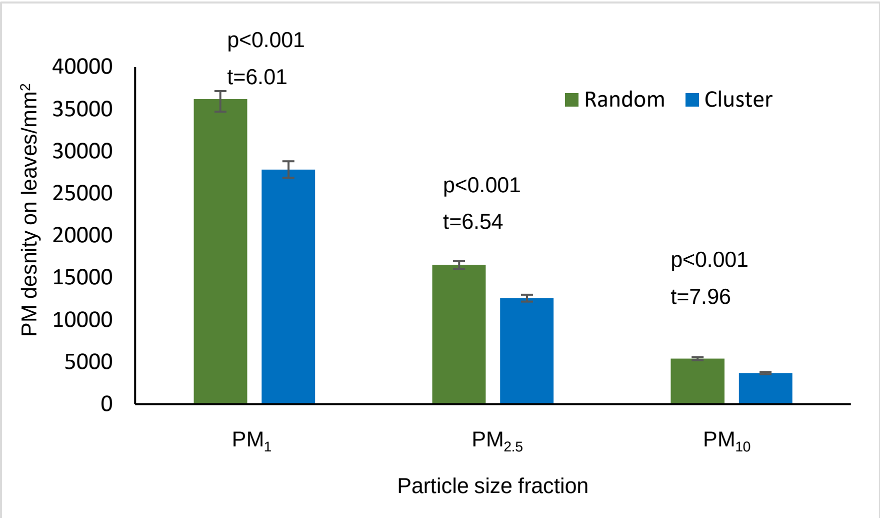


Figure 5-9 The mean PM density ± SE on leaves of tall *B. sempervirens* plants used in the living wall located along Leek Road, Stoke-on-Trent in planting designs where plants of the same height were grouped together to form a homogeneous topography 'Cluster' or interspersed with short plants to form a heterogeneous topography 'Random'.

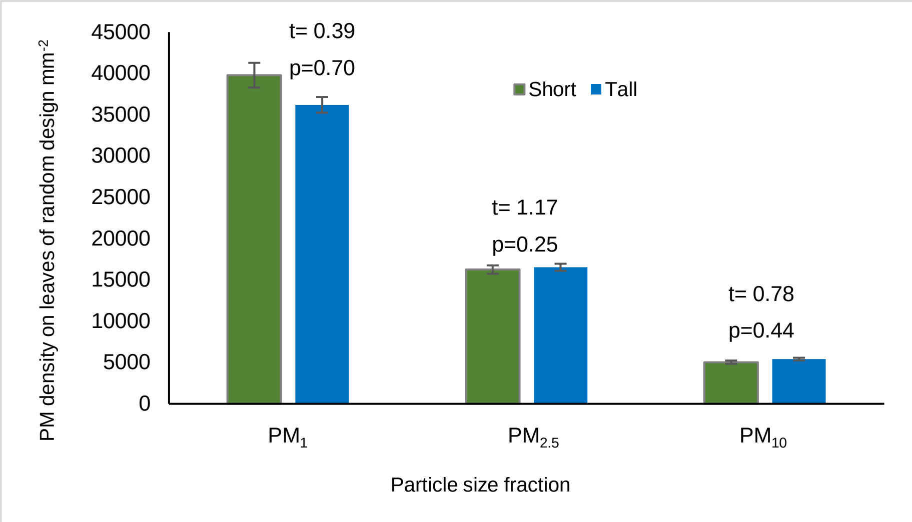


Figure 5-10 The mean PM density ± SE on leaves of short and tall plants of *B. sempervirens* plants used in the random planting design with heterogeneous topography in the living wall located along Leek Road, Stoke-on-Trent.

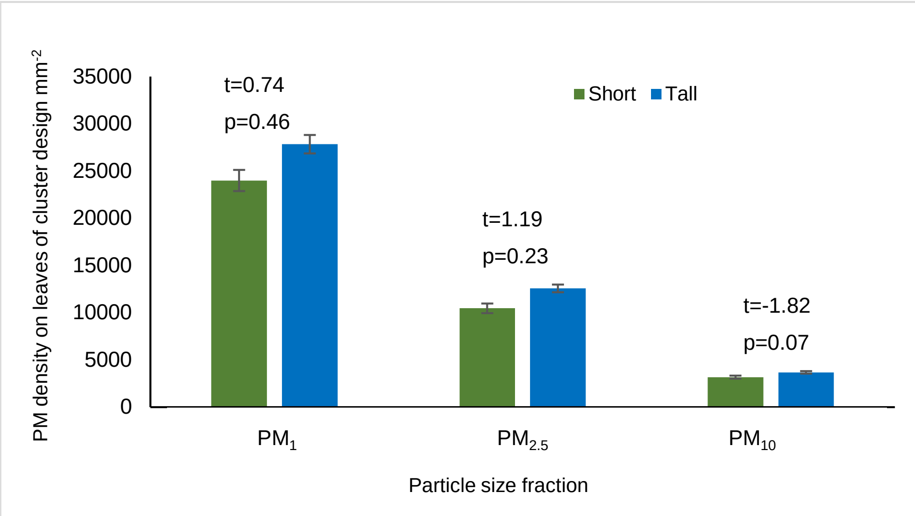


Figure 5-11 The mean PM density ± SE on leaves of short and tall plants of *B. sempervirens* plants used in the 'cluster' planting design with homogeneous topography in the living wall located along Leek Road, Stoke-on-Trent.

5.2.4 Discussion

Significantly higher PM densities were found on leaves of the plants in the random design compared to those in the cluster design, for both tall and short plants, and demonstrated the higher efficacy of a random planting design to capture and retain particulates. The random arrangement of plants with two different heights created an uneven wall-surface with more topographical variation, whereas the cluster design had a more level surface in each cluster and, hence, less topographical variation. Therefore, the random design has probably created a relatively more complex deposition surface, with a greater surface roughness, which consequently increased turbulence in the surrounding airflow (Manning and Feder, 1980; Slinn, 1982) resulting in higher PM accumulation on the leaves

(Davidson and Wu, 1990; Petroff *et al.*, 2008b; Slinn, 1982). Since there was no variability in PM accumulation on the plants with different heights within the random design, use of plants in different heights is unlikely to affect the PM capture ability of each other by shielding. Similarly, in the cluster design, absence of any variability between short and tall plant clusters suggest the importance of overall topography in PM accumulation regardless of their height.

Whilst higher PM levels were predicted to occur on leaves collected from the edges of the clusters compared to the middle of the clusters due to greater turbulence at the interface between short and tall plantings, no such difference was detected. The absence of any significant variability between leaves on the edges and the middle of clusters (Fig. 5.7) indicates that there was insufficient turbulence created at the interface and, hence, no differential accumulation of PM. This may have resulted from a boundary layer effect or back wind pressure, similar to that found with pesticide drift deposition at the base of hedges (Davis *et al.*, 1994) or be attributable to the relatively small dimensions of these plant clusters.

The results of this experiment suggest an important impact of planting design on the performance of living wall species in the capture of PM; for a purpose-built living wall to reduce atmospheric PM, use of a random design or a similar design with high topographical variation would be beneficial.

5.2.5 Conclusion

A planting design with topographical heterogeneity showed a higher positive impact on the ability of plants to capture and retain PM compared to a planting design with homogeneous topography. Therefore, use of a planting design with complex topography would enhance the PM reduction potential of living walls.

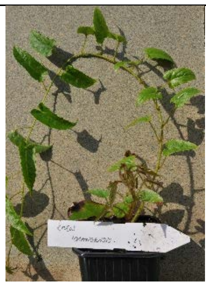
5.3 The potential use of scrambling *Rubus* species in living walls to immobilise PM pollutants

A recent trend has been observed around the use of different forms of vegetation on living walls. As the leaf orientation of scrambling species is pointed more towards the ground, and at a lower angle closer to the wall, their aerodynamic behaviour due to surrounding air flow may have a differential impact on PM deposition compared to those species studied in previous experiments. Therefore, the present experiment used scrambling species, belonging to the genus *Rubus L*. which are commonly propagating to use in VGSs, in a living wall system to assess their potential of PM immobilisation in order to understand their impact in this respect. Most scrambling species belonging to the genus *Rubus* have complex micromorphology and over 2,000 species, both evergreen and deciduous foliage, have been described so far (Sell and Murrell, 2014). By using seven plant species belonging to the same genus, inter-species variation in leaf PM accumulation within a single genus was also explored.

5.3.1 Site and species description

An existing modular living wall system (Mobilane ®) located on Staffordshire University campus was replanted with seven *Rubus spp*. (Table 5.4) in a random order (Fig. 5.12). *Rubus spp*. used in this study were specially propagated for this experiment by Hillier Nurseries, Southampton, UK, in order to assess their potential use in vertical greenery systems. The wall was north facing and the closest pollution source to this wall was a busy traffic road, College Road, 50 m distance from the wall (the installation was side-on to the road). This living wall comprised eight modular planting panels each containing 40 planting holes.

Table 5.4 Plant species used in this experiment, their images and brief species description [c]

Species name	Image	Species and leaf description
Rubus gongshanensis TT Yu and LT Lu		A semi evergreen shrub. Large, hairy, spiny, oval leaves with a serrated margin.
Rubus ichangensis Hemsl. & Kuntze		A deciduous shrub. Large, glabrous, narrowly ovate leaves with a slightly serrated margin.

Rubus lambertianus Ser.		A semi-evergreen shrub. Large, glossy, broadly ovate (triangular) leaves with a narrowly pointed tip and a slightly serrated margin.
Rubus nepalensis (Hook. f.) O. Kuntze		An evergreen, prone shrub. Medium, glossy, trifoliate leaves with a serrate margin.
Rubus rolfei J.E.Vidal		An evergreen prone spreading shrub. Large, deeply wrinkled, lobed leaves with a rough leaf surface and a serrated margin.
Rubus alceifolius forma. purpureus.		Large, slightly wrinkled, lobed leaves with a serrated margin.
Rubus yiwanus Sichuan		An evergreen prone shrub. Large, slightly wrinkled, broadly ovate leaves with a serrated margin.

[c] Leaf shape descriptions based on Hickey and King (2000) and Beentje (2012)

5.3.1 Material and methods

5.3.1.1. Analysing inter-species variation in PM capture

Leaf sampling and storage followed the same approach used in Weerakkody *et al.* (2017) (Chapter 4). Twenty leaves from each species were sampled immediately after replanting the wall to evaluate their baseline PM levels on the leaves and were evaluated using an ESEM/imageJ approach (Section 5.1). Plants were then left undisturbed for three weeks allowing for continuous exposure to weather changes. Plants on the top-most row, side rows, and three rows from the bottom were excluded from the sampling to avoid any potential edge-effect and soil contamination from the ground.

Figure 5-12 The experimental living wall located on Staffordshire University's campus Stoke-on-Trent, UK, planted with *Rubus spp.* (see further Table 5.4).

Twenty leaves from each species were randomly sampled on five different days during July and August 2016 under dry weather conditions. All the species were equally sampled on each sampling occasion as four leaves from each species per day. Density of PM_1, $PM_{2.5}$, and PM_{10} on both adaxial and abaxial surfaces of the leaves were estimated using the ESEM/imageJ approach (Weerakkody *et al.*, 2017, Chapter 4). Any inter-species variation in total PM density and, separately, on the adaxial and abaxial surfaces of the leaves, were identified using mixed-Anova in a Generalized Linear Mixed-effect Model (GLMM) following the same statistical analysis approach given in section 5.1. Any significant difference in PM density between adaxial and abaxial surfaces of the same species of plant was identified using Student's t-test after the Shapiro-Wilk test to confirm normality. The Leaf Area Index (LAI) was measured to estimate the total leaf area of the plants using the same approach and formula used in Weerakkody *et al.*, 2017 (Chapter 4). Inter-species variations in total PM capture levels (incorporating LAI) was identified using a mixed-Anova in GLMM including time as a factor.

5.3.1.3. Evaluating the correlation between leaf micromorphology and PM accumulation

Following visualization of particulates, a further set of micrographs were taken to image micro-morphological features from ten random leaves previously scanned to quantify particles. As surface structures such as leaf hairs and trichomes are substantially larger than particulates, leaf surfaces were scanned at 60 x, 250 x; in order to image leaf stomata, grooves and ridges, leaves were scanned at 300 x and 450 x (Fig. 5.13). As these characters were unevenly distributed on leaves, focus settings were changed accordingly and ten random micrographs per character for each species were taken. Ridges, grooves and elongated trichomes on leaves were estimated as the percentage area covered by the character with reference to total area of leaf surface using ArcGIS (ArcMap

10.4 © 2015 ESRI) and the number of simple leaf hairs/trichomes and stomata on leaves were manually quantified and expressed as number of characters per unit area (1 mm^2) (Chapter 2, Weerakkody *et al.,* 2018b/Section 5.1.3.5). The epidermal protrusions of the leaves except for dense elongated trichomes, were classified as hairs. Any significant relationship between PM densities on leaves and these leaf micro-morphological characters were identified using GLMM including species as a random factor; as these characters came from seven different species, plant species was included as a random factor in this model. Based on an uneven distribution of leaf morphological characters on the adaxial and abaxial surfaces, they were analysed separately.

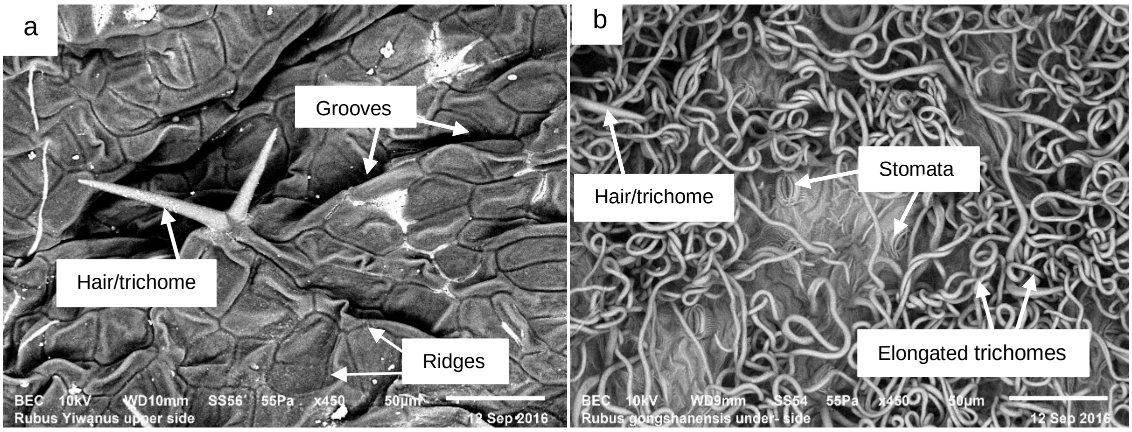


Figure 5-13 ESEM micrographs showing micromorphological characters examined on the a) adaxial and b) abaxial surfaces of the leaves of *Rubus spp.*

5.3.2. Results

5.3.2.1. Inter-species variation in PM capture

As there were no significant differences in baseline PM densities, they were not included in the analysis (Appendix 4). When all seven species of plants were considered, on average, 2.51 ± 0.29 x 10^6 of PM_1, 1.3 ± 0.18 x 10^5 of $PM_{2.5}$, and 2.68 ± 0.39 x 10^4 of PM_{10} were estimated to have been captured on 100 cm^2 of the living wall. Analysis of micrographs revealed a significant inter-species variation between *Rubus sp*. in both PM densities (PM_1: $F= 7.42$, $p < 0.001$; $PM_{2.5}$: $F= 6.342$, $p < 0.001$; PM_{10}: $F= 5.933$, $p < 0.001$) (Table 5.5) on leaves and in their overall ability to capture and retain PM when LAI (Table 5.6) was incorporated (PM_1: $F= 9.372$, $p < 0.001$; $PM_{2.5}$: $F= 7.035$, $p < 0.001$; PM_{10}: $F= 6.654$, $p < 0.001$) (Fig. 5.14). The highest mean density of all PM sizes (PM_1:1.78 ± 0.15 x 10^4; $PM_{2.5}$: 9.98 ± 1.21 x 10^2; PM_{10}: 2.18 ± 0.30 x 10^2) on the leaves and the highest ability to capture PM for all PM size fractions when LAI (Table 5.6) was incorporated (PM_1:3.37 ± 0.29 x 10^6; $PM_{2.5}$: 1.89 ± 1.21 x 10^5; PM_{10}: 4.13 ± 0.57 x 10^4) were shown by *R. rolfei*. These levels were significantly higher than for *R. lambertianus, R. ichangensis*, and *R. alceifolius* in all the size ranges (Table 5.5 and Fig. 5.14). The second and third highest PM densities on leaves and the overall ability to capture PM were both shown by *R. yiwanus* and *R. nepalensis,* and these levels were not significantly different from the highest R. rolfei. or from fourth highest *R. gongshanensis.*

The comparison of PM densities on the adaxial and abaxial surfaces of leaves showed that PM density on the adaxial surfaces were always higher than on the abaxial surfaces, and in *R. rolfei, R. yiwanus and R. gongshanensis* these differences were statistically significant (Fig. 5.15). The densities of all PM sizes on the adaxial surfaces were significantly different between species whereas, on the abaxial surfaces, $PM_{2.5}$ ($F= 2.10$, $p= 0.057$) and PM_{10} ($F= 1.33$, $p= 0.24$) did not show any significant variation between species. The highest mean on the adaxial surfaces of leaves were found in *R. rolfei* for all PM size fractions (PM_1:$1.59 \pm 0.13 \times 10^4$; $PM_{2.5}$: $9.15 \pm 1.23 \times 10^2$; PM_{10}: $1.77 \pm 0.19 \times 10^2$). On the abaxial surfaces of the leaves, the highest mean density of PM_1 was found on leaves of *R. nepalensis* ($6.23 \pm 0.43 \times 10^3$).

Table 5.5 Mean PM densities ± 1SE on leaves of *Rubus* species in the living wall used in this experiment [d]

Species	Mean PM_1 density on leaves ($\times 10^2$) ± SE	Group assigned by Tukey's HSD test	Mean $PM_{2.5}$ density on leaves ($\times 10^2$) ± SE	Group assigned by Tukey's HSD test	Mean PM_{10} density on leaves ($\times 10^2$) ± SE	Group assigned by Tukey's HSD test
R. gongshanensis	128.4 ± 15.35	ab	6.17 ± 1.05	bc	1.43 ± 0.26	ab
R. ichangensis	101.6 ± 7.97	bc	4.11 ± 0.40	c	0.86 ± 0.09	b
R. lambertianus	103.6 ± 13.48	bc	4.37 ± 0.28	c	0.78 ± 0.17	b
R. nepalensis	139.5 ± 9.95	ab	6.76. ± 1.10	abc	1.47 ± 0.22	ab
R. rolfei	178.0 ± 15.29	a	9.98 ± 1.21	a	2.18 ± 0.30	a
R. alceifolius	72.7 ± 4.58	c	4.88 ± 0.64	bc	0.97 ± 0.09	b
R. yiwanus	145.6 ± 17.18	ab	8.39 ± 0.87	ab	1.49 ± 0.13	ab

[d] PM density on leaves of species assigned with the same letter in each PM size range, were not significantly different within their respective PM size category (Tukey's HSD post hoc test, p> 0.05).

5.3.2.2 Correlation between leaf micromorphology and PM densities

The nature and distribution of micro-morphological characters on leaves varied between different species of plants, and between adaxial and abaxial surfaces of the same species (Table 5.6 and Fig. 5.16). The abaxial surfaces of the leaves of the majority of species were covered with dense elongated trichromes which obscured the leaf surfaces; as a result, ridges and grooves on the abaxial surfaces were not quantifiable (Fig. 5.16). On the adaxial surfaces of the leaves, the density of $PM_{2.5}$ showed positive relationships with surface grooves ($t = 5.52$ and $p < 0.0001$) and ridges ($t = 3.77$ and $p = 0.0004$), whilst the density of PM_1 showed a positive relationship only with surface grooves ($t = 3.65$, $p = 0.0005$). The density of PM_{10} on the adaxial surfaces did not show any significant relationship with any of the characters examined (hairs, grooves, or ridges) (Appendix 3). Similarly, even on the abaxial surfaces, the density of PM_{10} did not show any relationship with any of the leaf micro-morphological characters analysed (hair, stomata, elongated trichomes) (Appendix 3). Nevertheless, the density of PM_1 ($t = 4.35$ and $p = 0.0001$) showed a positive relationship with the density of leaf hairs, whereas the density of elongated hairs showed a negative relationship with both $PM_{2.5}$ ($t = - 2.41$ and $p = 0.018$) and PM_1 ($t = - 6.17$ and $p < 0.0001$).

Table 5.6 Mean leaf size ±1SE, LAI ±1SE, and mean quantities ±1SE of micro-morphological characters of the leaves of plant species used in this experiment

Species	Leaf area±SE (cm^2)	LAI±SE	Micromorphology on adaxial surface of the leaves ±SE			Micromorphology on abaxial surface of the leaves ±SE		
			Density of Hair (mm^{-2})	Grooves %	Ridges %	Elongated trichomes %	Stomatal density (mm^{-2})	Density of Hair (mm^{-2})
R .gongshanensis	57.3 ± 0.88	2.2 ± 0.09	39.3 ± 12.46	35.7 ± 1.57	37.1 ± 1.31	41.4 ± 1.85	681.4 ± 12.37	28.4 ± 1.76
R. ichangensis	34.3 ± 1.33	1.8 ± 0.11	5.2 ± 0.66	1.0 ± 0.39	3.3 ± 0.83	0.0	207.3 ± 13.17	10.3 ± 0.22
R. lambertianus	28.2 ± 1.43	1.6 ± 0.09	13.4 ± 0.48	39.4 ± 1.61	41.6 ± 1.57	0.0	335.4 ± 11.01	5.1 ± 0.56
R .nepalensis	9.7 ± 0.50	2.3 ± 0.12	21.6 ± 1.19	10.2 ± 1.37	17.2 ± 1.77	0.0	543.3 ± 13.28	24.1 ± 0.65
R. rolfei	26.4 ± 0.96	1.9 ± 0.11	5.1 ± 1.26	54.7 ± 1.86	47.8 ± 1.41	48.4 ± 1.28	372.4 ± 10.62	30.3 ± 1.08
R. alceifolius	67.9 ± 1.41	2.2 ± 0.11	19.8 ± 1.04	37.4 ± 1.41	40.4 ± 1.41	17.6 ± 1.47	672.1 ± 13.9	20.7 ± 1.36
R. yiwanus	68.9 ± 1.26	2.2 ± 0.12	24.2 ± 1.09	47.4 ± 1.19	42.4 ± 1.19	37.4 ± 1.32	672.1 ± 13.9	31.0 ± 1.35

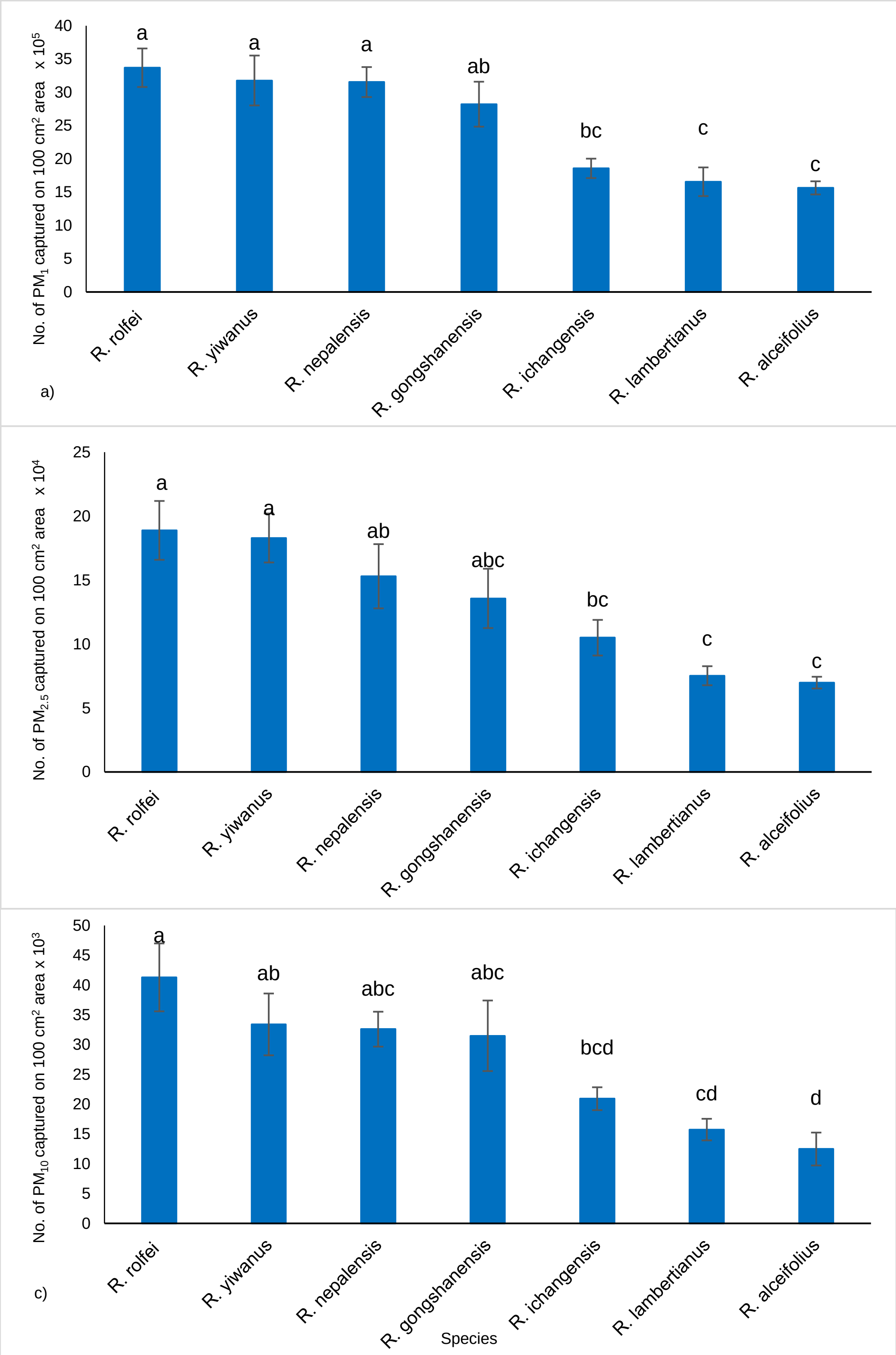


Figure 5-14 The total estimated mean number ±1 SE of PM captured on leaves in a 100 cm^2 area of living wall (incorporating the LAI) by different species of *Rubus* plants on the experimental living wall on Staffordshire University's Campus: a) PM_1, b) $PM_{2.5}$, and c) PM_{10}.

Data are arranged in descending order of PM accumulation for each size range. Note the different scales on the y-axes. Species marked with the same English letters were not significantly different within each particle size category (Tukey's HSD post hoc test, $p > 0.05$).

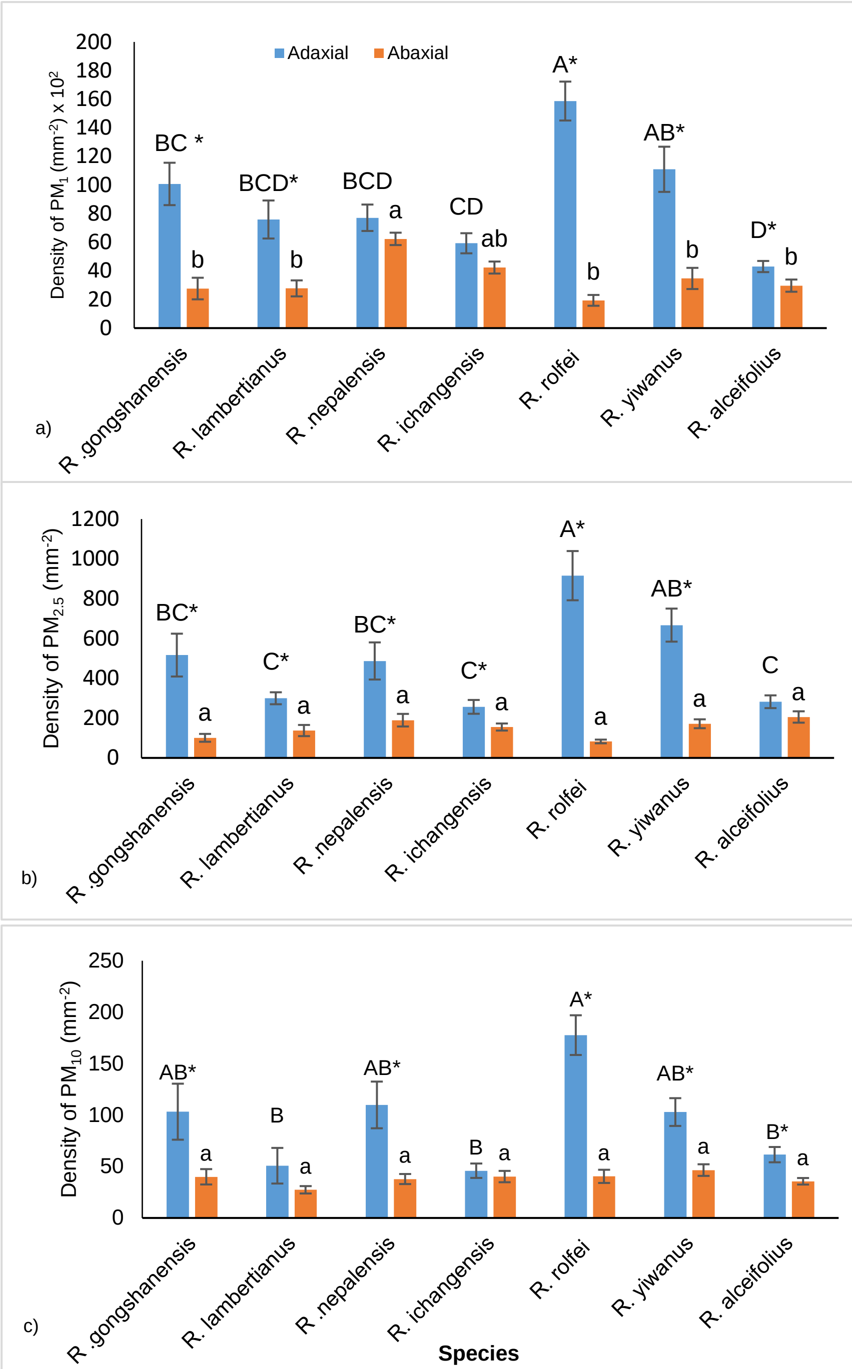


Figure 5-15 The mean PM density ±1 SE on 1 mm^2 of the adaxial and abaxial surfaces of the leaves of plant species on the living wall located on Staffordshire University's Campus (PM densities are given in different scales): a) PM_1, b) $PM_{2.5}$, and c) PM_{10}.

Plant species marked with the same letters are not significantly different from each other ($p > 0.05$). **Upper case letters** - variation between adaxial surfaces of the leaves; **lower case letters** - variation between abaxial surfaces of the leaves. Note different scales on the y-axes. ***** indicates that the PM densities on adaxial and abaxial surfaces were significantly different.

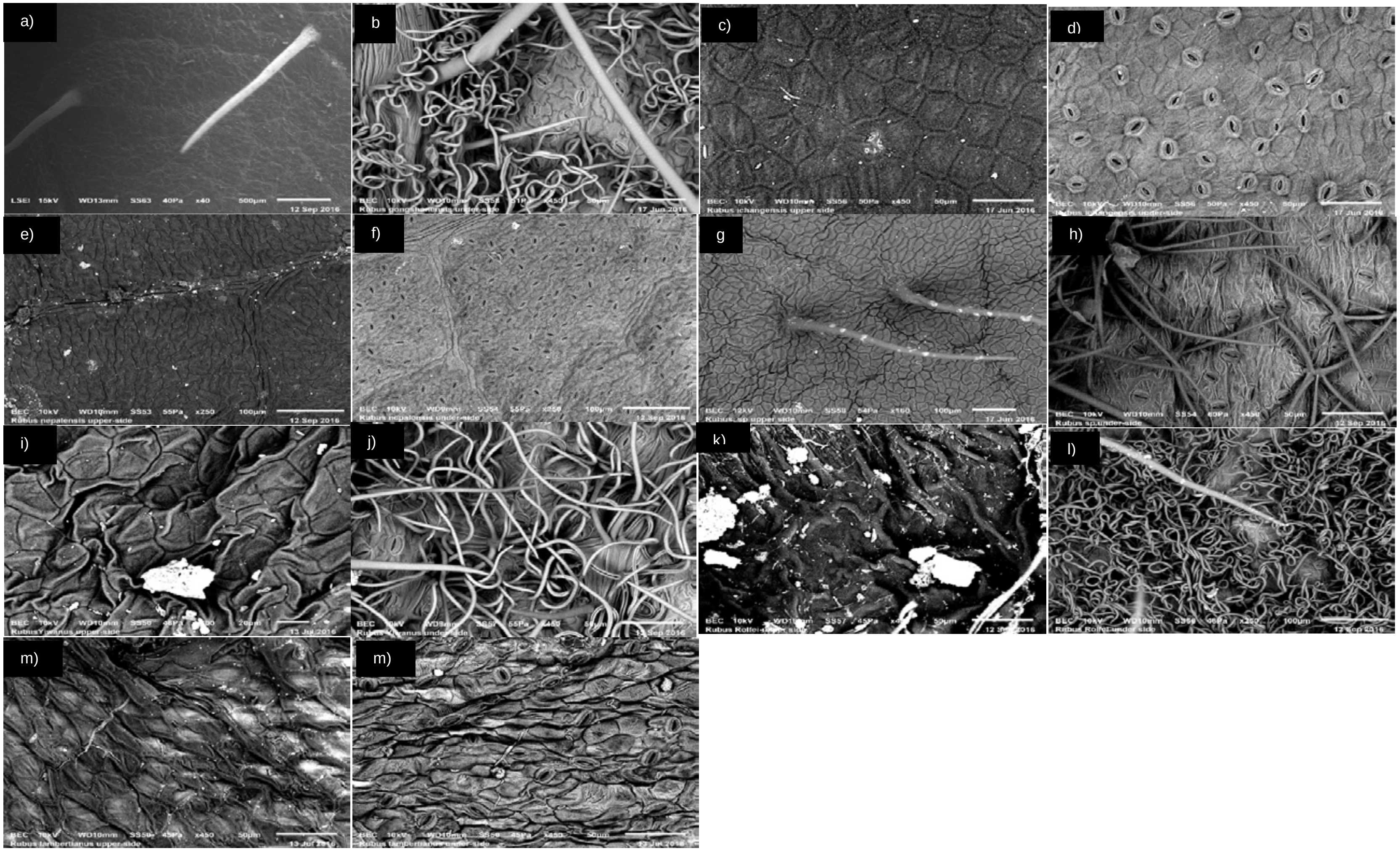


Figure 5-16 ESEM micrographs showing micro-morphological characteristics on the a) adaxial (x40) and b) abaxial surfaces (x450) of *R. gongshanensis*, c) adaxial (x450) and d) abaxial (x450) surfaces of *R. ichangensis*, e) adaxial (x250) and f) abaxial (x250) surfaces of *R. nepalensis*, g) adaxial (x160) and h) abaxial (x450) surfaces of *R. alceifolius*, i) adaxial (x400) and j) abaxial (x450) surfaces of *R. yiwanus,* k) adaxial (x450) and l) abaxial (x250) surfaces of *R. rolfei*, and m) adaxial (x450) and n) abaxial (x450) surfaces of *R. lambertianus*

5.3.3. Discussion

In agreement with the findings of Sæbø *et al.* (2012) on the possibility of having differential PM accumulation on different species in the same genus (*Tilia cordata* and *Tilia×europaea*), species in the genus *Rubus* used in the present study, showed a considerable variation in both PM densities and overall PM accumulation (incorporating LAI). Although the average PM capture by plants on this wall was lower compared to the other living walls studied relating to road traffic in Stoke-on-Trent (122.08 ± 6.9 x 10^7 PM_1, 8.24 ± 0.72 x 10^7 $PM_{2.5}$ and 4.45 ± 0.33 x 10^7 PM_{10} by 100 cm^2 area) (Section 5.1.4.1), they still showed a notable potential for capturing and retaining PM (2.51 ± 0.29 x 10^6 of PM_1, 1.3 ± 0.18 x 10^5 of $PM_{2.5}$ and 2.68 ± 0.39 x 10^4 by 100 cm^2 area). In addition to the differences in species used in the two walls, differences in pollutant concentration in the atmosphere surrounding the walls (caused by differences in distance to the source of pollutants and in traffic density) may be responsible for differences in PM removal observed (Nowak *et al.*, 2013; Nowak *et al.*, 2006; Sæbø *et al.*, 2012; Weber *et al.*, 2014). The distance between the living wall and the traffic-road is relatively large in the present experiment and the traffic density of College Road is lower compared to Leek Road. The orientation of this wall might also have a negative impact on PM capture, as the wall was not directly facing the road as in the previous case (Section 5.1.3). In agreement with the results of Freer-Smith *et al.* (2005), Ottele *et al.* (2010), Weerakkody *et al.* (2017) (Chapter 4) and results of the previous experiment (Weerakkody *et al.,* 2018b/Section 5.1), the total number of particles captured on leaves increased with decreasing particle diameter reflecting the different aerodynamics of different PM sizes (Davidson and Wu, 1990).

Similar to the findings in Ottelé, *et al.* (2010) and Ram *et al.* (2014) and in other experiments in this research (Weerakkody *et al.*, 2017 / Chapter 4 and Section 5.1) higher PM densities were found on the adaxial surfaces of the leaves compared to the abaxial surfaces, which can be explained by high PM exposure on the adaxial surfaces of the leaves due to the leaf orientation in VGS.

Leaf sizes between the *Rubus* species did not vary greatly (apart from slightly smaller leaves in *R. nepalensis*), and hence variabilities in PM densities on leaves can probably be attributed mostly to their micro-morphological variations and/or leaf-shapes. Although leaf hairs and stomatal density are known to have a positive impact on PM accumulation on leaves (Kardel *et al.*, 2012; Ram *et al.*, 2012; Räsänen *et al.*, 2013; Weerakkody *et al.*, 2017), densities of PM sizes analysed in the present experiment did not show any correlation with density of leaf hair or stomata, which can therefore probably be attributed to the influence of other variables. However, leaves of *R. gongshanensis,* which had the highest density of hair also had high PM densities. Indeed, the densities of PM_1 and PM_{10} on *R. gongshanensis* were not significantly different from those on the leaves *of R. rolfei* (the species with the highest accumulations) and the second highest density of $PM_{2.5}$ (found on the leaves of *R. yiwanus*). In the previous experiment (Section 5.1.4.2), the stomatal density of leaves showed a higher impact on PM accumulation compared to other micro-morphological features; however, the abaxial surfaces of the majority of the species used in this study (*R. rolfei, R. yiwanus, R. alceifolius* and *R. gongshanensis*) were mostly covered by elongated trichomes which formed a complex web (Fig. 5.16) which might have limited the potential influence of stomata on PM accumulation. Since dense elongated trichomes can only be found in certain species of plants (e.g. only in 4 *Rubus spp*. out of 40 species studied in this project), sometimes as an anti-herbivore defence adaptation (Karley *et al.*, 2016), their impact on PM accumulation has not been studied or discussed in the literature. Interestingly, the results of this experiment revealed a negative correlation between elongated trichomes and PM accumulation; dense elongated hair might have shielded the leaf surfaces from external contaminants. Reflecting the findings of previous research (Barima *et al.*, 2014; Weerakkody *et al.*, 2017; Zhang, *et al.*, 2017) surface roughness of the leaves showed a positive influence on accumulating smaller particle sizes (both ridges and grooves on accumulating $PM_{2.5}$ and grooves on PM_1). The highest densities of all PM sizes (on the adaxial surfaces of leaves of *R. rolfei* and the second highest PM densities on the adaxial surfaces of the leaves of *R. yiwanus*) can probably be attributed to their rough leaf surfaces, with a higher number of ridges and grooves compared to rest of the species (Table 5.6). However, their impact on the abaxial surfaces were not quantifiable as the majority of the species had their abaxial surfaces covered by dense elongated trichomes (Fig. 5.16). Irrespective of their lower surface roughness, *R. nepalensis* also showed relatively higher PM densities on their leaves that, were not significantly different from the highest densities found on *R. rolfei*, and which can probably attributed to their relatively smaller size, composite leaves, and complex shape (Leonard *et al.*, 2016; Popek *et al.*, 2013; Weerakkody *et al.*, 2017).

5.3.5. Conclusion

The use of brambles on a living wall system showed a notable potential for capturing and retaining atmospheric PM, and there was a considerable inter-species variation in PM accumulation within the same genus (*Rubus*). The levels of PM capture recorded from plants in this living wall were was relatively lower compared to the plant species used in the previous experiment (Section 5.1) and these differences can be attributed to their variable species characteristics, the type of vegetation, and differences in ambient pollution concentration. The dense elongated trichomes found on the abaxial surfaces of the leaves of some of the *Rubus* spp. were found to have a negative impact on PM accumulation, whereas leaf surface roughness showed a positive impact on accumulating small particle sizes.

Chapter 6 : Evaluating the impact of individual leaf traits on atmospheric particulate matter accumulation using natural and synthetic leaves

This chapter is presented as an article published in the *Journal of Urban Forestry and Urban Greening* (Weerakkody *et al.* 2018a) with slight modification to align with the thesis structure.

6.1 Abstract

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

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

6.2 Introduction

Three different size fractions of Particulate Matter (PM) are of particular concern based upon their ability to be inhaled and toxicity: coarse particles/PM_{10} (aerodynamic diameter ≤10 μm), fine particles/$PM_{2.5}$ (aerodynamic diameter ≤ 2.5 μm), and ultra-fine particles/$PM_{0.1}$ (aerodynamic diameter ≤0.1 μm) (Chow *et al.*, 2006; Solomon *et al.*, 2012). Long-term exposure to coarse particles diminishes lung function and increases cardiovascular mortality (Gilmour *et al.*, 1996). $PM_{2.5}$ can reach the narrower spaces in lungs (Brunkeef and Holgate, 2002) and cause lung cancer and cardio-vascular mortality associated with acute ischemic events (Solomon *et al.*, 2012). Ultra-fine particles are more dangerous than the other size ranges as they can cross cell membranes and influence intracellular functions (Riddle, 2009). According to Seaton *et al.* (1995), $PM_{0.1}$ can cause systemic inflammatory changes by entering the blood stream or influence phagocytosis by accumulating in alveolar macrophages. They can also enter the brain via the olfactory nerves (Solomon *et al.,* 2012) which may cause central nervous system disorders (e.g. Alzheimer's disease and Parkinson's disease) depending on their chemical composition and toxicity (Allsop *et al.*, 2008; Maher *et al.* 2013).

Vegetation has been known as a sink for atmospheric PM for some time (Smith, 1977; Zulfacar, 1979) and the PM filtering behaviour of different types of vegetation has been studied using a range of different techniques (Beckett *et al.*, 2000a; Dover, 2015; Freer-Smith *et al.*, 2004; Leonard *et al.*, 2016; Maher *et al.*, 2013; McDonald *et al.*, 2007; Ottelé *et al.*, 2010; Sternberg *et al.*, 2011; Terzaghi *et al.*, 2013; Zhang *et al.*, 2017). Vegetation has been found to be more effective in removing PM from air compared to other building/land surfaces due to high air turbulence created by their complex morphology and large surface area (Roupsard *et al.*, 2013b; Tallis *et al.*, 2011).

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

For example, coniferous species with leaf needles have been frequently cited as being good PM filters compared to broad-leaved species (Beckett *et al.*, 2000b; Dzierzanowski *et al.*, 2011; Wang *et al.* 2011), and Shackleton *et al.* (2010) found "grass-like" (linear leaved) species to be the best PM filters out of the 16 species they tested. In contrast, Leonard *et al.* (2016) found significantly higher PM levels on lanceolate leaves compared to needle-like or linear leaves. The effects of epicuticular wax and leaf hairs have also produced mixed results. According to Dzierzanowski *et al.* (2011), the relationship between PM deposition and epicuticular wax does not depend on the amount of wax but on the structure and composition of the wax. Liu *et al.* (2012) found a negative impact of epicuticular wax in capturing particles but identified stomata, deep grooves, and leaf size as critically important characters. In contrast, Sæbø *et al.* (2012) found that PM deposition was a function of epicuticular wax content. Hairy leaves were found to be effective in accumulating PM in several different studies (Beckett *et al.*, 2000b; Kardel *et al.*, 2012; Ram *et al.*, 2012); conversely, Perini *et al.* (2017) found a negative impact of leaf hair on PM capture.

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

6.3 Materials and Method

6.3.1 Site description

Stoke-on-Trent is a city located in Staffordshire, United Kingdom with an estimated population of 259,140 and population density of 6,640 persons/km^2 (ukpopulation, 2017). Leek Road is one of the busiest single carriageways in Stoke-on-Trent, categorised as an A Road (Fig. 5.1), with a traffic density of 20,251 Average Daily Flow in 2016 (Department for Transport, 2017). Given the continuous pollution generation due to road traffic, a linear, grassed area, 11.5 m in width at Staffordshire University, located parallel to Leek Road (4.5 m distance from the road) (Fig. 5.1) was selected to erect experimental rigs.

6.3.2 Manufacturing the synthetic leaves and sampling natural leaves.

Synthetic leaves used in these experiments were hand-made, using stiffened Poplin. A 1.0 m x 1.4 m section of commercially available Poplin (125.0 gm^{-2}), was stiffened by painting a sago solution, a commercially available starch extracted from stems of *Metroxylon sagu*, (15 g of sago boiled in a 1 L of water) on the fabric to ensure synthetic leaves had no pleats or folds. Cardboard templates of different sizes and shapes (sizes and shapes are detailed in sections 6.2.4 and 6.3.5) were used to outline and cut the leaves from the fabric as required. Commercially available floral stems (plastic-paper covered stem wire, Handicrafts Ltd.) were stuck on one side of each of the leaves using fabric glue (Hobby glue gun- 230 V,15 W, Powerbox International Ltd.) and dried for 20 minutes. Using the same fabric, without any pleats or folds, produced artificial leaves with exactly the same surface characteristics and roughness (Fig. 6.1). Natural leaves required for the experiments were obtained from a free-standing living wall system (an experimentally manipulated system designed for our work on PM pollution, manufactured and installed by Nemec Cascade Garden Ltd., Czech Republic) (Fig. 5.1) located at the same site, facing Leek Road. Sampling and experiments were conducted on several occasions during March and April 2017. The mean temperature, mean humidity and mean wind speed of the study site was recorded as 12.8 ^{0}C, 68% and 2.3 m s^{-1} during this period.

6.3.3 ESEM/ImageJ approach to quantifying PM densities on natural and artificial leaves

PM accumulation on both natural and synthetic leaves used in this study were quantified using an ESEM (Model: JSM-6610LV) and ImageJ image analysis software (Ottele *et al.*, 2010; Sternberg *et al.,* 2010; Weerakkody *et al.*, 2017). Sampling numbers and methods of each experiment are detailed in the relevant sections below. In experiments where complete leaves were exposed to pollution all the leaves were synthetic (sections 6.3.4 and 6.3.5) and three leaf sections (5 mm x 5 mm) from every leaf blade were cropped out and mounted on aluminium stubs using double-sided carbon adhesive tabs for microscopic analysis. In the experiment in which only leaf sections were exposed to pollution (all from natural leaves) (section 6.3.6), whole leaf sections were mounted without cropping.

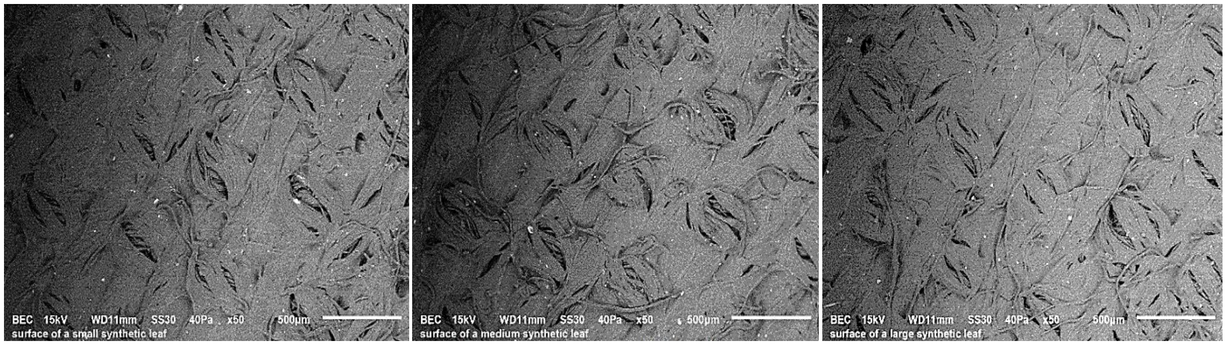


Figure 6-1 ESEM images (x50) of synthetic leaves used in manipulative experiments where size or shape were varied whilst holding other variables constant (see further Fig. 6.2).

In this series of images, the surface characteristics of small, medium, and large leaves (left to right) are visualized to demonstrate their similar surface characteristics.

Both natural and synthetic leaves were scanned under a low vacuum in the ESEM at x450 and x1,000 magnifications using Back Scattered Electrons, without any conductive coating following the same approach used in Weerakkody *et al.* (2017) (Chapter 4) to visualise natural leaves. The high carbon content of the fibres used in the synthetic leaves minimises conductive charging, and PM accumulation was also clearly imaged using this approach. Micrographs were taken at three random points on each leaf section (natural or synthetic), maintaining the same working distance and consistent contrast/brightness to help define the threshold in image processing. The smallest particle that could clearly be visualised using this approach was 0.1 µm in diameter. Considering their different health effects and different aerodynamics, the number of PM_1, $PM_{2.5}$, and PM_{10} on all the micrographs were quantified using ImageJ. The most appropriate threshold available in the auto-threshold menu was carefully selected using ten random micrographs to minimise potential human error of using a user-defined threshold. PM accumulation on each leaf section was estimated as PM density (number of PM per 1 mm^2) using the mean PM count on three random micrographs taken for each leaf section. In the experiments where whole leaves were used, mean PM density on each leaf was estimated taking the mean PM density of three leaf sections (see further, section 6.3.4 and 6.3.5).

6.3.4 The impact of leaf size on PM capture

Synthetic leaves were manufactured for the following three size ranges: small (1.7 cm^2) medium (28.9 cm^2) and large (59.6 cm^2) with 40 leaves in each range (Fig. 6.2a). The leaves were of the same basic elliptical shape and the same surface characteristics (i.e. same fabric). The size of natural leaves in the Nemec living wall outside the Science Centre were used as an approximate guide for the size ranges. Prior to use, the synthetic leaves were washed using a spray bottle and then dipped and shaken in deionised water in large beakers to remove existing particles. Subsequently, they were dried in a closed drying chamber (with controlled light and temperature regimes), thereby reducing contamination by indoor particulates. Twenty leaves from each size category were then scanned, to visualise any PM remaining on their surfaces, using the ESEM/imageJ approach; any significant variation in baseline PM levels between size ranges were identified using a one-way Anova (R statistical software version 3.2.5: R development Core Team, 2016). The remaining 20 leaves of each size category were then exposed to traffic pollution generated from Leek Road by mounting them on a wooden garden trellis. Leaves were attached to the trellis by their petioles using thin wires at 1.0 m -1.5 m height from the Road surface, keeping a similar configuration as vertical greenery systems (i.e. facing the road). Leaf arrangement was random using a Latin Square design to avoid any variation relating to columns and rows. The trellis was erected at the roadside edge of the grassed area, 4.5 m distance from the roadside and leaves were left exposed to traffic pollution for five consecutive dry days. Subsequently, leaves were taken to the laboratory using sealed storage boxes to provide minimal disturbance and PM densities were quantified using the ESEM/imageJ. Identification of baseline PM levels prior to exposure was carried out to identify if there were significant differences in the different leaf categories, and hence allow for adjustment of the experimental data by subtracting the mean baseline levels from the PM densities found on the roadside-exposed leaves (i.e. PM density on exposed leaves – mean baseline PM density) if required. Differences in PM levels captured by different sizes of leaves were identified using a one-way Anova followed by Tukey's pairwise comparison (n = 20). The leaf perimeter/leaf surface area ratio in each category was calculated to explain any differences in PM accumulation due to variable edge effects; the leaf perimeter was measured using ImageJ image analysis software.

6.3.5 The impact of leaf shape on PM capture

Synthetic leaves with exactly the same surface area (28.9 cm^2) and surface characteristics were made in three common leaf-shapes (elliptical, palmately-lobed, and linear) (Fig. 6.2b); 40 leaves were made for each shape category. Leaves were washed using the same approach followed in section 6.3.4 to remove existing particles and dried in a closed drying chamber avoiding contamination from indoor particulates. Twenty leaves from each shape category were then tested for their baseline PM levels using ESEM/imageJ analysis.

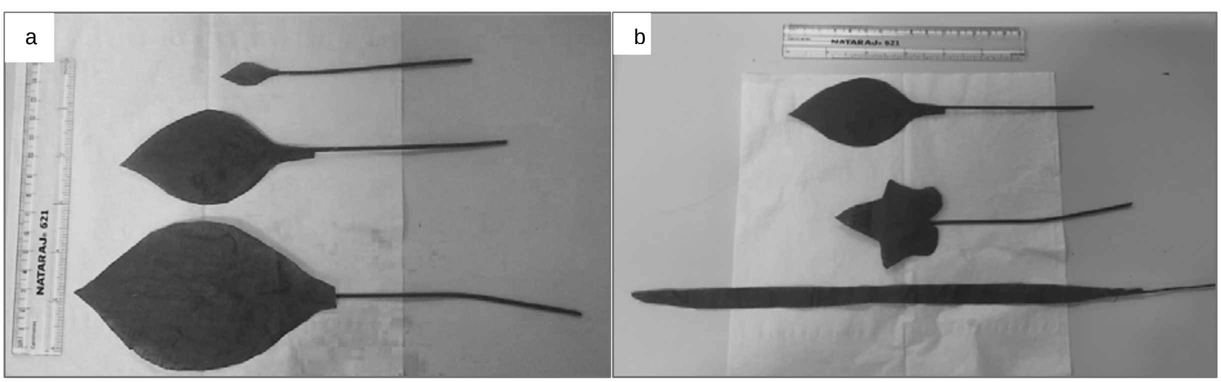


Figure 6-2 (a) an image of synthetic leaves designed with different leaf sizes but with the same shape and surface characteristics, and (b) an image of synthetic leaves designed with different shapes but with the same surface area and surface characteristics.

The remaining twenty leaves of each shape category were simultaneously exposed to traffic pollution on Leek Road for five consecutive days using a wooden garden trellis following the same approach given in section 6.3.4. Subsequent transfer of leaves to the lab, visualisation and counting of particulates, and statistical analysis followed the approach in Section 6.3.4 (n = 20). The Leaf perimeter/ surface area ratio in each category was calculated to explain any differences in PM accumulation due to variable edge effects.

6.3.6 The impact of leaf micro-morphology on PM capture

Sixteen square holes (1 cm x 1 cm) were cropped-out from plastic laminating pouches (ImageLast 125 Micron, Lyreco) in 4 equally spaced parallel rows. Four species of plant with different surface textures (e.g. hairy, smooth, rough and velvety) (*Geranium macrorrhizum* L., *Bergenia cordifolia* (L.) Fritsch, *Helleborus x sternii* and *Heuchera villosa* Michx. var. *macrorhiza*) used in the Nemec living wall (Fig. 5.1) were selected for this experiment. The leaf shape and size was standardised in this experiment so that differences in PM capture due to surface characteristics (micromorphology) could be identified. Forty leaves per species were randomly collected from the living wall and carefully washed by dipping and slowly shaking in cold deionised water to remove existing PM. Leaves were dried at 22 ^{0}C in a closed drying chamber for one hour. Subsequently, 1.5 cm x 1.5 cm sections were excised from every leaf blade and twenty leaf sections of each species were immediately tested for their baseline PM levels. Remaining leaf sections were attached to the back of the laminating paper replacing the empty squares in a randomised order (Latin Square design) using thin sticky tapes; this resulted in a grid of equal sized leaf squares with different micro-morphology (Fig. 6.3).

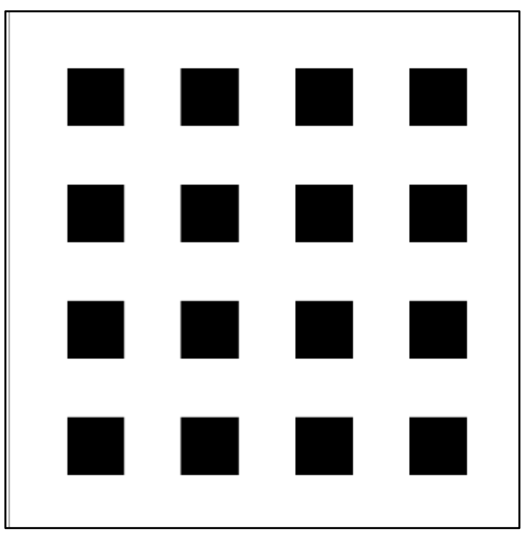


Figure 6-3 Schematic diagram of an experimental rig holding equally sized and shaped leaf sections with different micro-morphology arranged in a random order.

Five such rigs, each holding 16 leaf sections (4 leaf sections from each species) were made to hold a total of 80 leaf sections (20 leaf sections per species). The back of the laminating papers was then covered with wet layers of gauze to prevent dehydration of leaves, and inserted into plastic frames with water retentive backing. The plastic frames holding the leaf sections, with supportive material, were then exposed to traffic pollution generated from Leek Road (4.5 m distance from the roadside) for five consecutive days. Gauze layers were kept moist by spraying deionised filtered water through holes in the back of the plastic frames each day to avoid any structural changes in leaf sections due to dehydration. These experimental rigs were taken to the laboratory with minimal disturbance using the same approach followed in section 6.3.4 and 6.3.5. Leaf sections were carefully removed from the holding frames and PM densities on leaf sections were quantified using ESEM/ImageJ analysis. Statistical analysis followed the approach in Section 6.3.4 ($n = 20$). Specific leaf micromorphological characters (hairs, trichomes, epicuticular wax, ridges) of the leaf sections were observed using the ESEM at a range of magnifications as appropriate (Chapter 5).

6.4 Results

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

Baseline PM levels on synthetic leaves were not significantly different between different size categories for all particle size fractions ($p > 0.05$) hence they were not deducted from the PM levels on synthetic leaves exposed to roadside air pollution. Roadside-exposed leaves showed differential PM densities on leaves of different sizes (Fig. 6.4) (PM_1: $F = 114.9$, $p < 0.001$, $PM_{2.5}$: $F = 41.68$, $p < 0.001$, PM_{10}: $F = 76.53$, $p < 0.001$) in all particle size fractions. The highest mean PM densities in all particle size fractions (PM_1 =2,233 mm^{-2}, $PM_{2.5}$ =1,122 mm^{-2} and PM_{10} =303 mm^{-2}) were found on the smallest leaves and these levels were significantly higher than the PM levels in both medium and large-sized leaves ($p < 0.001$). The second highest mean PM densities in all PM size fractions were found on medium-sized leaves; the density of PM_{10} on medium sized leaves was significantly higher compared to larger leaves ($p < 0.001$). However, densities of PM_1 and $PM_{2.5}$ on medium sized leaves were not significantly different from larger leaves ($p = 0.06$ and $p = 0.22$ respectively). The leaf perimeter/surface area ratio was 0.2, 0.07 and 0.04 for small, medium and large sized leaves respectively (ratio between sizes, small: medium: large = 27 : 7 : 4).

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

Baseline PM densities on leaves of different shape categories were not significantly different ($p > 0.05$) and hence were not deducted from the PM densities on synthetic leaves exposed to roadside air pollution. There were differential PM levels on roadside-exposed leaves with different shapes (Fig. 6.5) in all PM size fractions (PM_1: $F = 21.06$, $p < 0.001$; $PM_{2.5}$: $F = 31.09$, $p < 0.001$; PM_{10}: $F = 37.9$, $p < 0.001$). The highest PM densities in all particle size fractions (PM_1 =2,226 mm^{-2}, $PM_{2.5}$ =652 mm^{-2} and PM_{10} =211 mm^{-2}) were found in palmately-lobed leaves and these densities were significantly higher than for both elliptical and linear leaves. The second highest PM densities were found on linear leaves in all particle size fractions and the density of PM_{10} on linear leaves was significantly higher compared to PM density on elliptical leaves (p =0.01). However, these levels were not significantly different for smaller PM sizes (PM_1: p =0.37 and $PM_{2.5}$: p =0.88). The leaf perimeter/surface area ratio was 0.07, 0.09 and 0.16 for elliptical, lobed and linear leaves respectively (ratio between shapes:- elliptical : lobed : linear = 7 : 9 : 16).

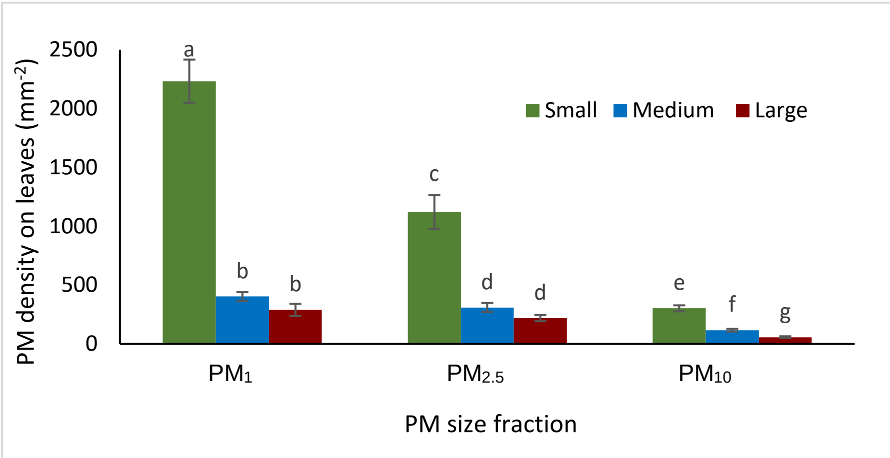


Figure 6-4 Mean±1SE PM densities on leaves of different sizes (small: 1.7 cm^2, medium: 28.9 cm^2, and large: 59.6 cm^2) but with the same shape and surface characteristics (see further Fig. 6.2a).
Species labeled with the same letter are not significantly different from each other within the given particle size fraction (PM_1: a,b; $PM_{2.5}$: c,d; PM_{10}: e,f,g).

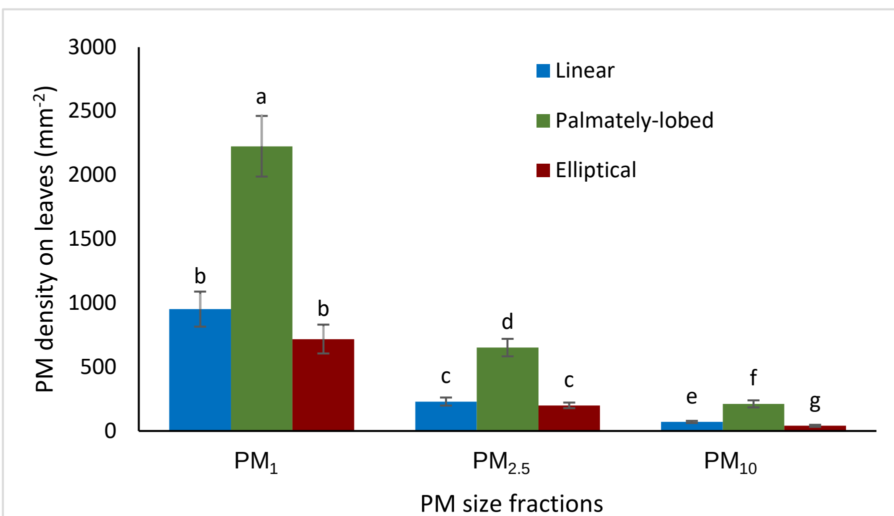


Figure 6-5 Mean ±1SE PM densities on leaves of different shapes but with the same surface area (28.9 cm^2) and surface characteristics (see further Fig. 6.2b).
Species labeled with same letters are not significantly different from each other within the given particle size fraction (PM_1: a,b; $PM_{2.5}$: c,d; PM_{10}: e,f,g).

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

Leaf micrographs of four species of plants (Fig. 6.6) showed their different surface micromorphologies and are detailed in Table 6.1. Baseline PM densities on leaf sections of different species were not significantly different ($p > 0.05$) after washing-off and hence were not deducted from the PM densities of leaf sections subsequently exposed to roadside air pollution. There were differential PM levels on roadside-exposed leaf sections of different species (Fig. 6.7) in all PM size fractions (PM_1: $F = 10.38$, $p < 0.001$; $PM_{2.5}$: $F = 12.27$, $p < 0.001$; PM_{10}: $F = 140.9$, $p < 0.001$). Leaf sections of *G. macrorrhizum* showed the highest mean PM densities in all particle size fractions (PM_1 =7,424 mm^{-2}, $PM_{2.5}$ =1,902 mm^{-2} and PM_{10} =383 mm^{-2}). PM_1 densities on *G. macrorrhizum H. villosa and H. sternii* were significantly higher than on *B. cordifolia* ($p < 0.001$) but not significantly different ($p > 0.05$) from one another. The density of $PM_{2.5}$ on *G. macrorrhizum* was significantly higher than *B. cordifolia (*p <0.001) and *H. villosa* ($p = 0.03$) but not significantly different from *H. sternii* ($p = 0.13$). *B. cordifolia showed* the lowest mean PM density in all particle size fractions (PM_1 =3,539 mm^{-2}, $PM_{2.5}$ =790 mm^{-2} and PM_{10} =69 mm^{-2}) which were significantly lower ($p < 0.05$) than the rest of the species. PM levels on *H. villosa* and *H. sternii* were not significantly different in the smaller PM size fractions ($p > 0.05$). PM_{10} levels were more varied between species compared to other size fractions and the densities of PM_{10} on leaf sections of each species were significantly different from each other.

Table 6.1 Micromorphological characteristics of different species of plants used in the study

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

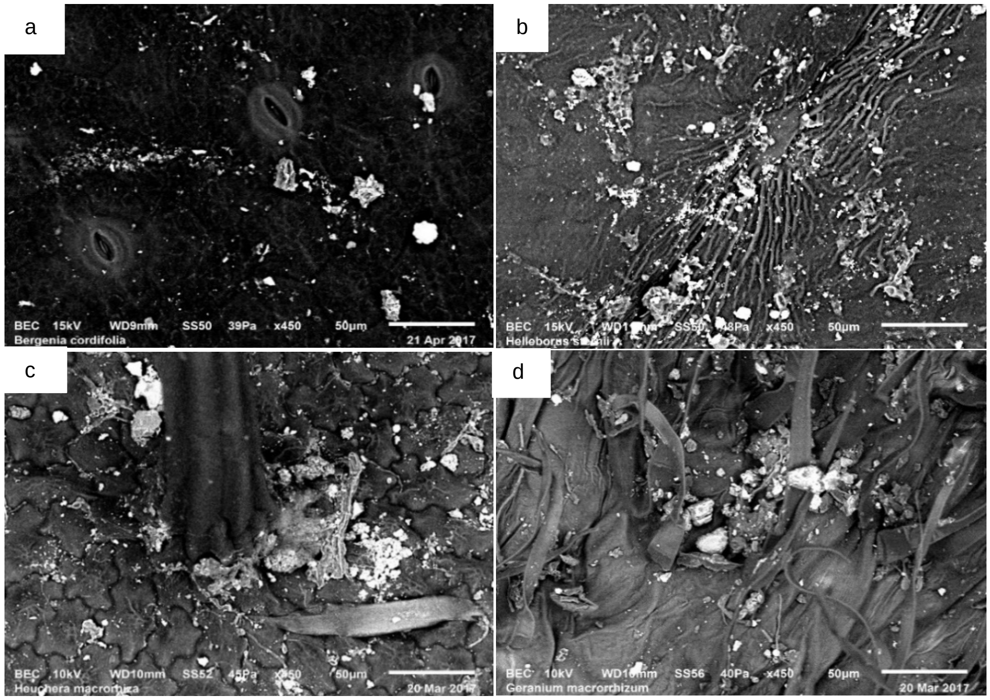


Figure 6-6 Scanning Electron Microscope images (450x) of leaf micromorphology of the adaxial surface of a) *B. cordifolia* b) *H. sternii* c) *H. villosa* (*Heuchera macrorhiza*) and d) *G. macrorrhizum*

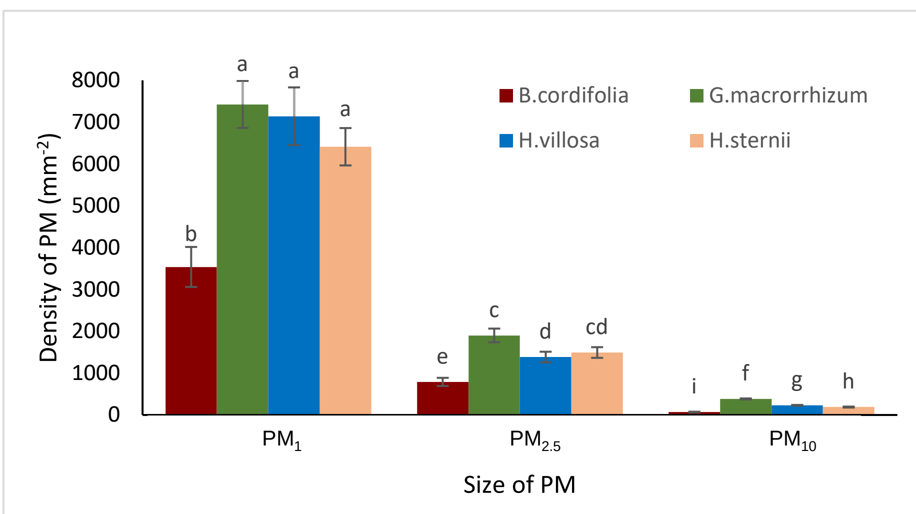


Figure 6-7 Mean ±1SE PM densities on real leaf sections with different micromorphology but with the same surface area and shape.

Species labeled with the same letters are not significantly different from each other within the given particle size fraction (PM_1: a,b; $PM_{2.5}$: c,d,e; PM_{10}: f,g,h,i).

6.5 Discussion

6.5.1 Impact of leaf size on PM accumulation on leaves

As synthetic leaves with the same leaf shape and same surface characteristics were arranged randomly, at the same height, and with the same pollution exposure levels and timings, the different PM capture levels that resulted (Fig. 6.4) can only be attributed to their size. In contrast to the findings of Sæbø *et al.* (2012), large differences in PM densities were found in all PM size fractions which demonstrates that size is an important trait to consider in assessing PM accumulation on leaves. Similar to the findings of Freer-Smith *et al.* (2005) and Leonard *et al.* (2016), high PM accumulation on smaller-sized leaves demonstrates their greater potential to capture and retain PM. An unpublished pilot study conducted (Chapter 3) on the distribution of particulates on leaf surfaces found elevated PM levels on leaf edges and tips except for linear-shaped leaves where there was no apparent change in density with distance from the leaf edge. The relatively larger edge effect present in smaller sized leaves (due to their high perimeter/surface area ratio) may have resulted in high levels of PM impaction on leaves. However, this pattern was only significant for PM_{10} and not for the smaller particles size fractions (PM_1 and $PM_{2.5}$); this difference may result from the different aerodynamic behaviour of different particle sizes (Slinn, 1982). Increased turbulence in the boundary layer around a deposition surface increases PM accumulation via turbulent transfer which is more important for smaller particle size fractions (Petroff *et al.*, 2008a; Slinn, 1982). More turbulence around leaf edges can result from leaves by swaying with the wind flow. There is a substantial difference in perimeter/surface area ratios between small and medium-sized leaves but this is much smaller between medium and larger leaves (small: medium : large = 27 : 7: 4). This might explain the large differences in PM levels between small and medium sized leaves and the smaller differences evident between medium and larger leaves.

6.5.2 Impact of leaf shape on PM accumulation on leaves

The shape of the deposition surface can directly influence the airflow pattern around the surface and hence, has a marked influence on PM deposition (Davidson and Wu, 1990; Petroff *et al.*, 2009), and this experiment with synthetic material showed that there were large differences in PM accumulation on leaves with different shapes. As additional factors (e.g. surface area, surface characteristics, exposure to weather and PM exposure levels) were held constant, the different levels of PM accumulation can only be attributed to the shape of the leaves. Although the different leaf shapes had different perimeter/surface area ratios (elliptical : lobed : linear = 7 : 9 : 16), they did not explain the differential PM accumulation by different shapes. Variable shapes generate different drag forces on them due to wind (Gemba, 2007), changing their influence on surrounding air flow patterns; the response of leaves to such forces can be by swaying, bending or fluttering (Gillies *et al.*, 2002) and hence different levels of turbulence can result. The pattern of PM accumulation between species was consistent for all PM size fractions (Fig. 6.5); palmately lobed leaves showed a greater potential to capture and retain PM, probably due to their complex shape. Lobed leaves create more than one “leaf-tip-like” area creating a more complex morphology; results of an unpublished pilot study (Chapter 3) showed elevated PM accumulation at leaf tips. Similar to the results of Leonard *et al.* (2016), PM accumulation on our elliptical and linear leaves was relatively poor. Even though linear leaves can be predicted to be good PM filters on the basis of their larger perimeter, they can also bend more readily with the wind flow (as these lengthy leaves are connected to a petiole with a narrow leaf base) without swaying with the wind currents, potentially resulting in lower levels of turbulence. However, despite having a linear shape, leaf needles are frequently cited as good PM filters (Beckett *et al.*, 2000a; Freer-Smith *et al.,* 2005; Mori *et al.*, 2015; Räsänen *et al.*, 2013), in addition to their epicuticular wax helping to provide a sticky surface to retain PM, this may also be attributed to their rigid/stiff nature compared to "grass-like" linear leaves. Further research in this area is clearly warranted.

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

Significant variations in PM levels on real leaf sections with different micromorphology but with area and shape held constant clearly demonstrated that leaf micromorphology has an impact on PM capture and retention and hence is an important leaf trait to consider where vegetation is being proposed for PM removal. Variation in PM_{10} levels were particularly interesting, where differential PM capture abilities were evident between all the species examined. In addition to turbulent deposition, capture by sedimentation is more important for PM_{10} compared to smaller particles (Petroff *et al.*, 2008a) and surface texture is known to be very important in preventing heavy PM rebounding from surfaces (Davidson and Wu, 1990). The greater potential of *G. macorrhizum* to capture and retain PM in all size fractions can be attributed to its complex micromorphology, with densely arranged trichomes and glandular hairs (Fig. 6.6) (Table 6.1). *H. villosa* is also slightly hairy and also showed relatively high PM accumulation supporting this argument. Leaf hairs are known to be important in increased PM accumulation (Beckett *et al.*, 2000b; Leonard *et al.*, 2016; Räsänen *et al.*, 2013; Ram *et al.*, 2012; Sæbø *et al.*, 2012) by increasing surface area (for capture) and by preventing re-suspension of captured PM (Prusty *et al.*, 2005; Qiu *et al.*, 2009). Having such protruding structures can also create complex micro-topography on leaf surfaces which also facilitates capture and retention of PM compared to smooth leaves. In addition, the hydrophobicity of some leaf hairs is known to have a positive impact on attracting metal-based charged particles (Fernández *et al.*, 2014). The poor ability of leaves of *B. cordifolia* to capture and retain PM can probably be attributed to their glossy smooth leaf surface. PM can readily rebound from smooth surfaces resulting in the retention of only a small fraction of captured particles (Davidson and Wu, 1990). *H. sternii* also showed relatively high PM levels compared to *B. cordifolia* which could probably be attributed to their deep surface ridges and epicuticular wax. While a positive impact of epicuticular wax on PM capture is frequently cited in the literature (Barima *et al.*, 2014; Räsänen *et al.*, 2013; Sæbø *et al.*, 2012), several studies found reduced PM levels on waxy surfaces due to their variable chemical structure and composition (Faini *et al.*, 1999; Kardel *et al.*, 2012; Leonard *et al.* 2016) and also due to the self-cleansing ability of wax tubules (Wang *et al.*, 2011). However, rough leaf surfaces with a large number of ridges were found to be effective in PM capture (Kardel *et al.*, 2012; Ram *et al.*, 2012; Zhang *et al.*, 2017) and

Barima *et al.* (2014) found significantly higher PM levels on ridged leaf surfaces compared to waxy surfaces. The relatively high levels of PM on *H. sternii* could either be attributed to their epicuticular wax or ridges or to the collective impact of both these characters. However, these levels did not significantly exceed the PM levels found on hairy leaves for any particle size fraction, highlighting the importance of leaf hairs in the capture and retention of PM. Nevertheless, the actual impact of these species of plants under field conditions, may be quite different when they stand as leaves instead of leaf sections. For example, irrespecive of their ridged surface, Weerakkody *et al.* (2017) found very low PM accumulation on leaves and plants of *H.sternii* citing their wide leaves and low leaf area index as potential reasons. Therefore, the potential of leaf surfaces for the capture and retention of PM can be enhanced or limited by other influencial variables such as leaf size, shape, location, configuration and leaf area index (Freer-Smith *et al.,* 2005; Leonard *et al.*, 2016; Weerakkody *et al.,* 2017). However, selecting species sharing at least a few of these micro-morphological characters (e.g. ridged hairy leaf surfaces) would proabably be beneficial to increase PM accumulation.

6.5.4 Implications of the findings

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

6.6 Conclusion

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

Chapter 7 : The impact of rainfall in remobilising particulate matter accumulated on leaves of evergreen species grown on a green screen and a living wall

This chapter is presented, with slight modification, as an article published in the *Journal of Urban Forestry and Urban Greening* (Weerakkody *et al.* in press).

7.1 Abstract

Green walls have recently been identified as a green infrastructure (GI) solution to the problem of particulate matter (PM) air pollution. Green wall systems mostly use evergreen plants as the leaves are retained throughout the year; however, researchers have argued that evergreen foliage becomes saturated with PM and fails to capture more due to a long retention time on the leaves. This study evaluated the potential of (simulated) rainfall to remobilise these captured PM and renew the capture ability of the leaf surfaces of four evergreen species (*Heuchera villosa* Michx., *Helleborus* × *sternii* Turrill, *Bergenia cordifolia* (Haw.) Sternb., *Hedera helix L.*) used in a living wall and a green screen located along a busy road in Stoke-on-Trent, UK. The approach used compared PM densities on pre- and post-rain exposed leaf surfaces (using leaf halves of the same leaf) and using a paired t-test to identify any significant reduction in PM due to the rainfall. An Environmental Scanning Electron Microscope (ESEM) and ImageJ image analysis software were employed to quantify the PM densities on leaves. The reduction of PM on leaves, following exposure to 16 mm.hr^{-1} simulated rain in six different rainfall durations was estimated in all four species in order to evaluate any variable impact of rainfall on different species of plants. PM wash-off levels on leaves of *H. helix* by 41 mm.hr^{-1} rain was also evaluated, using the same rainfall durations, to assess any differential impact of rainfall intensity on PM wash-off. This study revealed a significant impact of rainfall in washing the particles off the leaves in all rainfall durations used. A one-way Anova in a General Linear Model showed a differential impact of rainfall in remobilising PM on different species of plants. The rainfall with higher intensity (41 mm.hr^{-1}) showed a significantly higher impact on PM wash-off compared to 16 mm.hr^{-1} rain. The results of this study demonstrated the potential of green walls to act as good PM traps throughout the year by recycling their capture surfaces.

Key words: PM wash-off; simulated rain, evergreen species; leaf micromorphology; Green infrastructure

7.2 Introduction

Particulate matter (PM) less than 10 µm in aerodynamic diameter (PM_{10}) was graded as the most influential contributor to outdoor air pollution due to the higher prevalence of morbidity caused by PM in comparison to other air pollutants; 90% of urban dwellers were estimated to have been exposed to PM pollution which exceeded WHO standards in 2014 (WHO, 2016). An estimated 428,000 premature deaths were caused by $PM_{2.5}$ (PM less than 2.5 µm aerodynamic diameter) in Europe in 2014 (Guerreiro *et al.*, 2017), and 29,000 annual premature deaths in the UK were attributed to PM pollution (RCP, 2016). The International Agency for Research on Cancer categorized PM as a human carcinogen (IARC, 2015) mainly relating to its impact on lung cancer (Pope III, 2002; Raaschou-Nielsen *et al.*, 2016); in addition, childhood leukemia and bladder cancers are also known to be associated with PM pollution (Raaschou-Nielsen *et al.*, 2017). Short-term and long-term exposure to PM pollution is also a factor in several respiratory diseases such as asthma, cardio-vascular diseases (Anderson *et al.*, 2013; Pope III, 2002; Seaton *et al.*, 1995) and has also been linked to neurodegenerative diseases via particles in the ultrafine range entering the brain (Maher *et al.*, 2016).

Studies have revealed that vegetation has a great potential for removing PM pollutants from the atmosphere (Beckett *et al.*, 2000b; Blanusa *et al.*, 2015; Dover and Phillips, 2015; Freer-Smith *et al.*, 2005; Jin *et al.*, 2014; McDonald *et al.*, 2007; Nowak *et al.*, 2013; Song *et al.*, 2015; Weerakkody *et al.,* 2017). The use of vegetative barriers to filter PM pollutants, thereby improving human health and wellbeing, is of particular interest, especially when implemented at the city scale (Lin *et al.*, 2016; Baldauf, 2017; Tong *et al.*, 2016). Irrespective of the type of green Infrastructure (GI) used or the study location, many reports have demonstrated that some species of plant are better than others in capturing PM (Currie and Bass, 2008; Freer-Smith *et al.*, 2005; Leonard *et al.*, 2016; Liang *et al.*, 2017; Sæbø *et al.*, 2012; Song *et al.*, 2015; Weerakkody *et al.*, 2017) thus emphasising the need for careful species selection. Evergreen species can be particularly useful in countries where there is seasonal variation in, as their foliage is retained throughout the year, and their high PM removal efficiency has been repeatedly reported in previous studies (Beckett *et al.*, 2000b; Dochinger, 1980; Hwang *et al.*, 2011; Wang *et al.* 2011). However, in a comparison of evergreen conifers and deciduous species, Beckett *et al.* (2000b) stated that leaf needles of conifers are retained for several years without being shed and become saturated with particles. In addition, they have the potential to accumulate toxic chemicals over their extended lifespan whereas deciduous species, with broader leaves, have the advantage that captured particulates can be shed at leaf-fall and new shoots and buds provide new capture surfaces. It has also been argued that the accumulated PM on leaves can be remobilised either by re-suspension to the atmosphere by wind (Currie *et al.,* 2008; McPherson *et al.,* 1994) washed off by rain (McPherson, *et al.*, 1994; Przybysz *et al.*, 2014; Schaubroeck *et al.*, 2014; Wang *et al.*, 2015; Xu *et al.*, 2017) or concentrated at the tips of the leaves by rainfall (Van Bohemen *et al.*, 2008). If this is indeed the case, then the PM capture surfaces of leaves of both evergreen and deciduous species is probably regularly restored without saturation occurring – provided the local environment supports resuspension.

Vertical greenery systems have recently been recognised as a short term GI solution to PM pollution (Weerakkody *et al.*, 2017; Perini *et al.*, 2011; Cheetham *et al.*, 2012). Plant species used in these systems are mostly evergreen as their foliage is retained throughout the year maintaining their aesthetic value. Therefore, understanding the leaf PM wash-off behaviour of plants used in the various types of green wall, in order to be able to select the most appropriate species (i.e. those whose surfaces can continue trapping particles without saturation) is important.

Schaubroeck *et al.*, (2014) developed a multi-layered PM removal model to study the influence of weather on PM remobilisation in forest canopies of *Pinus sylvestris*; however, applying their finding to urban shrubs, herbs or vertical greenery systems is problematic due to their different configurations. There have been few attempts made to quantify the PM remobilisation ability of rainfall at the leaf level. Wang *et al.* (2015) quantified PM reduction on leaves of an evergreen tree *Ligustrum lucidum* by collecting its leaves after natural rainfall events; however, rainfall intensities, continuity, and intervals between rainfall and sampling were not specified. Although it is difficult to mimic a natural rainfall event to meet all potential conditions, rain simulation can mimic certain important attributes of rain to provide information on PM reduction under specific rainfall intensities and volumes. Perini *et al.* (2017) evaluated PM reduction due to rainfall by washing leaves in water, an approach which might not provide an accurate simulation of rainfall as washing in water cannot simulate specific/different rainfall intensities, amounts of rain, or the kinetic energy carried by raindrops (Neinhuis and Barthlott, 1998; Wang *et al.*, 2015; Xu *et al.*, 2017). Przybysz *et al.* (2014) attempted to provide a similar effect to rainfall by spraying 20 mm of water on leaf needles of *Pinus sylvestris* (Scots pine), though there was no information on rain intensity, duration or continuity. Xu *et al.* (2017) carried out a comprehensive study on PM wash-off on leaves of three deciduous species of trees and one evergreen shrub by simulating three rainfall intensities using an artificial simulator. In this experiment, plant twigs were exposed to simulated rainfall, the washed-off PM was collected, and the amount of PM in the wash-off was then estimated gravimetrically; unfortunately, they did not report information on the size ranges of PM washed-off/retained by leaves.

The aim of this study was to evaluate the impact of rainfall in remobilising PM accumulated on leaves of some evergreen species used in two vertical greenery systems: a green screen and a living wall. In these systems, the leaf configuration is similar, both have a more vertical leaf orientation (i.e. tips toward the ground) than plants grown at ground level which possibly enhances PM wash-off. Our rainfall simulation approach was similar to the approach of Xu *et al.*, (2017). However, in contrast to the gravimetric approach towards evaluation of PM wash-off used by Xu *et al.*, (2017), we used Particle Number Concentration (PNC) to ensure that the water soluble fraction of PM, which accounts for about 45% of particulates (Li *et al.*, 2012), was included in the analysis. Wash-off levels were estimated with reference to the main PM size fractions discussed above (i.e. PM_{10}, $PM_{2.5}$ and PM_1,) rather than to total suspended PM, due to their differential deposition rates/aerodynamic behaviour and health effects (Davidson and Wu, 1990; Slinn, 1982). The overall aim was to develop a greater understanding of the effectiveness and suitability of using vertical greenery systems in PM reduction.

7.3 Material and methods

7.3.1 Study site and species description

The study was conducted in a laboratory on the campus of Staffordshire University, Stoke-on-Trent, UK using artificial rain simulators (see Section 7.3.3). The leaves of four plant species, three from a living wall system (Nemec Cascade Garden Ltd.), *Heuchera villosa* Michx. (hairy alumroot), *Helleborus* × *sternii* Turrill (blackthorn strain) and *Bergenia cordifolia* (Haw.) Sternb. (Elephant's ears), and one from a green screen (Mobilane ®), *Hedera helix L*. (English ivy), both located on the University campus, were used in this study (Figures 2.1b and 2.1e). All four species are evergreen and are commonly found in vertical greenery systems in Europe. Plants were selected to represent different leaf morphological types: *H. villosa* with hairy and velvety leaves of, *H. sternii* with ridged and slightly waxy leaves, *B. cordifolia* with smooth and glossy leaves, and *H. helix* with leaves with thick epicuticular wax. The closest potential PM source for both the systems was road-traffic due to their close proximity (5 m) to a busy 'A' road, Leek Road with a 20,251 average daily traffic flow (Department for Transport, 2017). As both the systems were located at the same site, both facing the road, and both the same distance from the roadside, all four species were exposed to similar environmental and pollution conditions.

7.3.2 General experimental approach

The experimental approach was the estimation of PM wash-off by rainfall by comparing the PM held on two equal halves of leaves exposed to roadside pollution (Ottelé *et al.*, 2010b): one half was evaluated immediately after removal from the plant, and the other evaluated after exposure to simulated rainfall. A rainfall event simulating 16 $mm.hr^{-1}$ rain was used to evaluate the impact of rainfall on PM remobilization on leaves of all four species in six different rainfall durations; 16 $mm.hr^{-1}$ is within the normal range of rainfall experienced in the UK (Data.Gov.UK^{Beta}). As leaves of *H. helix* were predicted to show very low PM wash-off rates (Ottelé *et al.*, 2010), due to their waxy epicuticle retaining impacted particles, *H. helix* was selected to evaluate any differential impact of an intense rainfall event of 41 $mm.hr^{-1}$ in six different rainfall durations.

7.3.3 Design and operation of rainfall simulators

An Environmental Chamber (FMH1/900/-40/+150/P/R, JTS Ltd.) with 1.1 m wide x 1.5 m high x 1.2 m deep internal dimensions was used to simulate 41 $mm.hr^{-1}$ rainfall (Fig. 7.1a). The environmental chamber was equipped with four plastic spray nozzles, a humidity control fan, and a flow meter. The rain profile required was programmed and operated with a timer for specific rainfall conditions and durations. The minimum rainfall intensity that could be achieved with a consistent flow rate (41 $mm.hr^{-1}$), the mean ambient temperature in Stoke-on-Trent during the period of experimentation (17°C), and six rainfall durations (10, 20, 30, 40, 50 and 60 minutes) were used to run the rain profile. As the Environmental Chamber could not simulate more typical rainfall intensities with consistent flow rates, a handmade rain simulator (Fig. 7.1b) was designed to simulate an average rainfall event.

The simulator was set up in the laboratory using a commercially available watering kit containing small plastic spray nozzles, a Poly-ethylene (PE) hose with 4 mm diameter and 5 m length, and a quick-fit hose socket (hosepipe connector with a regulator) with 2 l hr^{-1} maximum flow rate (Yanqh48 Pvt. Ltd.).

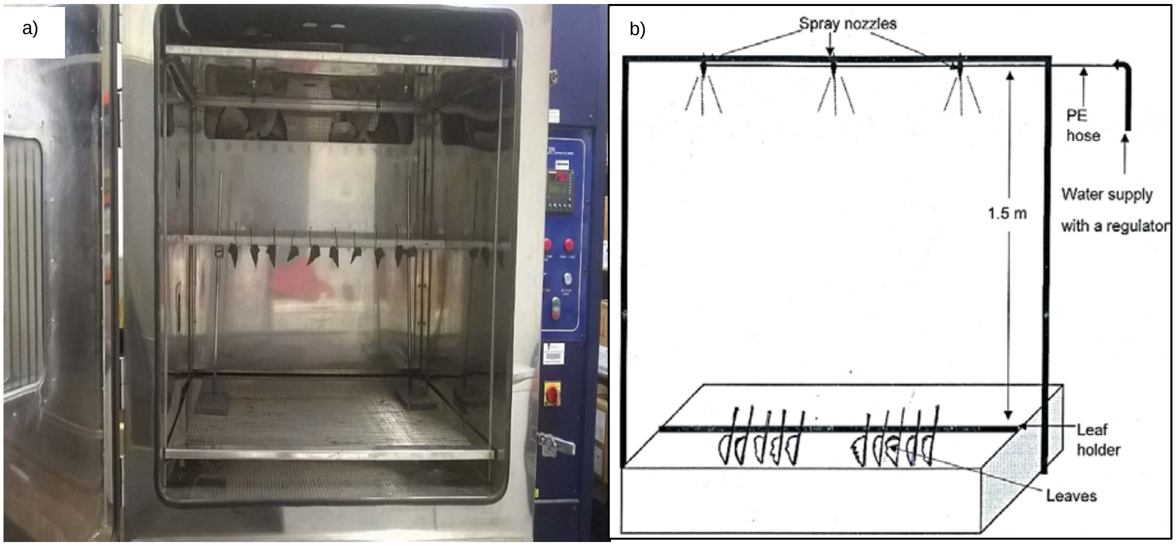


Figure 7-1 a) the Environmental Chamber used in the 41 mm.hr^{-1} rainfall study, and b) a schematic diagram of the 16 mm.hr^{-1} simulator used in this study. Note the semi-vertical leaf arrangement in the simulators

Three spray nozzles were connected to the PE hose and they were held at 1.5 m height above the leaf holder (Fig. 7.1b). The system was connected to the main water supply using the quick-fit hose socket. Calibrated glass beakers were arranged on a deep plastic tray completely covering the basal area of the simulator and the simulator was optimised to simulate a 16 mm.hr^{-1} rainfall by adjusting the regulator.

7.3.4 Leaf sample collection

Sample collection and storage in this study followed the same approach given in Weerakkody *et al.* (2017) (Chapter 4). Sampling was carried out in daylight under non-rainy weather conditions with at least three non-rainy days prior to sampling. In order to explore the effect of heavy rainfall (41 mm.hr^{-1}) on PM remobilization, 20 leaves of *H. helix* were removed from green screens on six separate days (120 leaves in total) during the period March-April 2016. To explore the impact of more typical rainfall levels (16 mm.hr^{-1}) on PM remobilisation, 120 leaves per species from all four species of plants were sampled on 12 separate days in March – April 2017. All the species were equally sampled on each occasion (ten leaves/day) to avoid any differential influence of weather on existing PM levels between sample days. Samples were arranged in plastic storage boxes and sealed with minimal disturbance and transferred to the laboratory.

7.3.5 Simulating the effect of rainfall on leaves

In both the experiments, all the leaves were sectioned into halves at the midrib, one half with the petiole attached and the other without. The latter half of each leaf was used to quantify existing PM levels using the ESEM/ImageJ approach (Section 7.3.6). The remaining halves were attached to a 1 m-long plastic ruler using adhesive tape via their petioles. In the first experiment, with higher rainfall intensity, the ruler holding the leaves was placed inside the environment chamber at 50 cm height from its floor (Fig. 7. 2a) with tips pointing down at a slight angle. The chamber door was locked and the rain profile was run for a particular time period on each sample day, i.e. a different time duration on each day. Time durations were 10, 20, 30, 40, 50 and 60 minutes producing 6.8 mm, 13.6 mm, 20.5 mm, 27.3 mm, 34.2 mm, and 41.0 mm rainfall amounts. Once the rain stopped, leaves were carefully removed and stored in clean plastic containers using blue tac to attach them by their petioles to the container walls (avoiding the leaf surfaces touching each other or the container surfaces). The storage containers, with leaves, were kept open (to aid evaporation of rain water) in a closed drying chamber at 25°C for two hours and PM levels retained on the leaf surfaces were then quantified using ESEM/imageJ analysis. In the experiment using 16 $mm.hr^{-1}$ rainfall, leaf halves of four species were attached to a ruler in random order following the same approach as above. The ruler, with leaves, was subsequently placed in the rainfall simulator by attaching it to the side-walls of the tray (Fig. 7.1b) and exposed to rain for the same six durations used in Experiment 1 producing 2.6 mm, 5.33 mm, 8 mm, 10.66 mm, 13.33 mm, and 16 mm of rainfall. Leaf drying and quantification of the retained PM followed the same approach as above.

7.3.6 ESEM/ImageJ approach to estimation of PM levels on leaves

The amount of PM on leaves before and after exposure to rain was quantified in terms of PM density (number of PM on a 1 mm^2 section of leaf) using an ESEM and ImageJ image analysis software, following a similar approach to that given in Weerakkody *et al.* (2017) (Chapter 4). Six leaf sections were excised from each half of leaf blade (avoiding edges, midrib, tip and leaf base) (Chapter 3), three sections to image the adaxial surface and three for the abaxial surface. Leaf sections were mounted on small aluminium sample holders using adhesive carbon tabs. Leaf sections were uncoated and scanned under a low vacuum. Three random micrographs per each leaf section were taken at 450x and 1,000x using Back Scattered Electron signals to estimate the mean PM density. Subsequently, micrographs were imported into ImageJ software and the amount of PM in the following size fractions PM_{10} ($PM_{2.5}$ - PM_{10} excluding $PM_{2.5}$), $PM_{2.5}$ (PM_1 - $PM_{2.5}$ excluding PM_1), and PM_1 ($PM_{0.1}$ - PM_1) were quantified using the auto-threshold tool. The mean density of each PM size fraction on both the adaxial and abaxial surfaces of every leaf half (before and after rain) were separately calculated using their respective micrographs (9 micrographs for each half leaf surface).

7.3.7. Statistical analysis

Any significant reduction in PM densities in a particular rainfall duration was identified by comparing the two halves of each leaf (pre- and post-rain exposure) using a paired t-test following a Shapiro-Wilk normality test (R statistical software version 3.2.5: R Development Core Team, 2016) and the results expressed as percentage reductions (log transformation of data was carried out where required). Any inter-species variation in PM reduction on the adaxial surfaces of the leaves by rainfall was then identified using a one-way Anova in a General Linear Model (GLM) (R Package: MASS). Tukey's HSD post-hoc test (package: Agricolae) was used for the pairwise comparison of species and to cluster them into groups based on significant differences in wash-off levels. The impact of different rainfall intensities on PM wash-off from the adaxial surfaces of leaves of *H. helix* was evaluated by comparing the PM wash-off percentages from 16 $mm.hr^{-1}$ and 41 $mm.hr^{-1}$ rainfall, using a student's t-test following a Shapiro-Wilk normality test; as residuals met the assumptions of normality, non-transformed data was used in the analysis (Betts *et al.,* 2007; Wilson *et al.,* 2010). Any difference in wash-off between the adaxial and abaxial surfaces of the leaves of *H. helix* from exposure to 41 $mm.hr^{-1}$ was also compared using a student's t-test.

7.4 Results

7.4.1 Reduction of PM density on leaves exposed to rain

Both rainfall intensities used in this study removed significant amounts of all particle size fractions from the adaxial surfaces of the leaves of all the species in all rainfall durations (Fig. 7.2, Fig. 7.3a and Appendix 5). On the abaxial surfaces of the leaves of all species, there was no significant reduction in PM density following exposure to 16 $mm.hr^{-1}$ after 60 mins (Appendix 5) and, in a few instances, there were slightly higher numbers of PM recorded on post-rainfall samples; the analysis was thus not continued for shorter rainfall durations on abaxial surfaces. Nevertheless, 41 $mm.hr^{-1}$ intensity rainfall did show significant reductions in all PM size densities on the abaxial surfaces of the leaves of *H. helix* at all rainfall durations (Fig 7.4b and Appendix 5).

The reduction of all PM size densities on the adaxial surfaces of the leaves of *H. helix* following exposure to 41 $mm.hr^{-1}$ rainfall was significantly greater than those reductions due to 16 $mm.hr^{-1}$ rain in all rainfall durations, except for PM_1 wash-off within 30 minutes and PM_{10} wash-off within 10 minutes (Fig. 7.4). The differences in PM_{10} reduction densities on the adaxial and abaxial surfaces of leaves of *H. helix* exposed to 41 $mm.hr^{-1}$ rainfall was substantially greater than for the smaller sized particles ($PM_{2.5}$ and PM_1) as a result of significantly higher wash-off levels on the adaxial surface in all rainfall durations (in 10 mins: t =2.66 and p=0.01, 20 mins: t=2.07 and p=0.04, 40 mins: t= 2.13 and p=0.04, 50 mins: t=2.33 and p=0.02, 60 min: t=4.26, p<0.001) apart from that lasting for 30 minutes (t=1.11 and p=0.27) (Fig. 7. 4). PM_1 wash-off levels were not significantly different between the two leaf surfaces except for higher wash-off levels on the adaxial surfaces when exposed for 60 minutes (t=2.23 and p=0.03); similarly, $PM_{2.5}$ densities were not significantly different between the two leaf surfaces except for higher wash-off levels on the adaxial surfaces exposed for 20 (t=2.44 and p=0.02) and 60 minutes (t=3.35 and p=0.002). Of the three particle size fractions examined, PM_{10} had the highest wash-off levels from the leaves of all four species of plants when exposed to 16 $mm.hr^{-1}$ rain in all rainfall durations and PM_1 showed lowest wash-off levels in most of the rainfall durations in all the species of plants (Fig. 7.2). However, this pattern was not consistent on leaves of *H. helix* exposed to 41 $mm.hr^{-1}$ rainfall; although PM_1 showed the lowest wash-off levels in most of the rainfall durations (apart from the abaxial surface in 60-minute duration), there were several overlaps between PM_{10} and $PM_{2.5}$ (Fig. 7.3).

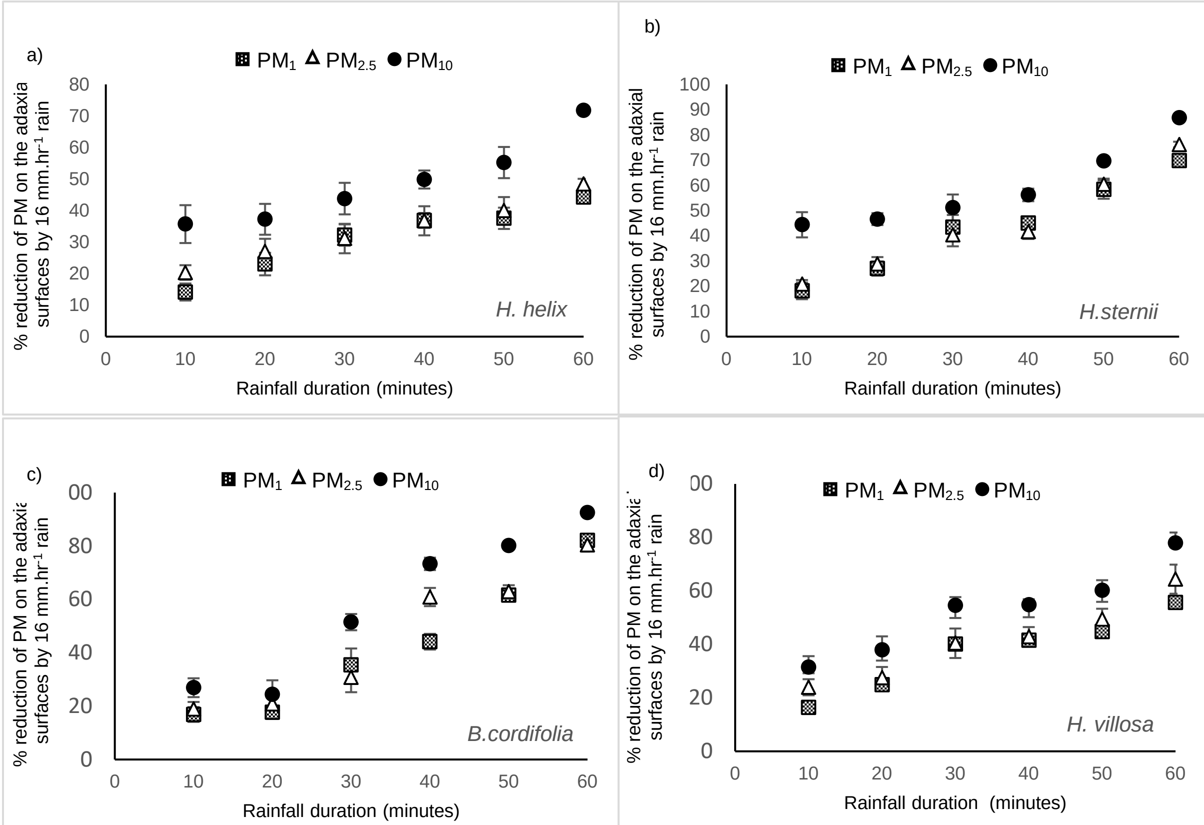


Figure 7-2 Reduction of PM densities (%) ± 1SE following exposure to 16 mm.hr^{-1} rainfall on the adaxial surfaces of leaves of a) *H. helix*, b) *H. sternii*, c) *B. cordifolia* and d) *H. villosa.* Note different scales on y-axis.

For statistical comparisons, see Appendix 5.

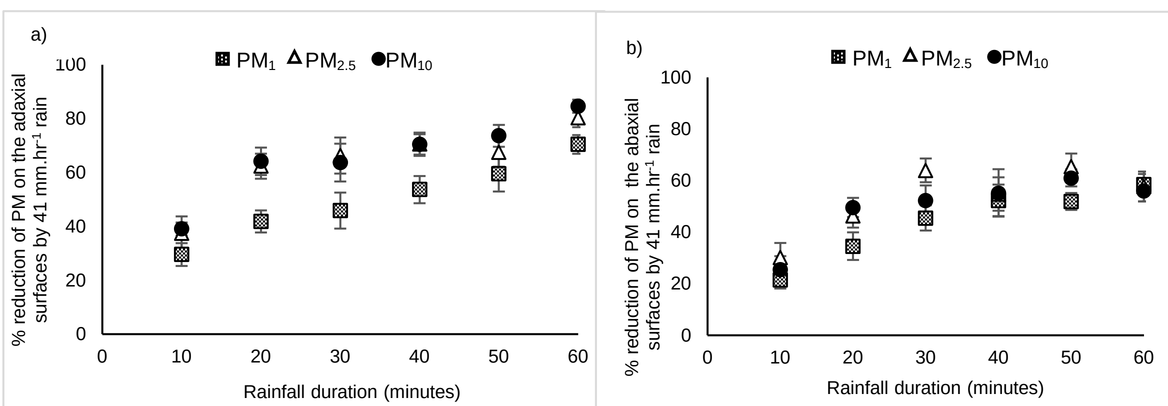


Figure 7- 3 Reduction of PM densities (%) ± 1SE following exposure to 41 mm.hr^{-1} rainfall on a) the adaxial surfaces of *H. helix* and b) the abaxial surfaces of *H. helix*.

For statistical comparisons, see Appendix 5.

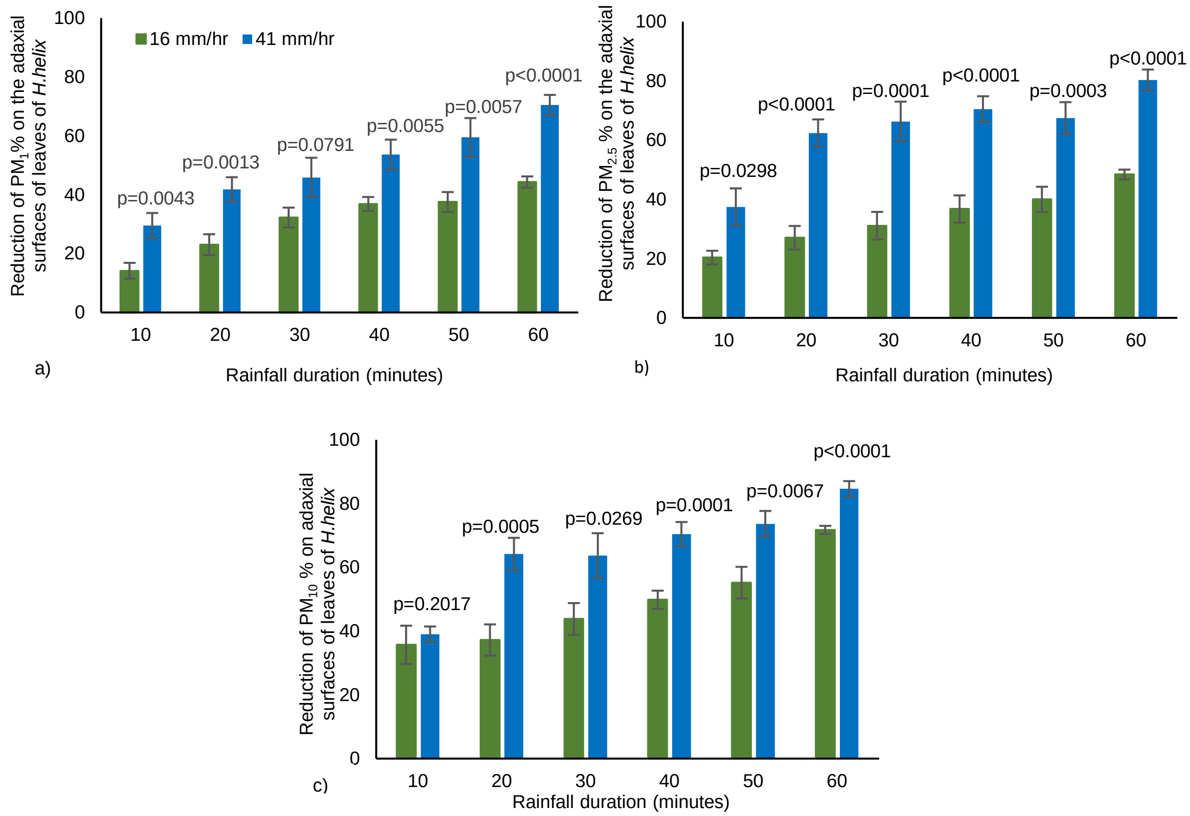


Figure 7-4 Reduction of PM densities (%) ± 1SE a) PM_1 b) $PM_{2.5}$, and c) PM_{10} on the adaxial surfaces of leaves of *H. helix* exposed to different durations and intensities of rainfall.

7.4.2 Inter-species variation in PM wash-off on leaves following exposure to 16 mm.hr^{-1} rainfall

Significant inter-species differences in PM_{10} reduction on the adaxial surfaces of leaves were apparent for all rainfall durations with the exception of 30 minutes (Fig. 7.5 and Table 7.1). However, $PM_{2.5}$ showed significant inter-species variation only for the longer time periods (for 40 mins: F=9.75 and p< 0.0001, 50 mins: F=10.15 and p< 0.0001, 60 mins: F=32.55 and p< 0.0001) and PM_1 showed significant inter-species variation only for the longest durations (for 50 mins: F=11.73 and p< 0.0001, 60 mins: F=27.44 and p< 0.0001) (Fig. 7.5, Table 7.1). Despite this, there was some consistency in PM wash-off levels on leaves of different species of plants at longer time durations, as rainfall durations become shorter this consistent pattern disappears (Table 7.1). At longer rainfall durations (40, 50, and 60 minutes) *B. cordifolia* showed the greatest wash-off levels for all particle size fractions and a reduction of 82.0% of PM_1, 80.4% of $PM_{2.5}$, and 92.5% of PM_{10} was found following exposure to 16 mm of rain (Fig. 7.5). The lowest wash-off levels of all particle sizes were mostly found on leaves of *H. helix* and a reduction of 44.30% of PM_1, 48.42% of $PM_{2.5}$, and 71.76% of PM_{10} was following exposure to 16 mm of rainfall.

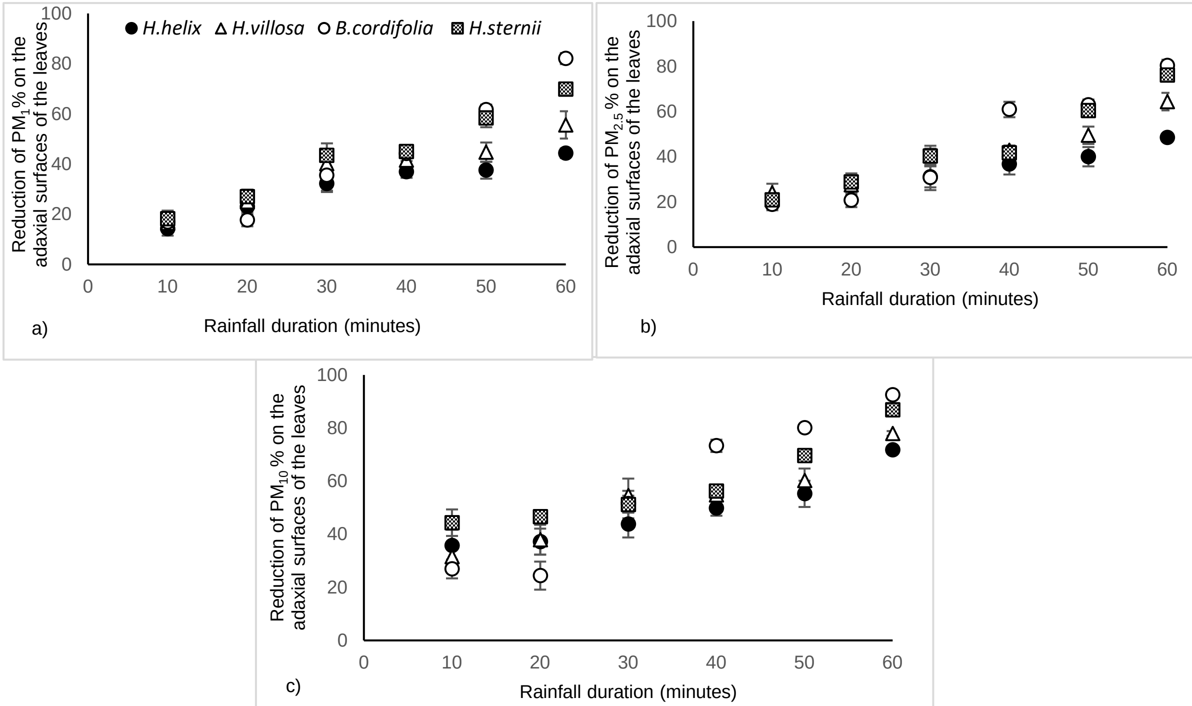


Figure 7-5 Reduction of PM densities (%) ± 1SE of a) PM_1 b) $PM_{2.5}$, and c) PM_{10} on the adaxial surfaces of the leaves of different species of plants following exposure to 16 $mm.hr^{-1}$ rainfall of different durations.

7.5 Discussion

Significant reductions in PM densities on the adaxial surfaces of leaves exposed to both rainfall intensities showed that rain can remobilise the particulates accumulated on leaves of all four evergreen species studied (Fig. 7.2), thereby preventing saturation of leaf surfaces and promoting the capture of particles throughout the year/life of the leaves. Przybysz *et al.* (2014), Xu *et al.* (2017), and Wang *et al.* (2015) reported similar findings using different rainfall volumes; in contrast, Perini *et al.* (2017) found that particles between $PM_{2.5}$ and PM_{10} were not washed-off by rain. The differences in findings between these studies can probably be attributed to different rainfall simulation methods used; washing leaves in water might not have provided enough intensity and kinetic energy carried by direct rainfall (Neinhuis and Barthlott, 1998) to remove particles from leaf surfaces. The PM wash-off found for the four evergreen species used in this study when exposed to 16 mm of rainfall was higher (Fig. 7.2) than the PM reduction of 28% and 48% by 10.4 mm and 31.9 mm rainfall (respectively) on leaves of *Ligustrum lucidum* (Wang *et al.*, 2015) and 30% to 40% reduction by 20 mm rainfall on leaf-needles of *Pinus sylvestris* (Przybysz *et al.*, 2014).

Table 7.1 Results of GLM in the analysis of inter-species variation in PM washed-off from the leaves of different species of plants following exposure to 16 $mm.hr^{-1}$ rainfall of different durations

Duration (min)	PM size fraction	Inter-species variation resulted by one-way Anova	Group assigned by Tukey's HSD test*			
			B. cordifolia	*H. sernii*	*H. villosa*	*H. helix*
10	PM_1	F=0.31, p=0.81	a	a	a	a
	$PM_{2.5}$	F=0.57, p=0.63	a	a	a	a
	PM_{10}	F=2.71, p=0.047	b	a	ab	ab
20	PM_1	F=1.51, p=0.22	a	a	a	a
	$PM_{2.5}$	F=0.87, p=0.46	a	a	a	a
	PM_{10}	F=3.76, p=0.01	b	a	ab	ab
30	PM_1	F=0.96, p=0.42	a	a	a	a
	$PM_{2.5}$	F=1.44, p=0.24	a	a	a	a
	PM_{10}	F=0.81, p=0.48	a	a	a	a
40	PM_1	F=1.67, p=0.18	a	a	a	a
	$PM_{2.5}$	F=9.75, p< 0.0001	a	b	b	b
	PM_{10}	F=17.42, p< 0.0001	a	b	b	b
50	PM_1	F=11.73, p< 0.0001	a	a	b	b
	$PM_{2.5}$	F=10.15, p< 0.0001	a	ab	bc	c
	PM_{10}	F=10.3, p< 0.0001	a	ab	bc	c
60	PM_1	F=27.44, p< 0.0001	a	b	c	c
	$PM_{2.5}$	F=32.55, p< 0.0001	a	a	b	c
	PM_{10}	F=74.9, p< 0.0001	a	b	c	d

*PM washed-off on from leaves of species assigned with the same letter, within each PM size fraction and within each rainfall duration, were not significantly different from each other (p > 0.05).

However, rainfall intensities used in these studies were not specified. A similar range of 51% to 70% reduction by 15 mm rainfall was reported by (Xu *et al.* (2017) on three deciduous species and one evergreen species, however, it was difficult comparing results of these two studies as they are based on reduction of PM masses rather than PM densities. Xu *et.al.* (2017) also reported that PM can only wash-off when the rainfall exceeds the leaf water-storing capacity. However, even the smallest amount of rain simulated, 2.7 mm (10 minutes of 16 $mm.hr^{-1}$ rainfall) in this study removed significant amounts of particles from the leaves (Fig. 7.2); in vertical greenery systems, the leaf water-storing capacity might have a reduced impact due to the leaves generally pointing towards the ground. Rainwater can probably more easily flow down the slope of such leaves resulting in higher PM removal rates even with modest amounts of rain. However, running experiments using lower rainfall intensities than 16 $mm.hr^{-1}$ (using more advanced rainfall simulators) might be useful to explore this further.

Following exposure to 16 mm.hr^{-1} rainfall over a full 60-minute period, PM densities on the abaxial surfaces of tested leaves did not show any significant wash-off levels, probably as the underside of the leaves were shielded from the rain (Xu *et al.*, 2017). However, intense rainfall (41 mm.hr^{-1}) carries more kinetic energy (Xu *et al.*, 2017) which can probably disturb leaf orientation sufficiently to cause them to move, twist, and turn and hence receive rainfall on their abaxial surfaces resulting in PM wash-off. In a real-world scenario, the wind state during rainfall might cause a similar effect and is likely to have an impact on PM wash-off from the leaf surfaces. Linking these explanations to the tree literature is problematic as studies have either analysed the total wash-off levels by rainfall (Wang *et al.*, 2015) or the abaxial surfaces were excluded from the experiment due to expected lower contact levels (Xu *et al.*, 2017).

In agreement with the studies of Przybysz *et al.* (2014) and Wang *et al.* (2015), larger particle sizes showed higher wash-off rates on exposure to 16 mm.hr^{-1} in most of the rainfall durations whilst PM_1 showed relatively low wash-off rates. This suggests that larger particles were more loosely bound to leaf surfaces compared to smaller particles and/or PM_{10} tends to have a higher fraction of water soluble ions than smaller fractions. However, there was not such a clear pattern in intense (41 mm.hr^{-1}) rainfall, with significant amounts of both $PM_{2.5}$ and PM_{10} being remobilised whilst PM_1 was largely retained. Intense rainfall can probably remove substantial amounts of particles irrespective of their size, unless they are strongly bound to epicuticular surface wax, as are the majority of finer particles (Terzaghi *et al.*, 2013b; Dzierzanowski *et al.*, 2011; Popek *et al.*, 2013), or entrenched in fine surface features (Weerakkody *et al.,* 2017). Significantly variable PM wash-off levels resulting from two different rainfall intensities (except for PM_1 wash off within 30 minutes and PM_{10} wash off within 10 minutes) on leaves of *H. helix* (Fig. 7.4) demonstrated that, a higher rainfall intensity has a positive impact on PM removal from the leaves compared to a lower intensity. Xu *et al.* (2017) found a similar positive impact of high rainfall intensities on PM wash-off; nevertheless, this influence became weaker with increased intensity at 50 mm.hr^{-1} indicating a retained fraction of PM even at higher rain intensities. However, higher PM removal rates on *B. cordifolia* (82.0% of PM_1, 80.4% of $PM_{2.5}$ and 92.5%) compared to other species, at 16 mm rain suggests that PM retention can probably be species specific.

Inter-species variation found in PM wash-off from leaves (Fig. 7.5, Table 7.1) can be, at least partly, attributed to their different surface micromorphologies. According to Xu *et al.* (2017) leaf roughness reduces the kinetic energy of rain that falls onto leaf surfaces. The higher levels of remobilisation of PM on the leaves of *B. cordifolia* compared to other species may thus have resulted from the lack of energy-dissipating structures on its surface. Whilst leaf roughness is likely to reduce PM remobilisation, and smooth surfaces promote it, dense epicuticular waxes on the leaves of *H. helix* may have a role to play in preventing wash-off if it strongly binds PM to the leaf surface compared to the species with sparsely arranged wax plates (Dzierzanowski *et al.*, 2011; Ottelé *et al.*, 2010; Terzaghi *et al.*, 2013) resulting in relatively lower PM wash-off levels. Despite their noticeable epicuticular wax plates, *B. cordifolia* showed the lowest PM retention rates. This may have been influenced by the localised distribution of the wax plates, potential differences in structure and chemical composition of wax (Barthlott *et al.,* 1998), especially compared to *H. helix*. According to Perini *et al.* (2017), PM (2.5-10 µm) captured on leaves of *H. helix* is not washed-off by rain; in contrast, we found significant wash-off levels in all particle sizes on leaves of *H. helix* with all rainfall durations. When exposed to 16 mm of rainfall (60 minutes duration) 44.30% of PM_1, 48.42% of $PM_{2.5}$, and 71.76% of PM_{10} were washed-off from leaves of *H. helix,* suggesting green screens should act as good PM traps throughout the year. Although leaves of *H. sternii* had localised dense epicuticular wax and a rough leaf surface, the hairy leaves of *H. villosa* retained greater levels of PM compared to *H. sternii* in most of the rainfall durations. The more complex microtopography of hairy leaves (Beckett *et al.*, 1998) and their potential secretions (Tomaszewski *et al.*, 2014) might have helped retain particles by interfering with the kinetic energy of rainfall resulting in a reduced PM removal rate. All four species used in this study, despite different leaf morphotypes, showed a considerable potential for rainfall-mediated remobilization of all important particle size fractions and for the restoration of capture surfaces. Hence, the use of evergreen species in living walls and green screens are unlikely to be diminished as PM traps through saturation as suggested by Becket *et al.* (2000b). In non-rainy seasons, spraying water on foliage using a sprinkler or watering hose with a moderate pressure could potentially enhance their benefits as PM filters.

7.6 Conclusion

A rainfall with 16 $mm.hr^{-1}$ intensity showed a considerable potential for washing-off PM_1, $PM_{2.5}$, and PM_{10} from the leaves of four evergreen species grown in a living wall and a green screen located at the campus of Staffordshire University, UK. However, there were significant inter-species variations in wash-off PM in most of the rainfall durations used, which can probably be attributed to their different micromorphologies. Larger particle sizes showed relatively higher wash-off levels compared to smaller particles. Smooth-leaved *B. cordifolia* had relatively higher PM wash-off levels from their leaves whilst *H. helix*, with a waxy epicuticle, had the lowest wash-off rates. A higher rainfall intensity of 41 $mm.hr^{-1}$ had a positive impact on PM wash-off from leaves of *H. helix,* resulting in significant wash-off levels on both adaxial and abaxial surfaces of its leaves. The results of this study demonstrated the potential of rainfall for restoring PM capture surfaces of evergreen species used in living walls and green screens suggesting that they have the ability to act as PM filters throughout the year without being saturated by pollutants.

Chapter 8 : General Discussion and Conclusion

This chapter will compile the key findings of this thesis reflecting upon the extent to which the specific objectives of this research were met. The potential use of living walls as PM traps will then be evaluated based on the outcome of this thesis and challenges in implementation will be discussed. The chapter will be concluded by proposing key areas for further research that would enhance the benefits of living wall systems in the reduction of PM.

The review of the literature on the ability of vegetation to mitigate PM pollution revealed that the value of living wall systems in the reduction of PM pollution was relatively overlooked. Although there were some studies that focused on the ability of PM capture by VGSs using climbing species (Dover and Phillips, 2015; Ottelé *et al.*, 2010; Perini *et al.*, 2017; Sternberg *et al.*, 2011), prior to this thesis, and to a study on a green façade with two species of shrubs (Perini *et al.*, 2017), there were no published studies specifically focused on living walls in this respect. Since traffic-generated PM has been predominantly identified as a serious threat to human health (Kelly and Fussell, 2012; Pant and Harrison, 2013; Ranft *et al.*, 2009; Stanek *et al.*, 2011), this research was carried out to explore the potential for living walls to serve as near-road PM filters and to optimise their overall PM capture efficiency. Each experiment conducted in this research achieved one or more of the specific objectives defined in Chapter 1.

8.1 Inter-species variation in PM accumulation on leaves and optimal species composition for living wall systems

Similar to the findings on ground and roof vegetation reviewed in the literature (Beckett *et al.*, 2000b; Blanusa *et al.*, 2015; Freer-Smith *et al.*, 2004; Perini *et al.*, 2017; Song *et al.*, 2015; Speak *et al.*, 2012), the results presented in this thesis revealed a considerable inter-species variation in PM capture and retention by plant species grown on all three living walls studied (Weerakkody *et al.*, 2017/ Chapter 4 and Chapter 5). These findings emphasised the importance of species choice in use of living walls to mitigate PM pollution. Some species, even within the same genus, were found to have differential levels of PM accumulation on their leaves. For example among *Rubus spp., R. rolfei, R. yiwanus and R. nepalensis* captured more PM than *R. ichangensis or R. lambertianus.* On the other hand, some species belonging to the same genus showed very similar PM capture performance; for example, both the species belonging to the genus *Berberis* showed similarly high performance, and of the four species tested in the genus *Veronica,* three species showed similarly high performance (Chapter 5). Closer observations of these species showed that similarly performing species shared similar leaf characteristics and structure (leaf arrangement and LAI), whilst the species with differential PM accumulation levels were morphologically different irrespective of their taxonomic relationship. For example, those species of *Veronica* with clusters of very small leaves and high LAI (*V. albicans, V. youngii* and *V. vernicosa*), showed very high performance in PM capture (Figures 4.3 and 5.2), whilst *V. salicifolia,* which had relatively elongated leaves and a slightly lower LAI (loosely arranged leaves compared to the rest), showed only a moderate level of PM accumulation (Fig. 4.3). Sæbø *et al.* (2012) reported similar observations in ground vegetation by finding similarly high PM capture levels on *Pinus mugo* and *Pinus sylvestris*, and differential levels on *Tilia cordata* and *Tilia* × *europaea*. Therefore, when selecting species based on their ability to capture and retain PM, characteristics exhibited at the species level is much more important compared to their taxonomic relationship at the level of genus. The inter-species variation in PM wash-off by rainfall (Chapter 7) suggested that this is also an important factor (see further to section 8.4) to be considered in the selection of species, to ensure plants function as PM filters throughout the year.

The variation in PM densities on the leaves of different species of plants, and between adaxial and abaxial surfaces of the leaves, demonstrated the significance of leaf characteristics on PM accumulation (further evaluated in Section 8.2), whilst the variation in overall PM removal ability by 100 cm^2 areas of the plants (once the LAI was incorporated) suggested the importance of the structural arrangement of leaf distribution on PM accumulation. Although some species showed relatively low PM densities on their leaves due to leaf surface characteristics, their overall PM capture abilities were enhanced by high LAI values. In Weerakkody *et al.,* 2017 (Chapter 4), despite having relatively low densities of $PM_{2.5}$ and PM_{10} on the leaves of *G. macrorrhizum*, their high LAI enhanced their ability to remove these PM. Similarly, the low LAI of some species negatively influenced their potential for removal of PM and hence they are unsuitable to use in living walls as PM filters. The smaller-leaved *H. officinalis* and *G. odoratum* (Chapter 4) had low PM densities compared to other smaller-leaved species and also had lower LAIs (due to the sparsely distributed leaves) which further reduced their PM capture ability and, hence, resulted in significantly lower PM capture levels for all particle size fractions (Fig. 4. 3).

PM dry deposition is a function of local weather conditions (Litschke and Kuttler, 2008; Petroff *et al.*, 2008b; Slinn, 1982; Tomasević *et al.*, 2005) and the experiments focused on inter-species variation were conducted in different locations under different weather conditions (Weerakkody *et al.,* 2017 and Chapter 5). Therefore, the quantities of captured PM on plant species in the different living wall systems we used were not directly comparable. There were four species used in both Birmingham and Stoke-on-Trent living walls; *T. vulgaris, G. macrorrhizum, H. sternii* and *P. scolopendrium*. Similar to the findings of Sæbø *et al.* (2012) on trees and shrubs, those species that performed well in PM capturing at one location showed similarly high performance in the other location (e.g. *T. vulgaris*) whereas those species with poor PM capture ability at one location showed poor performance in both locations (e.g. *H. sternii* and *P. scolopendrium*). These observations confirmed the conclusion made by Sæbø *et al.* (2012), that the PM deposition is more of a species-specific process and, regardless of their location, the ability of vegetation to capture and retain PM can, to a large extent, be determined based on the species present and the pollution concentration. Nevertheless, when the actual particle number concentrations (PNC) are considered, there were site-specific differences in PM accumulation in all four species, which could mainly be attributable to different concentrations of pollutants, influence of weather, distance to the pollution source, and orientation of the living wall relative to the source of pollution (Nowak *et al.*, 2006; Sæbø *et al.*, 2012).

Irrespective of different environmental conditions and variable PM sources (rail or road traffic), smaller-leaved species on living walls in both the Birmingham and Stoke sites showed the highest PM accumulation levels on their leaves (Weerakkody *et al.* 2017/Chapter 4 and Weerakkody *et al.* 2008b/Section 5.1). Although there was no comparable research on VGSs, Popek *et al.* (2013) reported similar results on shrubs and trees finding higher PM accumulation on species with small leaflets. Freer-Smith *et al.* (2005) also reported higher PM accumulation on smaller-leaved species based on a comparison between leaf needles of conifers and broad-leaved species. The coniferous species used in this study *J. chinensis* was particularly good at PM capture (Weerakkody *et al.*, 2008b/ Section 5.1). Previous studies that focused on woodland species in different environments supported this finding by reporting high performance of coniferous species in the reduction of PM pollutants (Beckett *et al.* 2000b; Chen *et al.*, 2017; Dochinger, 1980; Freer-Smith *et al.*, 2003; Hwang *et al.* 2011; Ram *et al.,* 2012; Tallis *et al.*, 2011). However, Beckett *et al.* (2000b) noted that evergreen conifers have the potential to accumulate toxic chemicals during their long needle-retention time and are less tolerant to high traffic pollution and to winter salt. Therefore, regular monitoring of plant health will probably be required in practice.

In addition to *J. chinensis*, almost all the other species with smaller leaves - *H. albicans, B. sempervirens, T. vulgaris*, *H. youngii, V. vernicosa*, *B. buxifolia*, *B. x media*, and *S. japonica* - showed greater levels of leaf PM accumulation compared to large leaved species, except for *H. officinalis* and *G. odoratum* (Weerakkody *et al.*, 2017/Chapter 4; Weerakkody *et al.,* 2018b /Section 5.1). Reflecting the findings of Beckett *et al.* (2000) and Hwang *et al.* (2011) on ground vegetation, most of the wide-leaved species used in these living walls - *H. sternii, P. veris, P. amplexicaulis, B. cordifolia, L. kamtschatica, H. americana and H. villosa* - accumulated lower PM levels compared to other species (Weerakkody *et al.*, 2017/Chapter 4; Weerakkody *et al.,* 2018b./Section 5.1); however, there was no such comparison possible in the study of *Rubus* since all the species examined were wide-leaved (Section 5.3).

According to Smith and Jones (2000) and Shackleton *et al.* (undated), grasses and “grass like” species are better PM filters compared to trees, shrubs and herbs, whereas Fowler *et al.* (2004) reported that grasslands in West Midlands of England had poor PM capture ability. A species of grass, *D. cespitosa* and two “grass like” species (*C. caryophyllea*, and A. gramineus) used in this research showed moderate abilities to capture and retain PM, which were higher than most of the wide-leaved species (Weerakkody *et al.,* 2018b. /Section 5.1); in contrast, the PM removal ability of the “grass like” *L. nivea* was extremely poorer (Weerakkody *et al.*, 2017/ Chapter 4). These differences suggest the importance of individual leaf traits in species choice for PM removal rather than their general growth form. Both species of ferns used in this study *P. scolopendrium* and *B. spicant* showed very poor PM removal ability from both rail and road traffic; however, these species are commonly found in living walls in the UK. As ferns have been recognised for their ability to remove some Volatile Organic Compounds (VOCs) from the atmosphere (Kim *et al.,* 2008), further research on ferns in order to identify species which are good at both PM and VOC removal is warranted.

This thesis identified the best performing species in PM capture and retention to use as PM filters in living walls, experimenting on 40 plant species used in the living wall industry in the UK and Europe. When all three PM size fractions were considered, nine plant species were consistently best performing (*J. chinensis, V. vernicosa*, *B. buxifolia*, *B. x media*, *T. vulgaris*, *S. japonica, B. sempervirens*, *H. albicans, and H. youngii)* while ten species showed consistently worst performance (*P. scolopendrium*, *H. sternii*, *P. veris, B. spicant, L. nivea, P. amplexicaulis*, *B. cordifolia*, *L. kamtschatica, R. ichangensis, R. lambertianus* and *R. alceifolius*). Performance of the rest of the species were moderate and variable in different occasions. Using combinations of most efficacious species in PM capturing as appropriate to local climatic conditions would be more effective in removing PM from air compared to VGSs with a single climbing species or living walls with the typical species combinations used by different companies.

8.2. Important leaf morphological characters in PM capture and retention

The differences in PM densities on leaves of different species of plants and on the adaxial/ abaxial surfaces of the leaves, and similar PM densities on leaves with similar morphology highlights the influence of species-specific leaf characters on PM capture. The literature revealed that PM dry deposition on vegetation is driven by the interactions between particles and plant surface characters (Chen *et al.*, 2016; Leonard *et al.*, 2016; Litschke and Kuttler, 2008; Tomasević *et al.* 2005). The analyses carried out to evaluate the impact of leaf characters on PM accumulation using natural leaves (Weerakkody *et al.*, 2017/Chapter 4; and Chapter 5) produced ambiguous results with reference to some leaf traits; the resulting correlations between PM densities and leaf characters were probably affected by other influential leaf traits (Leonard *et al.*, 2016; Weerakkody *et al.*, 2018a). The discrepancy of findings in the literature in this respect can probably also be attributable to the collective impact of leaf characters. However, synthetic/natural leaf models used in Weerakkody *et al.* (2018a) (Chapter 6) drew more robust conclusions on the impact of these leaf characters when the effect of other influential variables were standardised.

Of the leaf traits examined (leaf size, shape, and micromorphology), the effect of leaf size on accumulation of all particle sizes was most noticeable (Chapters 4, 5 and 6); smaller-leaved species (see Section 8.1) showed higher PM densities irrespective of the differences in their micromorphology (e.g. *T. vulgaris* and *B. sempervirens*) whilst the majority of the large-leaved species were found to have relatively low PM densities on their leaves. The increased level of turbulence that can be created in the airflow between small leaflets may enhance the PM deposition on their surfaces (Farmer, 2002). The results of the experiment with standardised leaf models in variable sizes, but with the same shape and morphology, confirmed this inverse relationship between leaf size and PM accumulation (Weerakkody *et al.*, 2018a/Chapter 6). A comparatively larger edge-effect in smaller-sized leaves, due to their high perimeter/surface area ratio, might have caused the high levels of PM impaction on leaves (Weerakkody *et al.,* 2018a/Chapter 6). Freer-Smith *et al.* (2005) and Leonard *et al.* (2016) reported similar findings of having higher PM accumulation in species with smaller leaves; in contrast, Sæbø *et al.* (2012) did not find any correlation between leaf size and PM accumulation, probably due to the effect of other influential variables. As mentioned in Section 8.1, two out of eleven smaller-leaved species (*H. officinalis* and *G. odoratum)* were found with poor PM accumulation. Sparsely arranged, soft, fragile leaves of *H. officinalis* and *G. odoratum* with low structural complexity and roughness might have failed to create sufficient turbulence in the surrounding airflow to promote high levels of impaction.

As different shapes generate different drag forces (Gemba, 2007), the shape of a deposition surface has a marked influence on the sorrounding airflow pattern which, in turn, directly affects PM depostion (Davidson and Wu, 1990). In the experiments conducted with natural leaves there was no noticable pattern of PM densities on leaves with reference to their shape except for slightly higher PM densities on lobed leaves among large-leaved species (e.g. *G. macrorhizzum, H. helix, G. renadii, H. americana*). However, an experiment with synthetic leaf models demonstrated a greater potential of palmately-lobed leaves to accumulate PM compared to elliptical and linear leaves (Weerakkody *et al.,* 2018a/Chapter 6). Similarly, Ingold (1971) reported high PM accumulation on leaves with complex shapes compared to simple leaves and Leonard *et al.* (2016) reported poor PM capture abilities of linear and elliptical leaves. Although needle-like linear leaves (e.g. *J. chinensis*) are known as good PM filters (Beckett *et al.*, 2000b; Freer-Smith *et al.,* 2005; Weerakkody *et al.*, 2008b/ Section 5.1), densities of small particle sizes on "grass like" linear leaf models were not different from those on elliptical leaf models. In contrast to leaf needles, grass-like leaves can readily bend with the wind without swaying to create sufficient turbulance for PM impaction (Weerakkody *et al.*, 2018a/Chapter 6). Therefore, even within similar shaped leaves there may be variable capture levels based on their rigidity or flexibility, length, and orientation. Further research in this area using a range of different leaf shapes is recommended in order to further refine our knowledge of effective leaf shapes.

Differential PM accumulation on standardised leaf sections (with equal shape and size) with variable micromorphology, showed an important impact of leaf micromorphology on PM accumulation (Weerakkody *et al.*, 2018a/Chapter 6). Although a positive impact of leaf hair/trichomes on leaf PM accumulation has frequently been reported (Beckett *et al.*, 2000b; Leonard *et al.*, 2016; Prusty *et al.*, 2005; Qiu *et al.*, 2009; Ram *et al.*, 2012; Sæbø *et al.*, 2012; Weber *et al.*, 2014), in the present study, a significant positive impact of leaf hair/trichomes on PM accumulation was found only on two occasions out of twelve (the density of PM_{10} on the adaxial surfaces of the leaves of plants used in the Nemec living wall and the density of PM_1 on the abaxial surfaces of the leaves of *Rubus spp*). In addition, dense elongated trichomes on the abaxial surfaces of *Rubus. spp.* showed a negative impact on leaf PM accumulation. Although Perini *et al.* (2017) also reported a negative impact of leaf hair on PM capture on leaves of façade greening, their findings were based on the typical leaf hair forms (categorised as hair/trichome in this study). These results suggested that the impact of leaf hair on PM accumulation could be variable depending on their structural differences such as dimensions, texture, hydrophobicity, and secretory/non-secretory nature. Although there was not consistent, statistically significant, relationship between leaf hair and PM densities common to all PM sizes or to findings from different sites, almost all of the species with hairy leaves used in this study, apart from *Heuchera villosa,* showed relatively high PM accumulation compared to the rest of the wide-leaved species. These observations still suggest a positive impact of leaf hair on PM accumulation, although it is probably less influential compared to smaller leaf size. The effect of leaf hair in *H. villosa* might have been overridden by its larger leaf size; high PM accumulation on hairy *G. macrorrhizum and H. villosa* used in Weerakkody *et al.* (2018a) (Chapter 6) reflected the positive impact of leaf hair when the size and shape were standardised.

The surface ridges of leaves did not show any significant relationship with PM accumulation except for positive relationships showed with the density of $PM_{2.5}$ on the adaxial surfaces of *Rubus spp* and $PM_{2.5}$ on the abaxial surfaces of the leaves of species used in Nemec living wall (Chapter 5). Similarly, when findings of all the experiments were brought together, surface grooves of leaves were positively correlated only with the densities of PM_1 and $PM_{2.5}$ on the adaxial surfaces of the leaves of *Rubus spp.* (for *Rubus spp.* abaxial surfaces were excluded in this analysis). However, even though there was no consistent pattern common to the findings of all the experiments, the results of some experiments still suggested a positive impact of leaf roughness at least on smaller PM sizes (Section 5.3). This ambiguity of the results may partially be attributable to the effect of other influencial variables and to the variable nature of ridges and grooves among different species of plants (Weerakkody *et al.*, 2017/Chapter 4; Weerakkody *et al.*, 2008/Section 5.1). Rough leaf surfaces with dense ridges and grooves have frequently been cited as favourable influences on PM capture and retention by many other researchers in studies relating to ground vegetation (Barima *et al.*, 2014; Kardel *et al.*, 2012; Liu *et al.* 2012; Ram *et al.,* 2012; Zhang *et al.*, 2017), although, Sæbø *et al.* (2012) did not find any correlation between leaf roughness (ridges and grooves) and PM accumulation. According to Chamberlain (1975), sticky surfaces are better at capturing coarser PM, whilst capturing of finer PM is favoured by surface roughness. The leaf sections with standardised shape and size used in Weerakkody *et al.* (2018a) (Chapter 6) also supported the positive impact of leaf roughness, by finding higher PM levels on rough leaf surfaces compared to smooth leaf surfaces. However, PM accumulation on rough-ridged leaf sections were lower compared to leaf sections excised from both hairy leaved species (*G. macrorrhizum* and *H. villosa*), suggesting a slightly higher impact of leaf hair compared to roughness created by ridges and grooves (Weerakkody *et al.*, 2018a/Chapter 6).

Similar to the findings of Liu *et al.* (2012) and Ram *et al.* (2012), stomatal density showed positive relationships with the densities of $PM_{2.5}$ and PM_{10} on the adaxial surfaces and with the density of PM_{10} on the abaxial surfaces of the leaves of species used in the Nemec wall, suggesting a more consistent impact compared to other micromorphological features (Weerakkody *et al.,* 2018b /Section 5.1). In contrast, the stomatal density of leaves of *Rubus spp.* (only found on the abaxial surfaces) did not show any significant relationship with PM accumulation. In addition to the effect of other influential variables, dense networks of elongated trichomes on the abaxial surfaces of leaves, which shield the surfaces, might have negatively influenced the effect of stomata (Section 5.3).

Although Sæbø *et al.* (2012) found a positive correlation between PM capture on leaves and the leaf-wax content, Dzierzanowski *et al.* (2011) reported that PM accumulation is not related to the depth/extent of wax but to its chemical composition and structure. In contrast to most of the findings, Liu *et al.* (2012) found a negative impact of epicuticular wax on leaf PM accumulation. Plant species with dense epicuticular wax used in this study (*J. chinensis, H. helix, T. vulgaris*) showed high or moderate PM accumulation levels. However, high PM densities found on leaves of *J. chinensis* and *T. vulgaris* and moderate PM densities on *H. helix* suggested that these differences can at least be partially explained by the differences in leaf sizes and hence, the precise impact of leaf wax on PM accumulation was not clear. Barthlott *et al.* (1998) described 23 types of cuticular wax, which were structurally varied from thin smooth films to crystalloid projections; hence, they probably have a differential impact on leaf roughness and stickiness. Therefore, to illustrate the impact of leaf wax on PM accumulation needs a comprehensive analysis on different types of wax including their structure and composition.

8.3 Elemental composition of PM, including those potentially hazardous to human health

Plant species grown on living walls in both Birmingham (Weerakkody *et al.,* 2017/Chapter 4) and Stoke-on-Trent (Weerakkody *et al.,* 2008b./Section 5.1) captured PM containing a wide range of important elements: 28 elements from the Stoke-on-Trent site and 22 elements from Birmingham. All the elements found on the plants at the Birmingham site, apart from Ni, were found in PM on plants in Stoke-on-Trent site as well. Additionally, Ag, Br, F, Sn, Tl, V and Zr were found in PM on plants in Stoke-on-Trent site. These differences in elemental composition of PM in different sites are attributed to sources of pollution (i.e. rail and road traffic), local environmental factors, meteorological conditions and anthropogenic activities in the area (Laongsri, 2012). The elemental composition of these PM were strongly correlated to PM released from rail (Abbasi *et al.*, 2013; Burkhardt *et al.*, 2008; Thornes *et al.*, 2016) and road traffic (Dong *et al.*, 2017; Lawrence *et al.*, 2013; Lin *et al.*, 2005; Sharma *et al.*, 2005) indicating that those plants can effectively immobilise traffic-generated PM. Studies that focused on the direct links between traffic-based PM and human health revealed that long-term exposure to these particles are responsible for premature mortality and detrimental health conditions such as lung cancer, cardio-pulmonary diseases, allergies (Brook *et al.*, 2010; Pascal *et al.*, 2014), neurodegenerative diseases (Maher *et al.*, 2016) and cognitive impairments (Ranft *et al.*, 2009). Therefore, removing more traffic-based PM is likely to be beneficial to human health. The most abundant element found in the particulates, C, probably mainly originates from elemental carbon and organic carbon of diesel exhaust particles (Kleeman *et al.*, 2000; Sharma *et al.*, 2005) and some from plant material. The organic component of traffic-generated PM is particularly important as it can contain potential geno-toxins and carcinogens such as PAHs (Fauser, 1999; Ji *et al.*, 2010; Maitre *et al.*, 2011). In addition to exhaust particles, a considerable proportion of hydrocarbons can be associated with non-exhaust PM (e.g. n-alkanes, benzothiazoles, n-alkanoic acids) (Pant and Harrison, 2013). Elevated levels of Fe in particulates at both sites showed that the living wall plants have a great potential to immobilise Fe-rich particles; capturing Fe in the PM_1 size-range is particularly important as they cause a serious threat to humans by creating oxidative stress when entering the brain which can lead to serious complications such as neurodegenerative diseases (Maher *et al.*, 2016).

8.4 The impact of rainfall on remobilisation of PM captured on leaves

The ability of rainfall to remobilise PM captured by plants on VGSs had not been studied prior to this research, and understanding this phenomenon is crucial in the use of evergreen plant species to capture pollution. The few studies that examined this aspect in ground vegetation, only focused on evergreen coniferous species (Przybysz *et al.*, 2014; Schaubroeck *et al.*, 2014), one broad-leaved evergreen species, and three deciduous species (Xu *et al.*, 2017) and thus there were no appropriate research findings that can be extrapolated to VGSs applications. Although the main focus of this thesis was living walls, PM remobilisation on green screens (Hedera screens) was also evaluated (Chapter 7). This was because PM remobilisation behaviour of façade greening had not been studied previously, and *H. helix* is frequently used in all types of VGSs.

The findings of this thesis showed that rainfall can wash-off a considerable fraction of PM captured on leaves of evergreen species, thus enabling them to work as effective PM sinks throughout the year. Therefore, the use of evergreen species in living walls and green screens does not hinder their ability to function as PM filters. However, the resulting inter-species variation in PM wash-off identified showed that some species (e.g. smooth glossy surfaces) can easily recover their capture surfaces by washing the particles off following a small rainfall event whereas waxy surfaces, followed by hairy surfaces, retained more particles even after 60 minutes of rain. Following exposure to 16 $mm.hr^{-1}$ rain for 60 minutes, the lowest wash-off levels were found in *H. helix* (44.3% of PM_1, 48.4% of $PM_{2.5}$ and 71.8% of PM_{10}) and more than 50% of the PM_1 and $PM_{2.5}$ were retained; this may have been because they were strongly bound to the dense epicuticular wax of *H. helix* (Terzaghi *et al.*, 2013; Dzierzanowski *et al.*, 2011; Popek *et al.*, 2013). Sixty minutes of exposure to increased rainfall intensity (41 $mm.hr^{-1}$) significantly increased the proportion of PM washed-off from *H. helix* (70.4% of PM_1, 80.3% of $PM_{2.5}$ and 84.6% of PM_{10}) leaving only a small fraction retained. However, rainfall as intense as 41 $mm.hr^{-1}$ can only be expected as an extreme weather event rather than under typical weather conditions. On the other hand, in rainy seasons rainfall durations can frequently be longer than the longest duration used here (60 minutes), which may cause higher wash-off levels compared to the estimations in this study. Although PM captured on leaf needles was also found to be washed-off by rainfall (Przybysz *et al.*, 2014), a similar or much higher PM-retaining level, as found for *H. helix*, can be expected on leaf needles of *J. chinensis* due to their thick epicuticular wax. Other species with a number of energy-dissipating structures (e.g. leaf hair/trichomes and ridges) also showed a slightly higher PM retaining rate compared to leaves with smooth surfaces. However, as leaf needles and leaves with hairy or rough surfaces were found to be accumulating significantly higher amounts of PM compared to leaves with smooth surfaces (Chapters 4, 5 and 6) use of these species is important to enhance the PM capture efficiency of living walls. In a real-world scenario, the influence of wind may have a positive impact on washing more particles off the leaves by increasing the kinetic energy of rain and by mechanical rubbing or twisting the leaves. Therefore, simulating an effect of intense rainfall with high kinetic energy (Xu *et al.*, 2017) using a watering hose or sprinkler system with a sufficient pressure, at least few times a year in non-rainy seasons, may enhance the functioning of both green screens and living walls. In situations where this approach is not technically feasible (e.g. for walls with large dimensions, lack of labour, and lack of sufficient water supply), including a large proportion of smaller leaved species with less surface roughness but higher capture efficiency (e.g. *H. albicans, B. sempervirens, H. youngii, V. vernicosa*, *B. buxifolia* and *B. x media*) and avoiding/minimising use of species with thick epicuticular wax or high surface roughness would be beneficial.

The experimental approach in this study was to estimate the PM wash-off levels comparing leaf halves and in order to provide adequate scanning samples to visualise using the ESEM, each rainfall duration was tested on a fresh set of leaves on each occasion. A further analysis using continuous rainfall on the same set of leaves and estimating the reduction at each time interval may lead to an explanation of the variation of PM wash-off over time for each species of plant. However, this will probably require a different PM quantification approach, such as use of a large number of leaves with standardised level of pollution (instead of comparing leaf-halves), in order to provide adequate scanning samples for the ESEM/imageJ approach.

8.5 An effective planting design to enhance PM capture efficiency

The findings of this thesis showed that the use of a planting design with heterogeneous topography, created by interspersing plants in different heights, has a significantly greater impact on immobilising atmospheric PM compared to a design with homogenous topography comprising plants of similar heights (Section 5.2). Similar to the literature on PM dry deposition on tree canopies (Manning and Feder., 1980; Slinn, 1982), complex topography, and greater surface roughness, on wall vegetation probably increased turbulence in the surrounding airflow resulting in higher levels of PM impaction. These findings demonstrate the importance of planting design in living wall applications to reduce PM pollution; when using multiple species, interspersing plant species with variable structural features would probably enhance their performance by increasing topographical variation.

8.6 Implications for the use of living walls in mitigating PM pollution: benefits and challenges

The research presented in this thesis revealed a great potential for living walls to immobilise atmospheric PM associated with both road and rail traffic. The living wall located along Leek Road, Stoke-on-Trent, and the living wall located adjacent to New Street Station, Birmingham, showed similarly higher performance in immobilising PM compared to the wall located inside the Staffordshire University campus, Stoke-on-Trent (with *Rubus spp.*). Based on the estimated mean PM removal ability by a 100 cm^2 area of a living wall, the highest potential for removing PM_1 ($122.08 \pm 6.9 \times 10^7$) and PM_{10} ($4.45 \pm 0.33 \times 10^7$) were shown by the living wall located along Leek road (Weerakkody *et al.,* 2008b/ Section 5.1) and the highest $PM_{2.5}$ removal potential ($9.9 \pm 5 \times 10^7$) was shown by the living wall located near the New Street station, Birmingham (Weerakkody *et al.*, 2017/Chapter 4). Even the wall with the lowest PM removal potential, located inside the University campus (the wall with *Rubus spp.*) captured a considerable amount of PM ($2.51 \pm 0.29 \times 10^6$ of PM_1, $1.3 \pm 0.18 \times 10^5$ of $PM_{2.5}$ and $2.68 \pm 0.39 \times 10^4$ of PM_{10}) (Section 5.3). However, due to the differences in location, distance to the pollution sources, pollution source, and concentration of PM in surrounding atmosphere, these values are not directly comparable (Nowak *et al.*, 2006; Sæbø *et al.*, 2012; Weber *et al.*, 2014). Weber *et al.* (2014) studied deposition of traffic-generated PM_{10} on herbaceous plants and capture levels were found to be varied between sites based on the traffic density, leaf traits, and plant height. Langner *et al.* (2011) also reported high PM deposition rates on vegetation adjacent to roads with a high traffic volume compared to the ones located on sites with low traffic volume. Although the values are not directly comparable due to site-specific variation, PM removal potential by a square metre of the living wall adjacent to Leek Road ($1.22 \pm 0.07 \times 10^{11}$ m^{-2} of PM_1, $8.24 \pm 0.72 \times 10^9$ m^{-2} of $PM_{2.5}$ and $4.45 \pm 0.33 \times 10^9$ m^{-2} of PM_{10}) was 5 to 10 times higher compared to near-road green facades with a single climbing species (*H. helix*) used in previous research 1.47×10^{10} m^{-2} of total PM (Ottele *et al.*, 2010), 2.9×10^{10} m^{-2} of $PM_{2.5}$ and PM_1 (Sternberg *et al.*, 2011) and 9.55×10^9 m^{-2} of PM_{10} and $PM_{2.5}$ (Dover and Phillips, 2015). The emission factor of a light vehicle for total PM has been estimated at 5.84×10^{13} Number. Km^{-1} (Jones and Harrison, 2006). Based on the results of the present study, a mean number of 1.34×10^{11} PM can be estimated to be captured on a square metre of the living wall adjacent to Leek Road. Focusing on the most important ultrafine PM, an hourly mean of 7.84×10^4 cm^{-3} PM has been estimated in the atmosphere in London (Schneider et al., 2015), and these levels in Stoke-on-Trent can be predicted to be lower due to the comparatively reduced pollution levels. Reflecting a great potential for living wall species to immobilise PM, an average of 3.32×10^{-4} cm^{-2}. hr^{-1} has been filtered by the leaves of the living wall located along Leek road. The overall mean PM capture potential of these living walls were estimated based on a mixture of better and poorly performing species. In some cases, PM capture levels on poorly performing species were 50 to 65 times lower than the best performing species (Weerakkody *et al.*, 2017/ Chapter 4) and, hence, use of the species with higher PM capture ability (smaller leaved species and species with complex morphology) may lead to a substantially higher PM removal potential than estimated in this thesis.

Similar to the findings of Freer-Smith *et al.* (2005), the smaller the diameter of particles the higher the number of particles being captured in all the experiments conducted in this thesis (Chapters 4 and 5). The number of PM_1 captured on leaves in some experiments was 27 times higher than the number of PM_{10} (Weerakkody *et al.,*2008b./Section 5.1). In addition to the presence of higher numbers of smaller particles in the atmosphere compared to larger particles (Sager and Castranova, 2009), leaves can be better at retaining fine particles (Przybysz *et al.*, 2014; Chapter 7). Immobilising a large number of small particles is particulaly beneficial as they are mainly anthropogenic in origin and have a higher toxicity and carcinogenic effect compared to larger particles (Manalis *et al.*, 2005).

The main challenges in the application of living wall systems are the high cost associated with installation and maintenance, and sustainability of the use of material. Perini and Rosasco (2013) carried out a cost benefit analysis of VGSs and reported that the cost of an indirect VGS using a climbing species in planter boxes is in the range of 100 - 150 €m^{-2} when plastic is used for the support structure, which can go up to 800 €m^{-2} for zinc-coated steel supports. The cost of modular living wall systems with subdivided small planters have some features of planter box systems (except that of using a single species of climbers) and mat-type living wall systems. Their costs are likely to be within the range between planter box systems and mat-type living walls and are estimated to be between 350 - 750 €m^{-2} (Dover, 2015). The estimated cost of a mat-type living wall system with pre-vegetated panel was reported to be as high as 400 -1200 €m^{-2}; in addition to the cost of material, this type of living wall is known to be associated with very high installation costs due to their pre-vegetated panels (Perini and Rosasco; 2013; Feng and Hewage, 2014). Therefore, they concluded that mat-type living walls were economically unstainable VGSs and, as a result, the use of modular living wall systems, particularly the ones with low cost design, may be more economically viable in PM filtration compared to mat-type walls.

According to Feng and Hewage (2014) and Ottele *et al.* (2011), modular living walls (only modular type living walls were used in this thesis) are mostly made up of recyclable material such as high-density polyethylene (HDPE) and are environmentally sustainable systems. Although the choice of material in mat-type (felt-based) systems have made these systems environmentally unsustainable (Feng and Hewage, 2014; Ottele *et al.,* 2011), replacing PVC with PE can probably reduce their environmental burden to a considerable extent (Feng and Hewage, 2014). The studies focused on the sustainability of living walls have drawn their conclusions by comparing the environmental burden with the environmental benefit profile of living wall systems. However, as there were no published studies conducted particularly on the ability of living walls in air purification, biodiversity conservation, and some other benefits (Chapter 1), the environmental profile has only been quantified using their energy saving and thermal performance (Feng and Hewage, 2014 and Ottele *et al.* 2011) or by extrapolating the air purification potential of green roofs to living walls (Perini and Rosasco; 2013). Therefore, the environmental benefit profiles used in these studies are probably an underestimation. Including recent findings such as reduction of PM pollution (Weerakkody *et al*, 2017; Weerakkody *et al.*, 2008b), improving biodiversity values (Chiquet, 2014), and reduction of noise pollution (Salmond *et al.*, 2016) would significantly improve the environmental benefit profile and sustainability of these systems, and this may probably easily offset the cost of these systems.

At the stage of maintenance, the service life of vegetation in modular systems was estimated at 10 years, whilst being only 3.5 years for mat walls (Ottelé *et al.*, 2011). Feng and Hewage (2014) recommended the use of vegetation with longer service life and low fertiliser consumption to reduce the number of replacements required in a long run and to minimise fertiliser usage. Plant health can be crucially important for a longer service life and for proper functioning of the living wall. Therefore, it is important to consider that some plant species have a low tolerance to environmental pollution; some species are adversely affected by PM deposition as they may interfere with important physiological processes such as photosynthesis, transpiration, and respiration (Farmer, 1993). PM can also influence the growth parameters of the plants (Gupta and Ghouse, 1987), moreover they are responsible for altering the chemical composition, anatomy, and morphology of leaves (Sharma *et al.*, 2017; Verma and Singh., 2006). Therefore, regular monitoring of plant health will be clearly warranted, but there is little information currently available on this aspect of living walls.

8.7 Recommendations for future research

Further exploration of several aspects relating to this thesis would be beneficial in enhancing the capacity of living walls in their reduction of PM pollution.

A complete life cycle analysis of a purpose-built living wall to reduce traffic-based PM, designed based on the findings of this thesis, would further explore economic benefits and sustainability aspects of living wall application, which can guide city planners and developers for large-scale implementation. A small-scale monitoring study to measure the variation of atmospheric PM concentration over time (starting from prior to living wall installation together with controls where no living wall was installed) using onsite air quality monitoring units would demonstrate the efficiency of living wall functioning as a PM filter.

The synthetic leaf models and the manipulation of natural leaf size/shape used in this study successfully simulated the effect of leaf size, shape, and overall micromorphology (Weerakkody *et al.,* 2018a). However, structure and dimensions of specific micromorphological features varied considerably between species (Weerakkody *et al.*, 2017; Weerakkody *et al.*, 2008b.) and hence, use of standardised leaf models simulating the effect of individual micromorphological features (e.g. the role of epicuticular wax) would lead to a better understanding of the role of these characters.

In evaluating PM remobilisation by rainfall, the minimum rainfall intensity that could be achieved by the techniques used in this thesis was 16 $mm.hr^{-1}$. Although this rain intensity was well within the range of natural rainfall, evaluating PM remobilisation at low rainfall intensities, and integrating the effect of wind may provide a deeper understanding of the effect of rainfall under real-world scenarios.

Developing a schematic model of particulate deposition based on the outcomes of this thesis will conceptualise the interactions between different aspects of particulate capture. A system dynamic modelling approach (e.g. using STELLA or Simile) to simulate particulate capture by living walls may enable the development of a predictive model.

8.8 Conclusion of the findings

The findings of this research revealed that living wall plants have a great potential to capture and retain atmospheric PM that are implicated in producing detrimental health effects (PM_1, $PM_{2.5}$ and PM_{10}). The elemental composition of PM captured on leaves showed a strong correlation with PM identified as being associated with road and rail traffic, emphasising the value of living walls as near-road PM traps. Nine, out of forty, species were identified as 'best performing' species while 21 species showed a moderate PM capture potential. PM accumulation on leaves was species-specific and leaf morphological characters were found to have an important impact on this process. The influence of leaf size on PM accumulation was found to be dominant over other examined characters (leaf size, shape, and micromorphology); smaller leaved species with high LAI were identified as the most efficacious plants in PM filtration. Leaf hair, surface roughness, and complex shapes also indicated a positive impact on PM capture and retention whilst elongated trichomes, which were only observed in *Rubus sp*. in this study, showed a negative impact on PM accumulation. Therefore, a thorough consideration on these specific morphological features is crucial in selecting the species to use as PM filters. The ability of simulated rainfall to remobilise captured PM on leaves of evergreen species, refreshing the capture surfaces, assures the long-term functioning of both living walls and green screens as near-road PM traps. This research further demonstrated an important positive impact of a planting design with heterogeneous topography to enhance the PM reduction ability of a living walls. Living wall systems designed with most effective PM-capturing species and effective planting designs, would substantially improve their capacity to immobilise PM and their environmental sustainability, which will potentially help in providing an effective solution to the problem of urban PM pollution and improve human wellbeing.

References

Abbasi, S., Jansson, A., Sellgren, U. and Olofsson U. (2013). Particle Emissions From Rail Traffic: A Literature Review. Critical Reviews in *Environmental Science and Technology*. 43: 2511–2544.

Abhijith, K.V., Kumar, P., Gallagher, J., McNabola, A., Baldauf, R., Pilla, F., Broderick, B., Di Sabatino, S. and Pulvirenti, B. (2017). Air pollution abatement performances of green infrastructure in open road and built-up street canyon environments – A review. *Atmospheric Environment*. 162: 71-86.

Alexandri, E. and Jones, P. (2007). Developing a one-dimensional heat and mass transfer algorithm for describing the effect of green roofs on the built environment: Comparison with experimental results. *Building and Environment*. 42: 2835–2849.

Allsop, D., Mayes, J. Moore, S. Masad, A. and Tabner, B. J. (2008). Metal- dependent generation of reactive oxygen species from amyloid proteins implicated in neurodegenerative disease. *Biochemistry Society Transactions*. 36: 1293−1298.

Anderson, H.R., Favarato, G. and Atkinson, R.W. (2013). Long-term exposure to air pollution and the incidence of asthma: meta-analysis of cohort studies. *Air Quality Atmosphere and Health*. 6:541–542.

Araujo, J.A. (2011). Particulate air pollution, systemic oxidative stress, inflammation, and atherosclerosis. *Air Quality, Atmosphere and Health.* 4: 79–93.

Atkinson, R.W., Anderson, H.R., Sunyer, J., *et al.* (2001). Acute effects of particulate air pollution on respiratory admissions: results from APHEA 2 project. Air pollution and health: a European approach. *American Journal of Respiratory and Critical Care Medicine* 164:1860-1866.

Bache, D.H. (1979). Particle transport within plant canopies-I. A framework for analysis. *Atmospheric Environment*. 13: 1257–1262.

Baldauf, R. (2017). Roadside vegetation design characteristics that can improve local, near-road air quality. Transportation Research Part D: *Transport and Environment*. 52: 354-361.

Balit, S. (2016). *A Breath of Fresh Air Addressing Climate Change and Air Pollution Together for Health*. © BMJ Publishing group Ltd.

Barima, Y.S.S., Angaman, D.M., N'Gouran, K.P., Koffi, N.A., Kardel, F., De Cannière, C. and Samson, R. (2014). Assessing atmospheric particulate matter distribution based on Saturation Isothermal Remanent Magnetization of herbaceous and tree leaves in a tropical urban environment. *Science of the Total Environment*. 470–471: 975–982.

Barros, A.P., Shrestha, P and Khlystov, A. (2010). Chemical composition and aerosol size distribution of the middle mountain range in the Nepal Himalayas during the 2009 pre-monsoon season. *Atmospheric Chemistry and Physics*. 10: 11605-11621.

Barthlott, W., Neinhuis, C., Cutler, D., Ditsch, F., Meusel, I., Theisen, I. and Wilhelmi, H. (1998). Classification and terminology of plant epicuticular waxes. *Botanical Journal of the Linnean Society.* 126: 237–260.

Becker, J.S., Bellis, D., McLeod, C.W., Dombovari, J., and Becker, J.S. (2000). Determination of trace elements including platinum in tree bark by ICP mass spectrometry. *Fresenius' Journal of* Analytical Chemistry. 368: 490-495.

Beckett, K.P., Freer-Smith, P.H. and Taylor, G. (1998). Urban woodlands: their role in reducing the effects of particulate pollution. *Environmental Pollution*. 99: 347–360.

Beckett, K.P., Freer-Smith, P.H. and Taylor, G. (2000a). Particulate pollution capture by urban trees: effect of species and windspeed. *Global Change Biology*. 6: 995–1003.

Beckett, K.P., Freer-Smith, P.H. and Taylor, G. (2000b). Effective tree species for local air-quality management. *Journal of Arboriculture*. 26:12–19.

Beeson,W.L., Abbey, D.E. and Knutsen, S.F. (1998). Long-term concentrations of ambient air pollutants and incident lung cancer in California adults: results from the AHSMOG study. *Environmental Health* Perspectives. 106:813-822

Bellouin, N., Boucher, O., Haywood, J and Reddy, M.S. (2005). Global estimate of aerosol direct radiative forcing from satellite measurements. *Nature*. 438:1138-1141.

Beentje, H. (2012). *The Kew Plant Glossary: An illustrated dictionary of plant terms*. Royal Botanic Garden. Kew.

Beerling, D.J. and Chaloner, W.G. (1993). The impact of Atmospheric CO_2 and temperature change on stomatal density: observations from *Quercus robur* Lammas leaves. *Annals of Botany*. 71: 231–235.

Berardi, U., GhaffarianHoseini, A. and GhaffarianHoseini, A. (2014). State-of-the-art analysis of the environmental benefits of green roofs. *Applied Energy.* 115:411–428.

Betts, M.G., Mitchell, D., Diamond, A.W. and Bety, J. (2007) Uneven rates of landscape change as a source of bias in roadside wildlife surveys. *Journal of Wildlife Management*. 2266-2273.

Birmili,W., Allen, A.G., Bary, F. and Harrison, R.M. (2006). Trace metal concentrations and water solubility in size-fractionated atmospheric particles and influence of road traffic. *Environmental Science and Technology.* 40: 1144-1153.

Blanc, P. (2008). *The Vertical Garden: In Nature and the City*. W. W. Norton and Company Ltd. London.

Blanusa, T., Fantozzi, F., Monaci, F. and Bargagli, R. (2015). Leaf trapping and retention of particles by holm oak and other common tree species in Mediterranean urban environments. *Urban Forestry and Urban Greening*.14: 1095–1101.

Boonyatumanond, R., Murakami, M., Wattayakorn, G., Togo, A. and Takada, H. (2007). Sources of polycyclic aromatic hydrocarbons (PAHs) in street dust in a tropical Asian mega-city, Bangkok. Thailand. *Science of the Total Environment*. 384: 420-432.

Bower, F .O. (1899). Studies in the morphology of spore-producing members. IV. The leptosporangiate ferns. *Philosophical Transactions of the Royal Society B*. 192: 29-138.

Braun-Fahrländer, C., Vuille, J., Sennhauser, F. *et al.* (1997). Respiratory health and long-term exposure to air pollutants in Swiss Schoolchildren. *American Journal of Respiratory and Critical Care Medicine*. 155: 1042–1049.

Brockhaus, A., Dolgner, R., Ewers, U., Krämer, U., Soddemann, H. and Wiegand, H. (1981). Intake and health effects of thallium among a population living in the vicinity of a cement plant emitting thallium containing dust. *International Archives of Occupational and Environmental Health*. 48: 375–389.

Bruse, M. and Fleer, H. (1998). Simulating surface-plant-air interactions inside urban environments with a three dimensional numerical model. *Environmental Modelling & Software.* 13: 373-384.

Brook, R.D., Rajagopalan, S., Pope, C.A. *et al.* (2010). Particulate Matter Air Pollution and Cardiovascular Disease. An Update to the Scientific Statement From the American Heart Association. *Circulation.*121: 2331–2378.

Burkhardt, J., Peters, K. and Crossley, A. (1995). The presence of struc- tural surface waxes on coniferous needles affects the pattern of dry deposition of fine particles. *Journal of Experimental Botany*. 46: 823-831.

Burkhardt, J. and Grantz, D.A. (2016). Plants and Atmospheric Aerosols. *Progress in Botany*. 78: 369–406.

Burkhardt, M., Rossi, L. and Boller, M. (2008). Diffuse release of environmental hazards by railways. *Desalination.* 226: 106–113.

Brunkeef, B and Holgate, S.T. (2002). Air pollution and health. *Lancet*. 360: 1233–1242.

Cadle, S.H., Mulawa, P.A., Ball, J., Donase, C., Weibel, A., Sagebiel, J.C., Knapp, K.T and Snow, R. (1997). Particulate emission rates from in-use high-emitting vehicles recruited in rrange county, California. *Environmental Science and Technology*. 31: 3405-3412.

Castelli, F., Librando, V. and Sarpietro, M.G. (2002). Calorimetric approach of the interaction and absorption of polycyclic aromatic hydrocarbons with model membranes. *Environmental Science and Technology*. 36: 2717–2723.

Chan, T., Dong, G., Leung, C., Cheung, C. and Hung, W. (2002). Validation of a two- dimensional pollutant dispersion model in an isolated street canyon. *Atmospheric Environment* 36: 861-872.

Cheetham, N., Woods, A. and Chesterton, V. (2012). *Delivering Vertical Greening*. Transport for London Surface Transport. Greater London Authority.

Chen, L., Liu, C., Zou, R., Yang, M. and Zhang, Z. (2016). Experimental examination of effectiveness of vegetation as bio-filter of particulate matters in the urban environment. *Environmental Pollution*. 208: 198–208.

Chen, L., Liu, C., Zhang, L., Zou, R. and Zhang, Z. (2017). Variation in Tree Species Ability to Capture and Retain Airborne Fine Particulate Matter (PM2.5). *Scientific Reports*. 7: 3206.

Cheng, Y., Lee, S.C., Ho, K.F., Chow, J.C., Watson, J.G., Louie, P.K.K., Cao, J.J. and Hai, X. (2010). Chemically-speciated on-road $PM_{2.5}$ motor vehicle emission factors in Hong Kong. *Science of the Total Environment.* 408:1621-1627.

Chiquet, C., Dover, J.W. and Mitchell, P. (2013). Birds and the urban environment: the value of green *walls. Urban Ecosystems. 16: 453–462.*

Chiquet, C. (2014). 'The animal biodiversity of green walls in the urban environment'. Unpublished PhD thesis. Staffordshire University, UK

Chow, J.C., Watson, J.G., Mauderly, J.L., Costa *et al.,* (2006). Health Effects of Fine Particulate Air Pollution: Lines that Connect. *Journal of the Air and Waste Management Association*. 56: 1368–1380.

Collins, R., Schaafsma, M. and Hudson, M.D. (2017). The value of green walls to urban biodiversity. *Land Use Policy.* 64:114–123.

Collins T. (2007) ImageJ for microscopy*. Biotechniques.* 43: 25–30.

Coma, J., Pérez, G., de Gracia, A., Burés, S., Urrestarazu, M. and Cabeza, L.F. (2017). Vertical greenery systems for energy savings in buildings: A comparative study between green walls and green facades. *Building and Environment*. 111: 228–237.

Chai, Z.F., Xu, D.D., Dan, M., Song, Y. and Zhuang, G.S. (2005). Concentration characteristics of extractable organohalogens in $PM_{2.5}$ and PM_{10} in Beijing, China. *Atmospheric Environment*. 39: 4119-4128.

Chamberlain, A. C. (1975). The movement of particles in plant communities. In: *Vegetation and the Atmosphere*. Academic Press: London.United Kingdom.

Chen, D., Wang, X., Thatcher, M., Barnett, G., Kachenko, A. and Prince, R. (2014). Urban vegetation for reducing heat related mortality. *Environmental Pollution*. 192: 275–284.

Chestnut, M.A., Schwartz, J., Savitz, D.A. and Burchfield, C.M. (1991). Pulmonary function and ambient particulate matter: epidemiological evidence from NHANES1. *Archives of Environmental Health*. 46:135-144.

Coward, M., Ross, D., Coward, S., Cayless, S. and Raw, G. (1996). *Pilot Study to Assess the Impact of Green Plants on NO_2 Levels in Homes.* Building Research Establishment, Watford, UK.

Croxford, B., Penn, A. and Hillier, B. (1996). Spatial distribution of urban pollution: civilizing urban traffic. *Science of the Total Environment*. 190:3-9.

Currie, B.A. and Bass, B. (2008). Estimates of air pollution mitigation with green plants and green roofs using the UFORE model. *Urban Ecosystems*. 11: 409–422.

Czemiel Berndtsson, J. C. (2010). Green roof performance towards management of runoff water quantity and quality: A review. *Ecological Engineering*. 36: 351–360.

Data.Gov.UK[Beta.] (2016). *Rainfall data.UK*. [Online]. https://data.gov.uk/data/search?q=rainfall. [Accessed 16 January 2018]

Davidson, C. and Wu, Y. L. (1990). Dry deposition of particles and vapors. In: Lindberg, S.E., Page, A.L., Norton, S.A. (Eds.), *Acidic Precipitation*. Springer, New York.

Davis, B.N.K., Brown, M.J., Frost, A.J., Yates, T.J., and Plant, R.A. (1994) The effects of hedges on spray deposition and on the biological impact of pesticide spray drift. *Ecotoxicology and Environmental Safety,* 27: 281-293.

Department for Environment Food and Rural Affairs (DEFRA). (2015). Valuing impacts on air quality: *Updates in valuing changes in emissions of oxides of nitrogen (NO_x) and concentrations of nitrogen dioxide (NO2).* DEFRA. London.

Department for Environment Food and Rural Affairs. (2015). *City of London Air Quality Strategy 2015-2020*. London. DEFRA. London.

Department for Environment Food and Rural Affairs. (2017). *Air Pollution in the UK 2016*. DEFRA. London.

Department for Transport. (2015). *Road traffic forecasts 2015*. DfT. London.

Department for Transport. (2017). *Traffic counts. AADF database.* DfT West midlands.

De Maerschalck, B., Maiheu, B., Janssen, S. and Vankerkom, J. (2010). CFD-Modelling of complex plant-atmosphere interactions: direct and indirect effects on local turbulence. *HARMO 2010 – Proceedings of International Conferecne on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes.* 839–842.

Dean, J., van Dooren, K. and Weinstein, P. (2011). Does biodiversity improve mental health in urban settings?. *Medical Hypotheses*. 76: 877–880.

Deutsch, B., Whitlow, H., Sullivan, M. and Savineau (2005). *Re-greening Washington, DC: A Green Roof Vision Based on Quantifying Storm Water and Air Quality Benefits.* CTE Fund. WASHINGTON, DC.

Dochinger, L.S. (1980). Interception of airborn particles by tree plantings. *Journal of Environmental Quality.* 9: 265–268.

Dockery, D. W. and Pope, C. A. (1994) Acute respiratory effects of particulate air pollution. Annual Review of Public Health, 15: 107-132

Dong, S., Ochoa Gonzalez, R., Harrison, R.M., Green, D., North, R., Fowler, G. and Weiss, D. (2017). Isotopic signatures suggest important contributions from recycled gasoline, road dust and non-exhaust traffic sources for copper, zinc and lead in PM_{10} in London, United Kingdom. A*tmospheric Environment.* 165: 88–98.

Dover, J.W. (2015). *Green Infrastructure: Incorporating Plants and Enhancing Biodiversity in Buildings and Urban Environments*. Routledge, Stoke-on-Trent, pp. 120–282.

Dover, J. and Phillips, S. (2015). *Particulate Pollution Capture by Green Screens along the A38 Bristol Street in Birmingham*. The Green Wall Centre, Staffordshire University. Stoke-on-Trent.

Dunnett, N. and Kingsbury, N. (2008). *Planting green roofs and living walls.* Portland/ London: Timber Press. London.

Dzierzanowski, K., Popek, R., Gawrońska, H., Saebø, A. and Gawroński, S.W. (2011). Deposition of particulate matter of different size fractions on leaf surfaces and in waxes of urban forest species*. International Journal of Phytoremediation*: 13: 1037–1046.

Ensikat, H.J., Ditsche-Kuru, P. and Barthlott, W. (2010). Scanning electron microscopy of plant *surfaces: simple but sophisticated methods for preparation and examination.* In: A. Méndez-Vilas and J. Diaz (Eds.) Microscopy: Science, Technology, Applications and Education. Formatex Research Center: Badajoz, Spain.

Environmental Protection UK. (2018). *Car Pollution Environmental Protection Uk.* [Online] http://intelligence.masterofmalt.com/car_pollution_environmental_protection_uk.pdf. [Accessed 24th January 2018]

European Environment Agency (EEA). (2016). *Air quality in Europe — 2016*. Publications Office of the European Union. Luxembourg.

Faini, F., Labbé, C. and Coll, J. (1999). Seasonal changes in chemical composition of epicuticular waxes from the leaves of Baccharis linearis. *Biochemical Systematics and Ecology*. 27: 673–679.

Fantozzi, F., Monaci, F., Blanusa, T. and Bargagli, R. (2015). Spatio-temporal variations of ozone and nitrogen dioxide concentrations under urban trees and in a nearby open area. *Urban Climate.* 12:119–127.

Farmer, A.M. (1993). The effects of dust on vegetation—a review. *Environmental Pollution*. 79: 63–75.

Farmer, J.G., Eades, L.J., Atkins, H. and Chamberlain, D.F. (2002). Historical trends in the lead isotopic composition of archival Sphagnum mosses from Scotland (1838-2000*). Environmental Science and Technology*. 36:152–157.

Ferreira, T. and Rasband, W. (2012). *ImageJ User Guide User Guide ImageJ*. Image J user Guide. 1.46r.

Fauser, P. (1999). 'Particulate Air Pollution with Emphasis on Traffic Generated Aerosols'. A PhD thesis. Kongens Lyngby: Technical University of Denmark.

Feng, H. and Hewage, K. (2014). Lifecycle assessment of living walls: air purification and energy performance. *Journal of Cleaner Production*. 69:91–99.

Feng, J. and Yang, W. (2012). Effects of particulate air pollution on cardiovascular health: A population health risk assessment. *PLoS ONE*. 7: 333-385.

Fernández, V., Sancho-Knapik, D., Guzmán, P., Peguero-Pina, J., Gil, L., Karabourniotis, G., Khayet, M., Fasseas, C., Heredia-Guerrero, J.A., Heredia, A. and Gil-Pelegrin, E. (2014). Wettability, polarity and water absorption of holm oak leaves: effect of leaf side and age. *Plant Physiology*. 166: 168–180.

Fowler, D. (2002). Pollutant deposition and uptake by vegetation. In: J. N. B. Bell & M. Treshow (eds.). *Air Pollution and Plant Life*. Chichester: Wiley, pp. 43–67.

Fowler, D., Skiba, U., Nemitz, E., Choubedar, F., Branford, D., Donovan, R. E. and Rowland, P. (2004). Measuring aerosol and heavy metal deposition on urban woodland and grass using inventories of 210PB and metal concentrations in soil. *Water, Air and Soil Pollution. 4: 483–499.*

Francis, R.A. and Lorimer, J. (2011). Urban reconciliation ecology: The potential of living roofs and walls. *Journal of Environmental Management*. 92: 1429–1437.

Franck, U., Odeh, S., Wiedensohler, A., Wehner, B. and Herbarth, O. (2011).The effect of particle size on cardiovascular disorders - the smaller the worse. *Science of the Total Environment*. 409: 4217–21.

Freer-Smith, P.H; El-khatib, A.A and Taylor, G. (2003). Capture of Particulate Pollution By Trees : a Comparison of Species Typical of Semi-Arid Areas and North American Species. *Water, Air, and* Soil Pollution.155:173–187.

Freer-Smith, P.H., El-Khatib, A.A. and Taylor, G. (2004). Capture of particulate pollution by trees: a comparison of species typical of semi-arid areas (Ficus nitida and Eucalyptus globulus) with European and north American species. *Water Air and Soil Pollution*. 155: 173–187.

Freer-Smith, P.H., Beckett, K.P. and Taylor, G. (2005). Deposition velocities to *Sorbus aria, Acer campestre, Populus deltoides* × *trichocarpa 'Beaupré', Pinus nigra and* × *Cupressocyparis leylandii* for coarse, fine and ultra-fine particles in the urban environment. *Environmental Pollution*. 133:157–167.

Fritschen, L.J. and Edmonds, R. (1976). *Dispersion of fluorescent particles into and within a douglas fir forest.* In: Symposium on Atmos- phere--Surface Exchange of Particulate and Gaseous Pollutants. Energy Research & Development Administrative Symposium Series No. 38, USA.

Fuchs, N.A. (1964). *The Mechanics of Aerosols.* Pergamon Press, New York.

Gago, E.J., Roldan, J., Pacheco-Torres, R. and Ordez, J. (2013). The city and urban heat islands: A review of strategies to mitigate adverse effects. Renew. *Sustainable Energy Reviews*. 25: 749–758.

Gallagher, J., Baldauf, R., Fuller, C.H., Kumar, P., Gill, L.W. and McNabola, A. (2015). Passive methods for improving air quality in the built environment: A review of porous and solid barriers. *Atmospheric Environment.* 120: 61–70.

Gallagher, J., Gill, L.W. and McNabola, A. (2012). Numerical modelling of the passive control of air pollution in asymmetrical urban street canyons using refined mesh discretization schemes. *Building and Environment*. 56: 232–240

Gallagher, J., Gill, L.W. and McNabola, A. (2013). The passive control of air pollution exposure in Dublin, Ireland: A combined measurement and modelling case study. *Science of the Total Environment.* 458–460: 331–343.

.Gautam, P., Blaha, U. and Appel, E. (2005). Magnetic susceptibility of dust-loaded leaves as a proxy of traffic-related heavy metal pollution in Kathmandu city, Nepal. *Atmospheric Environment*. 39: 2201–2211.

Gehring, U., Wijga, A.H., Brauer, M., Fischer, P., de Jongste, J.C., Kerkhof, M., Oldenwening, M., Smit, H.A. and Brunekreef, B. (2010). Traffic-related Air Pollution and the Development of Asthma and Allergies during the First 8 Years of Life. *American Journal of Respiratory and Critical Care Medicine.* 181:596–603.

Gemba, K. (2007). *Shape effects on drag.* Department of Aerospace Engineering, California State University.

Gillies, J.A., Nickling, W.G. and King, J. (2002). Drag coefficient and plant form response to wind speed in three plant species: Burning Bush (*Euonymus alatus*), Colorado Blue Spruce (*Picea pungens glauca*), and Fountain Grass (*Pennisetum setaceum*). *Journal of Geophysical Research Atmospheres.* 107: 1–15.

Gilmour, P.S., Brown, D.M., Lindsay, T.G., Beswick, P.H., MacNee, W. and Donaldson, K. (1996). Adverse health effects of PM_{10} particles: involvement of iron in generation of hydroxyl radical. *Occupational and environmental medicine*. 53: 817–822.

Godzik, S., Florkowski, T., Piorek, S. and Sassen, M.M.A. (1979). An attempt to determine the tissue contamination of *Quercus robur L*. and *Pinus sylvestris L.* foliage by particulates from zinc and lead smelters. *Environmental Pollution.* 18: 97–106

GOOGLE MAPS. (2016). Map of Birmingham New Street Station. [Online]. https://www.google.com/maps/d/viewer?mid=1bbpvY39gTk2kBo2GAJU6s9rtObYandhl=enandll=52.47786350006138%2C-1.896131500000024andz=17 [Accessed 13 February 2016].

Gower, S.T., Kucharik, C.J. and Norman, J.M. (1999). Direct and Indirect Estimation of Leaf Area Index, fAPAR, and Net Primary Production of Terrestrial Ecosystems. *Remote Sensing of Environment.* 70: 29-5.

Gowers, A.M., Miller, B.G. and Stedman, J.R. (2014). *Estimating Local Mortality Burdens associated with Particulate Air Pollution.* Public Health England. Oxfordshire.

Gray, H.A., Cass, G.R., Huntzicker, J.J., Heyerdahl, E.K. and Rau, J.A. (1986). Characteristics of atmospheric organic and elemental carbon particle concentrations in Los-Angeles. *Environmental Science and Technology*. 20: 580-589

Gregory, P.H. (1973). *The Microbiology of the Atmosphere*. Clarke, Doble and Brendon, Plymouth.

Grochowicz, E. and Korytkowski, J. (1996). *Air protection.* Polish Educational Publisher. Poland. 2: 17.

Gromke, C. and Ruck, B. (2009). On the impact of trees on dispersion processes of traffic emissions in street canyons. *Boundary-Layer Meteorology*. 131:19–34.

Gromke, C., Jamarkattel, N. and Ruck, B. (2016). Influence of roadside hedgerows on air quality in urban street canyons. *Atmospheric Environment*. 139: 75–86.

Guerreiro, C., González Ortiz, A. and Frank de Leeuw (2017). *Air quality in Europe - 2017*. Publications Office of the European Union. Denmark: Luxembourg.

Gupta, M.C., and Ghouse, A.K.M. (1987). The effect of coal smoke pollutants on growth yield and leaf epidermis features of Abelmoschus esculenus Moench. *Environmental Pollution*. 43:263–270.

Gustafson, W.I., Qian, Y. and Fast, J.D. (2011). Downscaling aerosols and the impact of neglected subgrid processes on direct aerosol radiative forcing for a representative global climate model grid spacing. *Journal of Geophysical Research: Atmospheres*.116.

Ha E.H., Lee, J.T., Kim, H., Hong, Y.C., Lee, B.E. and Park, H.S., *et al.* (2003). Infant susceptibility of mortality to air pollution in Seoul, South Korea. *Pediatrics*. 111:284–290.

Halonen, J.I., Lanki, T., Yli-Tuomi, T., Kulmala, M., Tiittanen, P., Pekkanen, J. (2008). Urban air pollution, and asthma and COPD hospital emergency room visits. *Thorax*. 63: 635-641.

Han, S. Q., Bian, H., Zhang, Y. F.,Wu, J.H.,Wang, Y.M., Tie, X. X. and Yao, Q. (2012). Effect of aerosols on visibility and radiation in spring 2009 in Tianjin, China. *Aerosol and Air Quality* Research.12: 211–217.

Harrison, R.M. and Yin, J.X. (2000). Particulate matter in the atmosphere: which particle properties are important for its effects on health?. *Science of the Total Environment*. 249: 85–101.

Harrison, R.O.Y.M. & Jones, A.M. (n.d.). A MULTI-SITE STUDY OF PROCESSES INFLUENCING PARTICLE NUMBER CONCENTRATIONS IN URBAN AIR Division of Environmental Health & Risk Management School of Geography , Earth and Environmental Sciences.

Hennebo, D. (1955). *Staubfilterung durch Grünanlagen*. – VEB Verlag Technik, Berlin, 79 pp.

Hickey, M. and King, C. (2000). *The Cambridge Illustrated Glossary of Botanical Terms*. Cambridge University Press, New York, pp. 105–109.

Hofman, J., Wuyts, K., Van Wittenberghe, S. and Samson, R. (2014). On the temporal variation of leaf magnetic parameters: Seasonal accumulation of leaf-deposited and leaf-encapsulated particles of a roadside tree crown. *Science of the Total Environment*. 493: 766–772.

Hofman, J., Staelens, J., Cordell, R., Stroobants, C., Zikova, N., Hama, S.M.L., Wyche, K.P., Kos, G.P.A., Van Der Zee, S., Smallbone, K.L., Weijers, E.P., Monks, P.S. and Roekens, E. (2016). Ultrafine particles in four European urban environments: Results from a new continuous long-term monitoring network. *Atmospheric Environment*. 136: 68–81.

Holzinger, R., Kasper-Giebl, A., Staudinger, M., Schauer, G. and Rockmann, T. (2010). Analysis of the chemical composition of organic aerosol at the Mt. Sonnblick observatory using a novel high mass resolution thermal-desorption proton- transfer-reaction mass-spectrometer (hr-TD-PTR-MS). *Atmosphric Chemistry and Physics*. 10: 10111-10128.

Hwang, H.J., Yook, S.J. and Ahn, K.-H. (2011). Experimental investigation of submicron and ultrafine soot particle removal by tree leaves. *Atmospheric Environment.* 45: 6987–6994.

Ingold, C.T. (1971). *Fungal spores*. Clarendon Press, Oxford.

International Agency for Research on Cancer (IARC) (2015). *Outdoor air pollution. Monographs on the evaluation of carcinogenic risks to humans*. IARC Press. Lyon.

Hyun Cho, S., Haiyan, T., John, K.M., Baldauf, R.W., Krantz, Q.T. and Gilmour, M.I. (2005). Comparative toxicity of size-fractionated airborne particulate matter collected at different distances from an urban highway. *Environmental Health Perspectives*. 117: 1682–1689.

Intergovernmental Panel for Climate change (IPCC). (2013). *Assessment report of the intergovernmental panel on climate change*. Cambridge University Press, Cambridge, UK

Impens, R.A. and Delcarte, E. (1979). Survey of urban trees in Brussels. Belgium. *Journal of Arboriculture.* 5:169-176.

Janhäll, S. (2015). Review on urban vegetation and particle air pollution – Deposition and dispersion. *Atmospheric Environment*. 105: 130–137.

Jayasooriya, V.M., Ng, A.W.M., Muthukumaran, S. and Perera, B.J.C. (2017). Green infrastructure practices for improvement of urban air quality. *Urban Forestry and Urban Greening*. 21: 34–47.

Jepson, P. (2016). A rewilding agenda for Europe: creating a network of experimental reserves. *Ecography*. 39: 117–124.

Ji, G.X., Gu, A.H., Zhou, Y., Shi, X.G., Xia, Y.K., Long, Y., Song, L.,Wang, S.L. and Wang, X.R. (2010). Interactions between exposure to environmental polycyclic aromatic hydrocarbons and DNA repair gene polymorphisms on bulky DNA adducts in human sperm. *Plos One.* 5: 13145

Jin, S., Guo, J., Wheeler, S., Kan, L. and Che, S. (2014). Evaluation of impacts of trees on $PM_{2.5}$ dispersion in urban streets. *Atmospheric Environment*. 99:277-287.

Johnston, J. and Newton, J. (2004). *Building Green A Guide to Using Plants on Roofs, Walls and Pavements.* Greater London Authority. London. pp. 121.

Jones, A. and Harrison, R. (2006). Estimation of the emission factors of particle number and mass fractions from traffic at a site where mean vehicle speeds vary over short distances. *Atmospheric Environment*. 40: 7125-7137.

Karagulian, F., Belis, C.A., Dora, C.F.C., Prüss-Ustün, A.M., Bonjour, S., Adair-Rohani, H. and Amann, M. (2015). Contributions toh cities' ambient particulate matter (PM): A systematic review of local source contributions at global level. *Atmospheric Environment*. 120: 475–483.

Karanasiou, A., Sitaras, I., Siskos, P. and Eleftheriadis, K. (2007). Size distribution and sources of trace metals and n-alkanes in the Athens urban aerosol during summer. *Atmospheric Environment.* 41: 2368-2381.

Kardel, F., Wuyts, K., Maher, B.A., Hansard, R. and Samson, R. (2011). Leaf saturation isothermal remanent magnetization (SIRM) as a proxy for particulate matter monitoring: Inter-species differences and in-season variation. *Atmospheric Environment.* 45: 5164–5171.

Kardel, F., Wuyts, K., Maher, B. a. and Samson, R. (2012). Intra-urban spatial variation of magnetic particles: Monitoring via leaf saturation isothermal remanent magnetisation (SIRM). *Atmospheric Environment*. 55: 111–120.

Karley, A. J., Mitchell . C., Brookes, C., McNicol, J., O'Neill, T., Roberts, H., Graham. J., Johnson, S.N. (2016). Exploiting physical defence traits for crop protection: Leaf trichomes of Rubus idaeus have deterrent effects on spider mites but not aphids. *Annals of Applied Biology*. 168: 159–172.

Karottki, D.G., Bekö, G., Clausen, G., Madsen, A.M., Andersen, Z.J., Massling, A., Ketzel, M., Ellermann, T., Lund, R., Sigsgaard, T., Møller, P. and Loft, S. (2014). Cardiovascular and lung function in relation to outdoor and indoor exposure to fine and ultrafine particulate matter in middle-aged subjects. *Environment international.* 73: 372–381.

EEA, F.J. and Fussell, J.C. (2012). Size, source and chemical composition as determinants of toxicity attributable to ambient particulate matter. *Atmospheric Environment.* 60: 504–526.

Kim, K.J., Kil, M.J., Song, J.S., Yoo, E.H., Son, K.C., and Kays, S.J. (2008) Efficiency of volatile formaldehyde removal by indoor plants: contribution of aerial plant parts versus the root zone. *Journal of the American Society for Horticultural Science*. 133: 521-526.

Kleeman, M.J., Schauer, J.J. and Cass, G.R. (2000). Size and composition distribution of fine particulate matter emitted from motor vehicles. *Environmental Science and Technology*. 34: 1132–1142.

Köhler, M. (2008). Green facades-a view back and some visions. *Urban Ecosystems.* 11: 423–436.

Kreyling, W.G., Hirn, S. and Schleh, C. (2010). Nanoparticles in the lung. *Nature Biotechnology.* 28:1275–1276

Künzli, N., Kaiser, R., Medina, S., Studnicka, M., Chanel, O., Filliger, P., Herry, M., Horak, F., Puybonnieux-Texier, V., Quénel, P., Schneider, J., Seethaler, R., Vergnaud, J.C. and Sommer, H. (2000). Public-health impact of outdoor and traffic-related air pollution: a European assessment. *Lancet.* 356:795–801.

Laden, F., Schwartz, J., Speizer, F.E. and Dockery, D.W. (2006). Reduction in fine particulate air pollution and mortality: Extended follow-up of the Harvard Six Cities study. *American Journal of Respiratory and Critical Care Medicine*.173: 667–72.

Lafortezza R, Davies C, Sanesi G and Konijnendijk CC, (2013). Green Infrastructure as a tool to support spatial planning in European urban regions. *iForest.* 6: 102-108.

Lamaud, E., Chapuis, A., Fontan, J. and Serie, E. (1994). Measurements and parameterisation of aerosol dry deposition in a semi-arid area. *Atmospheric Environment*. 28: 2461–2472.

Langner, M., Kull, M. and Endlicher, W.R. (2011). Determination of PM10 deposition based on antimony flux to selected urban surfaces. *Environmental Pollution*. 159: 2028–2034.

Laongsri, B. (2012). 'Studies of the properties of particulate matter in the UK atmosphere'. A PhD thesis. University of Birmingham, UK.

Lawrence, S., Sokhi, R., Ravindra, K., Mao, H., Prain, H.D. and Bull, I.D. (2013). Source apportionment of traffic emissions of particulate matter using tunnel measurements. *Atmospheric Environment.* 77: 548–557.

Lee, J.H. and Sim, W.K. (1999). 'Biological absorption of SO_2 by Korean native indoor species'. In: Burchett, M.D., *et al.* (Eds.), *Towards a New Millennium in People- plant Relationships, Contributions from International People-plant Symposium*", Sydney. pp. 101-108.

Legg, B.J. and Powell, F.A., (1979). Spore dispersal in barley crop: a mathematical model. *Agricultural Meteorology.* 20: 47–67.

Leonard, R.J., McArthur, C. and Hochuli, D.F. (2016). Particulate matter deposition on roadside plants and the importance of leaf trait combinations. *Urban Forestry and Urban Greening*. 20: 249–253.

Lohr, V.I. and Pearson-Mims, C. (1996). Particulate matter accumulation on horizontal surfaces in interiors: influence of foliage plants. *Atmospheric Environment*. 14: 2565-2568.

Lombaert, K., Morel, S., Le Moyne, L., Adam, P., De Maleissye, J.T. and Amouroux, J. (2004). Non-destructive analysis of metallic elements in diesel soot collected on filter: Benefits of laser induced breakdown spectroscopy. *Plasma Chemistry and Plasma Processing*. 24: 41–56.

Li, X., Wang, L., Wang, Y., Wen, T., Yang, Y., Zhao, Y. and Wang, Y. (2012). Chemical composition and size distribution of airborne particulate matters in Beijing during the 2008 Olympics. *Atmospheric Environment*. 50: 278–286.

Liang, J., Fang, H.L., Zhang, T.L., Wang, X.X. and Liu, Y.D. (2017). Heavy metal in leaves of twelve plant species from seven different areas in Shanghai, China. *Urban Forestry and Urban Greening*. 27: 390-398.

Lin, C.c., Chen, S.J and Huang, K.L. (2005). Characteristics of Metals in Nano/Ultrafine/Fine/Coarse Particles Collected Beside a trafficked road. *Environmental Science and Technology.* 39:8113–8122.

Litschke, T. and Kuttler, W. (2008). On the reduction of urban particle concentration by vegetation - a review. *Meteorologische Zeitschrift*. 17: 229–240.

Liu, L., Guan, D. and Peart, M.R. (2012). The morphological structure of leaves and the dust-retaining capability of afforested plants in urban Guangzhou, South China. *Environmental Science and Pollution Research*. 19:3440–3449.

Macmillan, G. (2004). *York University Rooftop Garden Stormwater Quantity and Quality Performance Monitoring Report*. Greening Rooftops for Sustainable Communities. Portland.

Madders, M. and Lawrence, M.I.G. (1981). The role of woodland in air pollution control. *Quarterly Journal of Forestry. 76: 256-261.*

Maher, B.A., Ahmed, I.A.M., Davison, B., Karloukovski, V. and Clarke, R. (2013). Impact of Roadside Tree Lines on Indoor Concentrations of Traffic- Derived Particulate Matter. *Environmental science and Technology.* 47:13737–13744.

Maher, B.A., Ahmed, I.A.M., Karloukovski, V., MacLaren, D.A., Foulds, P.G., Allsop, D., Mann, D.M.A., Torres-Jardón, R. and Calderon-Garciduenas, L. (2016). 'Magnetite pollution nanoparticles in the human brain'. Proceedings of the National Academy of Sciences of the United States of America. 113:797–801.

Maher, B.A., Moore, C. and Matzka, J. (2008). Spatial variation in vehicle-derived metal pollution identified by magnetic and elemental analysis of roadside tree leaves. *Atmospheric Environment*. 42: 364–373.

Maitre, A., Tarantini, A., Lefebvre, E., Marques, M., Rajhi, A. and Douki, T. (2011). Polycyclic aromatic hydrocarbons in binary mixtures modulate the efficiency of benzo[a] pyrene to form DNA adducts in human cells. *Toxicology*. 279: 36-44

Manalis, N., Grivas, G., Protonotarios, V., Moutsatsou, A., Samara, C. and Chaloulakou, A. (2005). Toxic metal content of particulate matter (PM_{10}), within the Greater Area of Athens. *Chemosphere.* 60:557–566.

Manning, W. J., and W. A. Feder. (1980). *Biomonitoring Air Pollutants with Plants*. Applied Science Publishers. London. pp 142.

Manoli, E., Voutsa, D. and Samara, C. (2002). Chemical characterization and source identification/apportionment of fine and coarse air particles in Thessaloniki, Greece. *Atmospheric Environment*. 36: 949-961.

Manso, M. and Castro-Gomes, J. (2015). Green wall systems: A review of their characteristics. *Renewable and Sustainable Energy Reviews*. 41: 863–871.

Manso, M. and Castro-Gomes, J.P. (2016). Thermal analysis of a new modular system for green walls. *Journal of Building Engineering*. 7: 53–62.

Mårtensson, L.M., Fransson, A.M. and Emilsson, T. (2016). Exploring the use of edible and evergreen perennials in living wall systems in the Scandinavian climate. *Urban Forestry and Urban Greening.* 15:84–88.

Matthews, T., Lo, A.Y. and Byrne, J.A. (2015). Reconceptualizing green infrastructure for climate change adaptation: Barriers to adoption and drivers for uptake by spatial planners. *Landscape and . Urban Planing*. 138:155–163.

Matzka, J. and Maher, B.A. (1999). Magnetic biomonitoring of roadside tree leaves: identification of spatial and temporal variations in vehicle- derived particulates. *Atmospheric Environment.* 33: 4565–4569.

McDonald, A.G., Bealey, W.J., Fowler, D., Dragosits, U., Skiba, U., Smith, R.I., Donovan, R.G., Brett*,* H.E., Hewitt, C.N. and Nemitz, E. (2007). Quantifying the effect of urban tree planting on concentrations and depositions of PM_{10} in two UK conurbations. *Atmospheric Environment*.41:8455–8467.

McPherson, E.G., Nowak, D.J. and Rowntree, R.A. (1994). *Chicago's Urban Forest Ecosystem: Results of the Chicago Urban Forest Climate Project.* US Deprtment of Agriculture.

Mir, M. A. (2011). 'Green Facades and Building Structures'. A MSc thesis. Delft University of Technology.

Mishra, A., Jain, K. and Garg, K. (1995). Role of higher plants in the deterioration of historic buildings. *Science of the Total Environment*. 167:375-392.

Mitchell, R. and Maher, B.A. (2009). Evaluation and application of biomagnetic monitoring of traffic-derived particulate pollution. *Atmospheric Environment*. 43: 2095–2103.

Mo, L., Ma, Z., Xu, Y., Sun, F., Lun, X., Liu, X., Chen, J. and Yu, X. (2015). Assessing the capacity of plant species to accumulate particulate matter in Beijing, China. *PLoS ONE*. 10:10.

Mori, J., Hanslin, H.M., Burchi, G. and Sæbø, A. (2015). Particulate matter and element accumulation on coniferous trees at different distances from a highway. *Urban Forestry and Urban Greening.* 14: 170–177.

Morris, W.A., Versteeg, J.K., Bryant, D.W., Legzdins, A.E., McCarry, B.E. and Marvin, C.H. (1995). Preliminary comparisons between mutagenicity and magnetic-susceptibility of respirable airborne particulate. *Atmospheric Environment.* 29: 3441–3450

Mouga. T. and Almeida M. (1997). Neutralization of herbicides. Effects on wall vegetation. *International Biodeterioration and Biodegradation.* 40:141-149.

Mulawa, P.A., Cadle, S.H., Knapp, K., Zweidinger, R., Snow, R., Lucas, R. and Goldbach, J. (1997). Effect of ambient temperature and E-10 fuel on primary exhaust particulate matter emissions from light-duty vehicles*. Environmental Science and Technology.* 31:1302–1307.

Natural England. (2009). *Green infrastructure guidance Report* NE.176. pp.1–107.

Naumann, S., Davis. Mc., Kaphengst, T., Pieterse, M. and Rayment, M. (2011*). Design, implementation and cost elements of Green Infrastructure projects.* Final report Brussels,European Commission.

Neinhuis, C. and Barthlott,W. (1998). Seasonal changes of leaf surface contamination in beech, oak, and ginkgo in relation to leaf micromorphology and wettability. *New Phytologist*. 138:91–98.

Nowak, D.J. (1994). *Air pollution removal by Chicago's urban forest.* General technical report. United States Department of Agriculture, Forest Service, Northeastern Forest Experimental Station. pp. 63–81.

Nowak, D.J., Crane, D.E. and Stevens, J.C. (2006). Air pollution removal by urban trees and shrubs in the united States. *Urban Forestry and Urban Greening*. 4:115–123.

Nowak, D.J., Hirabayashi, S., Bodine, A. and Hoehn, R. (2013). Modeled $PM_{2.5}$ removal by trees in ten U.S. cities and associated health effects. *Environmental Pollution.* 178:395–402.

Oberdorster, G., Ferin, J., Lehnert, B. (1994). Correlation between particle size, in vivo particle persistence, and lung injury. *Environmental Health Perspectives.* 102:173-179.

Onishi, A., Cao, X., Ito, T., Shi, F. and Imura, H. (2010). Evaluating the potential for urban heat-island mitigation by greening parking lots. *Urban Forestry and Urban Greening*. 9:323–332.

Ottelé, M., van Bohemen, H.D. and Fraaij, A.L.A. (2010). Quantifying the deposition of particulate matter on climber vegetation on living walls. *Ecological Engineering.* 36: 154–162.

Ottelé, M., Perini, K., Fraaij, A.L.A., Haas, E.M. and Raiteri, R. (2011). Comparative life cycle analysis for green facades and living wall systems. *Energy and Buildings*. 43: 3419–3429.

Ould-Dada, Z. (2002). Dry deposition profile of small particles within a model spruce canopy. *The Science of the Total Environment.* 286: 83–96

Oxford Instruments. (2006). *INCA Energy Operator Manual.* Issue 2.1. Abingdon.

Pant, P. and Harrison, R.M. (2013). Estimation of the contribution of road traffic emissions to particulate matter concentrations from field measurements: A review. *Atmospheric Environment*. 77:78–97.

Pakzad, P. and Osmond, P. (2016). Developing a Sustainability Indicator Set for Measuring Green Infrastructure Performance. *Procedia - Social and Behavioral Sciences.* 216: 68–79.

Parkes,W.R. (1983). *Inhaled Particles and Their Fate in the Lungs, Occupational Lung Disorders*. second edition. Butterworths, Boston.

Parker, J.D., Akinbami, L.J. and Woodruff, T.J. (2009). Air pollution and childhood respiratory allergies in the United States. *Environmental Health Perspect*ives. 117:140–147.

Pascal, M., Falq, G., Wagner, V., Chatignoux, E., Corso, M., Blanchard, M., Host, S., Pascal, L. and Larrieu, S. (2014). Short-term impacts of particulate matter (PM_{10}, $PM_{10\text{-}2.5}$, $PM_{2.5}$) on mortality in nine French cities. *Atmospheric Environment*. 95: 175–184.

Pauchard, A. Aguayo, M. Pena, E. and Urrutia, R. (2006). “Multiple Effects of Urbanization on the Biodiversity of Developing Countries: The Case of a Fast-growing Metropolitan Area (Concepcion, Chile),” *Bio- logical Conservation*. 127: 272–281.

Perez, G., Rincon, L., Vila, A., Gonzalez, J.M. and Cabeza, L.F. (2011) Behaviour of green facades in Mediterranean Continental climate. *Energy Conversion and Management*. 52: 1861–1867.

Pérez-Urrestarazu, L., Fernández-Cañero, R., Franco-Salas, A. and Egea, G. (2015). Vertical Greening Systems and Sustainable Cities. *Journal of Urban Technology*. 22: 65–85.

Perini, K., Ottele, M., Fraaij, A.L.A., Haas, E.M. and Raiteri, R.(2011). Vertical greening systems and the effect on airflow and temperature on the building envelope. *Building and Environment*. 46: 2287–2294.

Perini, K., Ottelé, M., Giulini, S., Magliocco, A. and Roccotiello, E. (2017). Quantification of fine dust deposition on different plant species in a vertical greening system. *Ecological Engineering.* 100: 268–276.

Perini, K., Ottelé, M., Haas, E.M. and Raiteri, R. (2012). Vertical greening systems, a process tree for green façades and living walls. *Urban Ecosystems*. 16: 265-277

Perini, K. and Rosasco, P. (2013). Cost-benefit analysis for green façades and living wall systems. *Building and Environment*. 70:110–121.

Petroff, A., Mailliat, A., Amielh, M. and Anselmet, F. (2008a). Aerosol dry deposition on vegetative canopies. Part I: Review of present knowledge. *Atmospheric Environment*. 42: 3625–3653.

Petroff, A., Mailliat, A., Amielh, M. and Anselmet, F. (2008b). Aerosol dry deposition on vegetative canopies. Part II: A new modelling approach and applications. *Atmospheric Environment*. 42: 3654–3683.

Petroff, A., Zhang, L., Pryor, S.C. and Belot, Y. (2009). An extended dry deposition model for aerosols onto broadleaf canopies. *Journal of Aerosol Science*. 40:218–240.

Pierse, N., Rushton, L., Harris, R.S., Kuehni, C.E., Silverman, M. and Grigg, J., (2006). Locally generated particulate pollution and respiratory symptoms in young children. *Thorax*. 61:216-220.

Pilinis, C. and Pandis, S. N. (1995). Physical, Chemical and Optical Properties of Atmospheric Aerosols. In *Airborne Particulate Matt*er, Volume D, edited by Kouimtzis, T. and C. Samara, Springer, 100-124.

Pio, C., Mirante, F., Oliveira, C., Matos, M., Caseiro, A., Oliveira, C., Querol, X., Alves, C., Martins, N., Cerqueira, M. and Camoes, F. (2013). Size-segregated chemical composition of aerosol emissions in an urban road tunnel in Portugal. *Atmospheric Environment*. 71:15-25.

Pope III, C.A., Dockery, D.W., Spengler, J.D. and Raizenne, M.E. (1991). Respiratory health and PM_{10} pollution. *The American Review of Respiratory Disease*. 144: 668-674.

Pope III, C.A. and Kanner, R.E. (1993). Acute effects of PM_{10} pollution on pulmo- nary function of smokers with mild to moderate chronic obstructive pulmonary disease. *American Review of Respiratory Disease*. 147:1336-1340.

Pope III, C.A. (2002). Lung Cancer, Cardiopulmonary Mortality, and Long-term Exposure to Fine Particulate Air Pollution. *JAMA*. 287: 1132-1141.

Pope III, C.A., Brook, R.D., Burnett, R.T. and Dockery, D.W. (2011). How is cardiovascular disease mortality risk affected by duration and intensity of fine particulate matter exposure? An integration of the epidemiologic evidence. *Air Quality, Atmosphere and Health.* 4: 5–14.

Popek, R., Gawrońska, H., Wrochna, M., Gawroński, S.W. and Sæbø, A. (2013). Particulate matter on foliage of 13 woody species: deposition on surfaces and phytostabilisation in waxes – a 3-Year Study. *International Journal of Phytoremediation*. 15:245–256.

Prajapati, S.K. and Tripathi, B.D. (2008). Seasonal variation of leaf dust accumulation and pigment content in plant species exposed to urban particulates pollution. *Journal of Environmental Quality*. 37:865–870.

Prusty, B.A.K., Mishra, P.C. and Azeez, P.A. (2005). Dust accumulation and leaf pigment content in vegetation near the national highway at Sambalpur, Orissa, India. *Ecotoxicology and Environmental Safety*. 60: 228–235,

Przybysz, A., Sæbø, A., Hanslin, H.M. and Gawronski, S.W. (2014). Accumulation of particulate matter and trace elements on vegetation as affected by pollution level, rainfall and the passage of time. *Science of the Total Environment. 481: 360-369.*

Pugh, T.A.M., Mackenzie, A.R., Whyatt, J.D. and Hewitt, C.N. (2012). Effectiveness of Green Infrastructure for Improvement of Air Quality in Urban Street Canyons. *Environmental Science and Technology*. 46:7692-7699.

Pye, K. (1987). *Aeolian Dust and Dust Deposits*. Cambridge University Press, Cambridge. pp. 29-62.

Qiu, Y., Guan, D., Song, W. and Huang, K. (2009). Capture of heavy metals and sulphur by foliar dust in urban Huizhou, Guangdong Province, China. *Chemosphere.* 75: 447–452.

R Development Core Team (2016). R: *A language and environment for statistical computing. R Foundation for Statistical Computing*, Vienna, Austria.

Raaschou-Nielsen, O., Beelen, R., Wang, M. et al. (2015). Particulate matter air pollution components and risk for lung cancer. *Environmental International*. 87: 66–73.

Raaschou-Nielsen, O., Pedersen, M., Stafoggia, M. *et al.* (2017). Outdoor air pollution and risk for kidney parenchyma cancer in 14 European cohorts. *International Journal of Cancer*. 140: 1528–1537.

Raizenne, M., Neas, L.M., Damokosh, A.I., Dockery, D.W., Spengler, J.D., Koutrakis, P., Ware, J.H. and Speizer, F.E. (1996). Health effects of acid aerosols on North American children: pulmonary function. *Environmental Health Perspectives*.104:506-514.

Ram, S.S., Majumder, S., Chaudhuri, P., Chanda, S., Santra, S.C., Maiti, P.K., Sudarshan, M. and Chakraborty, A. (2012). Plant canopies: bio-monitor and trap for re-suspended dust particulates contaminated with heavy metals. *Mitigation and Adaptation Strategies for Global Change*. 19:499–508.

Ranft, U., Schikowski, T., Sugiri, D., Krutmann, J. and Krämer, U. (2009). Long-term exposure to traffic-related particulate matter impairs cognitive function in the elderly. *Environmental Research*. 109: 1004–1011.

Randrup, T.B., McPherson, E.G. and Costello, L.R. (2001). Tree root intrusion in sewer systems: review of the extent and costs. *Journal of Infrastructure Systems*. 7:26–31.

Räsänen, J. V., Holopainen, T., Joutsensaari, J., Ndam, C., Pasanen, P., Rinnan, Å. and Kivimäenpää, M. (2013). Effects of species-specific leaf characteristics and reduced water availability on fine particle capture efficiency of trees. *Environmental Pollution.*183: 64–70.

Ries, K. and Eichhorn, J. (2001). Simulation of effects of vegetation on the dispersion of pollutants in street canyons. *Meteorologische Zeitschrift*. 10: 229–233.

Riddle, S.G., Robert, M.A., Jakober, C.A., Fine, P.M., Hays, M.D., Schauer, J.J. and Hannigan, M.P. (2009). Source Apportionment of Fine Airborne Particulate Matter during a Severe Winter Pollution Episode. *Environmental Science and Technology*. 43:272–279.

Roehr, D. and Laurenz, J. (2008) Living skins: environmental benefits of green envelopes in the city context. In *Eco- Architecture: Harmonisation between Architecture and Nature.* WIT press, Boston. Pp: 149–158

Roy, S., Byrne, J. and Pickering, C. (2012). A systematic quantitative review of urban tree benefits, costs, and assessment methods across cities in different climatic zones. *Urban Forestry and Urban Greening.* 11:351–363.

Royal College of Physicians. (2016) *Every breath we take: the lifelong impact of air pollution*. Report of a working party. RCP. London.

Roupsard, P., Amielh, M., Maro, D., Coppalle, A., Branger, H., Connan, O., Laguionie, P., Hébert, D. and Talbaut, M. (2013). Measurement in a wind tunnel of dry deposition velocities of submicron aerosol with associated turbulence onto rough and smooth urban surfaces. *Journal of Aerosol Science*. 55: 12–24.

Sæbø, a, Popek, R., Nawrot, B., Hanslin, H.M., Gawronska, H. and Gawronski, S.W. (2012). Plant species differences in particulate matter accumulation on leaf surfaces. *The Science of the total environment.* 427–428: 347–54.

Sabatino, S. Di, Buccolieri, R., Pappaccogli, G. and Leo, L.S. (2015). The effects of trees on micrometeorology in a real street canyon: consequences for local air quality. *International Journal of Environmental Pollution.* 58:100.

Sager, T.M. and Castranova, V. (2009). Surface area of particle administered versus mass in determining the pulmonary toxicity of ultrafine and fine carbon black: comparison to ultrafine titanium dioxide. *Particle and Fibre Toxicology. 6:15.*

Salmond, J.A., Tadaki, M., Vardoulakis, S., Arbuthnott, K., Coutts, A., Demuzere, M., Dirks, K.N., Heaviside, C., Lim, S., Macintyre, H., McInnes, R.N. and Wheeler, B.W. (2016). Health and climate related ecosystem services provided by street trees in the urban environment. *Environmental Health*. 15: 36

Sadeghian, M.M. (2016). A Review on Green Wall , Classification and Function. *International Journal of Scientific Research in Science and Technology*. 2:47–51.

Schaubroeck, T., Deckmyn, G., Neirynck, J., Staelens, J., Adriaenssens, S., Dewulf, J., Muys, B. and Verheyen, K. (2014). Multilayered modeling of particulate matter removal by a growing forest over time, from plant surface deposition to washoff via rainfall. *Environmental Science and Technology*. 48: 10785–10794.

Schneider, I.L., Teixeira, E.C., Silva Oliveira, L.F. and Wiegand, F. (2015). Atmospheric particle number concentration and size distribution in a traffic–impacted area. *Atmospheric Pollution Research*.6: 877–885.

Schwartz, J., Dockery, D.W., Neas, L.M. (1996). Is daily mortality associated with fine particles? *Journal of the Air & Waste Management Association*. 46:927–939

Seaton, A., Godden, D., MacNee, W. and Donaldson, K. (1995). Particulate air pollution and acute health effects. *The Lancet.* 345: 176–178.

Sehmel, G.A. (1980). Particle and gas deposition - a review. *Atmospheric Environment*. 14: 983–1011.

Seinfeld, J. H. and Pandis, S. N. (2006). *Atmospheric Chemistry And Physics: From Air Pollution To Climate Change*. Wiley-VCH: New York.

Sell, P. and Murrell, G. (2014). *Flora of Great Britain and Ireland* . Cambrdridge University press. United Kingdom.

Sgrigna, G., Sæbø, A., Gawronski, S., Popek, R. and Calfapietra, C. (2015). Particulate Matter deposition on Quercus ilex leaves in an industrial city of central Italy. *Environmental Pollution*. 197: 187-194.

Sharma, M., Agarwal, A.K. and Bharathi, K.V.L. (2005). Characterization of exhaust particulates from diesel engine. *Atmospheric Environment*. 39: 3023–3028.

Shaughnessy, W.J., Venigalla, M.M. and Trump, D. (2015). Health effects of ambient levels of respirable particulate matter (PM) on healthy, young-adult population. *Atmospheric Environment*. 123: 102–111.

Shackleton, K., Bell, N., Smith, H. and Davies, L. (not dated). *The role of shrubs and perennials in the capture and mitigation of particulate air pollution in London.* Centre for Environmental Policy. Imperial College, London.

Shah, S., Cocker, D., Miller, J.W. and Norbeck, J.M. (2004). Emission rates of particulate matter and elemental and organic carbon from in use diesel engines. *Environmental Science and Technology*. 38:2544-2550

Sheweka, S. and Magdy, N. (2011). The living walls as an approach for a healthy urban environment. *Energy Procedia.* 6:592–599.

Silverman, D.T., Samanic, C.M., Lubin, J.H., Blair, A.E., Stewart, P.A., Vermeulen, R., Coble, J.B., Rothman, N., Schleiff, P.L., Travis, W.D., Ziegler, R.G., Wacholder, S. and Attfield, M.D. (2012). The diesel exhaust in miners study: A nested case-control study of lung cancer and diesel exhaust. *Journal of the National Cancer Institute*. 104: 855–868.

Šíp, V. and Beneš, L. (2016). Modelling the Effects of a Vegetation Barrier on Road Dust Dispersion. *Applied Mechanics and Materials*. 821: 105–112.

Slezakova, K., Reis, M. a., Pereira, M.C. and Alvim-Ferraz, M.C. (2007). Influence of traffic on the elemental composition of PM10 and PM2.5 in Oporto region. *Air Pollution*. 1: 59–68.

Slinn, W.G.N. (1982). Predictions for particle deposition to vegetative canopies. *Atmospheric Environment.* 16:1785–1794.

Smith, W.H. (1977) Removal of atmospheric particulates by urban vegetation: implications for human and vegetative health. *The Yale Journal of Botany and Medicine,* 50:185-197.

Smith, K.E.C. and Jones, K.C. (2000). Particles and vegetation: implications for the transfer of particle-bound organic contaminants to vegetation. *The Science of the Total Environment*. 246: 207–236.

Solomon, P.A., Costantini, M., Grahame, T.J. *et al.* (2012). Air pollution and health: bridging the gap from sources to health outcomes: conference summary. *Air Quality Atmosphere and Health*. 5:9–*62.*

Song, Y., Maher, B.A., Li, F., Wang, X., Sun, X. and Zhang, H. (2015). Particulate matter deposited on leaf of five evergreen species in Beijing, China: Source identification and size distribution. *Atmospheric Environment*. 105: 53–60.

Speak, A.F., Rothwell, J.J., Lindley, S.J. and Smith, C.L. (2012). Urban particulate pollution reduction by four species of green roof vegetation in a UK city. *Atmospheric Environment*. 61: 283–293.

Spencer, M.T., Holecek, J.C., Corrigan, C.E., Ramanathan, V. and Prather, K.A. (2008). Size-resolved chemical composition of aerosol particles during a monsoonal transition period over the Indian Ocean. *Journal of Geophysical Research: Atmospheres*. 113.

Stanek, L.W., Sacks, J.D., Dutton, S.J. and Dubois, J.J.B. (2011). Attributing health effects to apportioned components and sources of particulate matter: an evaluation of collective results. *Atmospheric Environment*. 45: 5655-5663.

Stern, A. c, Wohlers, H.C., Boubel, R.W. and Lowry, W.P. (2014). *Fundamentals of Air Pollution*. New York, London.

Sternberg, T., Viles, H., Cathersides, A. and Edwards, M. (2010). Dust particulate absorption by ivy (*Hedera helix* L) on historic walls in urban environments. *Science of the Total Environment*. 409:162–168.

Sternberg, T., Viles, H. and Cathersides, A. (2011). Evaluating the role of ivy (*Hedera helix*) in moderating wall surface microclimates and contributing to the bioprotection of historic buildings. *Building and Environment*. 46: 293-297.

Swietlicki, E., Puri, S., Hansson, H.C. and Edner, H. (1996) Urban air pollution source apportionment using a combination of aerosol and gas monitoring techniques. *Atmospheric Environment*. 30: 2795 – 2809.

Taha H (1997) Urban climates and heat islands: albedo, evapotraspiration, and antropogenic heat. *Energy and Buildings*. 25:99–103

Tallis, M., Taylor, G., Sinnett, D. and Freer-Smith, P. (2011). Estimating the removal of atmospheric particulate pollution by the urban tree canopy of London, under current and future environments. *Landscape and Urban Planning.* 103:129–138.

Terzaghi, E., Wild, E., Zacchello, G., Cerabolini, B.E.L., Jones, K.C. and Di Guardo, A. (2013). Forest Filter Effect: Role of leaves in capturing/releasing air particulate matter and its associated PAHs. *Atmospheric Environment*. 74: 378–384.

Thompson, J. R, Mueller, P. W., FIUckiger, W. and Rutter, A. J. (1984) The effect of dust on photosynthesis and its significance for roadside plants*. Environmental Pollution.* 34: 171-190.

Thornes, J.E., Cai, X., Hickman, A., Maria, J., Saborit, D. and Baker, C. (2016). Air quality in enclosed railway stations, *Institution of Civil Engineers*. 1–9.

Thornes, J.E. (2016). *'Breathe easy – engineering air quality solutions'.* Proceedings-ARCC network.

Thorpe, A. and Harrison, R.M. (2008). Sources and properties of non-exhaust particulate matter from road traffic: A review. *Science of the Total Environment*. 400: 270–282.

Thottathil, V.J., Balamuralikrishna, C. and Ghosh, S. (2011). Role of Green Facades in Creating Sustainable Environments : Comparing Chemical Analyses and Sem Images to Quantify the Atmospheric Cleansing by Green Creepers. World Academy of Science, *Engineering and Technology.* 53:343–346.

Timmers, V.R.J.H. and Achten, P.A.J. (2016). Non-exhaust PM emissions from electric vehicles. *Atmospheric Environment.*134: 10–17.

Tiwary, A., Reff, A. and Colls, J.J. (2008). Collection of ambient particulate matter by porous vegetation barriers: Sampling and characterization methods*. Journal of Aerosol Science*.39: 40–47.

Tomasevic, M. Vukmirovic, Z. Rajsic, S. Tasic, M. and Stevanovic, B. (2005). Characterization of trace metal particles deposited on some deciduous tree leaves in an urban area. *Chemosphere*. 61:753–60.

Tomaszewski, D., Zieliński, J. and Gawlak, M. (2014). Foliar indumentum in central-European *Rubus* species (*Rosaceae*) and its contribution to the systematics of the group. *Nordic Journal of Botany*. 32: 1–10.

Tong, S.T.Y. (1991). The retention of copper and lead particulate matter in plant foliage and forest soil. *Environment International.* 17: 31-37.

Tong, Z., Whitlow, T.H., Macrae, P.F., Landers, A.J. and Harada, Y. (2015). Quantifying the effect of vegetation on near-road air quality using brief campaigns. *Environmental Pollution*. 201:14-149.

Tong, Z., Baldauf, R.W., Isakov, V., Deshmukh, P. and Max Zhang, K. (2016). Roadside vegetation barrier designs to mitigate near-road air pollution impacts. *Science of the Total Environment*. 541:920–927.

UKpopulation (2017) *Population of Stoke-on-Trent*. [Online] http://ukpopulation2017.com/population-of-stoke-on-trent-2017.html. [Accessed 22nd June 2017]

Ulrich, R.S and Simons, R. (1986). 'Recovery from stress during exposure to every day outdoor environments'. In: Wineman J, Barns R, Zimring C (eds) The costs of not knowing. Proceedings of the seventieth annual conference of the environmental research association, Washington DC.

Van Bohemen, H.D., Fraaij, A.L.A. and Ottele, M. (2008). 'Ecological engineering, green roofs and the greening of vertical walls of buildings in urban areas'. Proceedings of Ecocity World Summit. p. 1–10.

Varshney, C.K. and Mitra, I. (1993). Importance of hedges in improving urban air-quality. *Landscape and Urban Planning.* 25: 75–83.

Velali, E., Papachristou, E., Pantazaki, A., Choli-Papadopoulou, T., Planou, S., Kouras, A., Manoli, E., Besis, A. and Voutsa, D. (2016). Redox activity and in vitro bioactivity of the water-soluble fraction of urban particulate matter in relation to particle size and chemical composition*. Environmental Pollution.* 208: 774-786.

Viles, H., Sternberg, T. and Cathersides, A. (2011) Is Ivy Good or Bad for Historic Walls? *Journal of Architectural Conservation*, 17: 25-41.

Verma, A. and Singh, S.N. (2006). Biochemical and ultrastructural changes in plant foliage exposed to auto pollution. *Environmental Monitoring and Assessment*. 120:585–602.

Vose, J.M., Dougherty, P.M., Long, J.N., Smith, F.W., Gholz, H.L. and Curran, P.J. (1994). Factors influencing the amount and distribution of leaf area of pine stands. *Ecological Bulletins*. 43:102–114.

Wåhlin, P., Berkowicz, R. and Palmgren, F. (2006). Characterisation of traffic-generated particulate matter in Copenhagen*. Atmospheric Environment*. 40: 2151–2159.

Walters, S., Griffiths, R.K. and Ayres, J.G. (1994). Temporal association between hospital admissions for asthma in Birmingham and ambient levels of sulphur dioxide and smoke. *Thorax* 49:133-40.

Wang, L., Liu, L., Gao, S., Hasi, E. and Wang, Z. (2006). Physicochemical characteristics of ambient particles settling upon leaf surfaces of urban plants in Beijing. *Journal of Environmental Science.* 18:921-926

Wang, Y.F., Huang, K.L., Li, C.T., Mi, H.H., Luo, J.H. and Tsai, P.J. (2003). Emissions of fuel metals content from a diesel vehicle engine. *Atmospheric Environment*. 37:4637–4643.

Wang, H., Shi, H. and Li, Y. (2011). Leaf dust capturing capacity of urban greening plant species in relation to leaf micromorphology. *International Symposium on Water Resource and Environmental Protection* (ISWREP). 2198–2201.

Weber, F., Kowarik, I. and Säumel, I. (2014). Herbaceous plants as filters: immobilization of particulates along urban street corridors. *Environmental pollution.* 186: 234–40.

Weckwerth, G. (2001). Verification of traffic emitted aerosol components in the ambient air of cologne (Germany). *Atmospheric Environment* 35: 5525–5536.

Weerakkody, U., Dover, J.W.,Mitchell, P. and Reiling, K. (2017) Particulate Matter pollution capture by leaves of seventeen living wall species with special reference to rail-traffic at a metropolitan station. *Urban Forestry and Urban Greening*. 27: 173-186.

Weerakkody, U., Dover, J.W.,Mitchell, P. and Reiling, K. (2018a). Evaluating the impact of individual leaf traits on atmospheric particulate matter accumulation using natural and synthetic leaves. *Urban Forestry and Urban Greening.* 30: 98-107.

Weerakkody, U., Dover, J.W.,Mitchell, P. and Reiling, K. (2018b). Quantification of the traffic-generated particulate matter capture by plant species in a living wall and evaluation of the important leaf characteristics. *Science of The Total Environment.* 635:1012-1024.

Weerakkody, U., Dover, J.W., Mitchell, P., Reiling, K., (2018c). The impact of rainfall in remobilising particulate matter accumulated on leaves of evergreen species grown on a green screen and a living wall. Submitted to *Urban Forestry and Urban Greening* (in press).

Wesely, M.L., Cook, D.R., Hart, R.L. and Speer, R.E. (1985) Measurements and parameterisation of particulate sulfur dry deposition over grass. *Journal of Geophysical Research.* 90: 2131–2143.

WHO. (2005). *Health effects of transport-related air pollution*. Regional Office for Europe. Copenhagen.

WHO. (2014). *WHO' s Ambient Air Pollution database* - Update 2014 Data summary of the AAP database. [Online] http://www.who.int/phe/health_topics/outdoorair/databases/cities/en/. [Accessed 5th January 2015]

WHO (2016). *WHO' s Ambient Air Pollution database* - Update 2016 Data summary of the AAP database. [Online]. http://www.who.int/phe/health_topics/outdoorair/databases/cities/en/. [Accessed 9th December 2017]

Williamson, B., Mikhailova, I., Purvis, O. and Udachin, V. (2004). SEM-EDX analysis in the source apportionment of particulate matter on Hypogymnia physodes lichen transplants around the Cu smelter and former mining town of Karabash, South Urals, Russia. *Science of The Total Environment*. 322:139–154.

Willmer, C.M. (1983). Stomata. New York: Longman Inc.

Wilson, E., Underwood, M., Puckrin, O., Letto, K., Doyle, R., Caravan, H., Camus, S. and Bassett, K. (2010). *The Arcsine Transformation: Has the time come for retirement?* A technical report. The University of New South Wales. Australia.

Winkler, P. (1988). The growth of atmospheric aerosol particles with relative humidity. *Physica Scripta*. 37: 223–230.

Wiman, B.L.B. and Agren, G.I. Lannefors, H.O. (1985) Aerosol concentration profiles within a mature coniferous forest-model versus field results. *Atmospheric Environment* 19:363–367.

Witherspoon, J.P. and Taylor, F.G.J. (1969). Retention of a fallout simulant containing 134Cs by pine and oak trees. *Health Physics*. 17: 825–829.

Wolverton, B.C. and Wolverton, J.D. (1993). Plants and soil microorganisms: removal of formaldehyde, xylene, and ammonia from the indoor environment. *Journal of the Mississippi Academy of Sciences.* 38: 11-15.

Wong, N.H., Tan, A.Y.K., Chen, Y., Sekar, K., Tan, P.Y., Chan, D., Chiang, K. and Wong, N.C. (2010a). Thermal evaluation of vertical greenery systems for building walls. *Building and Environment.* 45:663–672.

Wong, N.H., Tan, A.Y.K., Tan, P.Y., Sia, A. and Wong, N.C. (2010b). Perception Studies of Vertical Greenery Systems in Singapore. *Journal of Urban Planning and Development*. 136:330–338.

Woodruff TJ, Grillo J and Schoendorf KC. (1997). The relationship between selected causes of postneonatal infant mortality and particulate air pollution in the United States. *Environ Health Perspect*. 105:608–612.

Woodruff, T.J., Parker, J.D. and Schoendorf, K.C. (2006). Fine Particulate Matter ($PM_{2.5}$) Air Pollution and Selected Causes of Post-neonatal Infant Mortality in California. *Environmental Health Perspectives*. 114:786–790.

Xia, L., Lenaghan, S.C., Zhang, M., Wu, Y., Ahao, X., Burris, J.N. and Stewart, C.N.J. (2011). Characterization of English ivy (*Hedera helix*) adhesion force and imaging using atomic force microscopy. *Journal of Nanoparticle Research.* 13: 1029–1037.

Xu, X., Zhang, Z., Bao, L., Mo, L., Yu, X., Fan, D. and Lun, X. (2017). Influence of rainfall duration and intensity on particulate matter removal from plant leaves. *Science of the Total Environment.* 609: 11–16.

Yang, J., Yu, Q. and Gong, P. (2008). Quantifying air pollution removal by green roofs in Chicago. *Atmospheric Environment*: 42: 7266–7273.

Yang, J., Chang, Y. and Yan, P. (2015). Ranking the suitability of common urban tree species for controlling PM 2.5 pollution. *Atmospheric Pollution Research*. 6: 267-277.

Yan, J., Lin, L., Zhou, W., Ma, K. and Pickett, S.T.A. (2016a). A novel approach for quantifying particulate matter distribution on leaf surface by combining SEM and object-based image analysis. *Remote Sensing of Environment*. 173: 156–161.

Yan, J., Lin, L., Zhou, W., Han, L. and Ma, K. (2016b). Quantifying the characteristics of particulate matters captured by urban plants using an automatic approach. *Journal of Environmental Sciences (China).*39: 259–267.

Yin, J.X. and Harrison, R.M. (2008). Pragmatic mass closure study for $PM_{1.0}$, $PM_{2.5}$ and PM_{10} at roadside, urban background and rural sites. *Atmospheric Environment*. 42: 980-988.

Zhang, W.K., Wang, B. and Niu, X. (2015). Study on the adsorption capacities for airborne particulates of landscape plants in different polluted regions in Beijing (China). *International Journal of Environmental Research and Public Health.* 12: 9623–9638.

Zhang, W., Wang, B. and Niu, X. (2017). Relationship between Leaf Surface Characteristics and Particle Capturing Capacities of Different Tree Species in Beijing. *Forests*. 8:92.

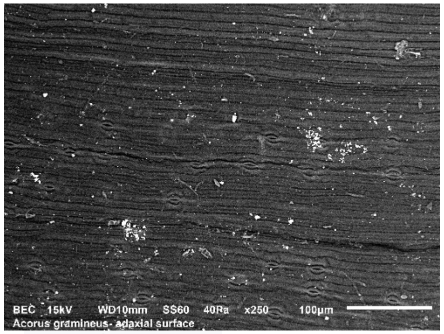
Zhang, Z.H., Khlystov, A., Norford, L.K., Tan, Z.K. and Balasubramanian, R. (2017). Characterization of traffic-related ambient fine particulate matter ($PM_{2.5}$) in an Asian city: Environmental and health implications. *Atmospheric Environment.* 161: 132–143.

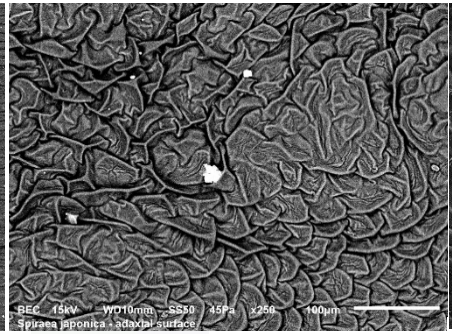
Zheng, G. and Moskal, L.M. (2009). Retrieving Leaf Area Index (LAI) using remote sensing: theories, methods and sensors. *Sensors.* 9: 2719–45.

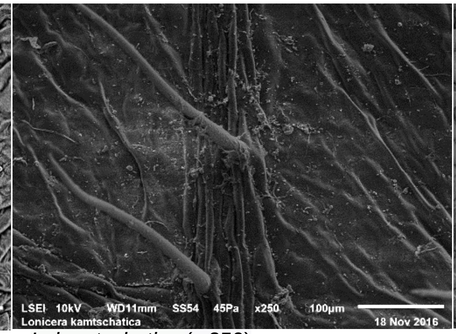
Zulfacar, A. (1975). Vegetation and urban environment. J*ournal of Urban Planning and Development*. 101: 21-33.

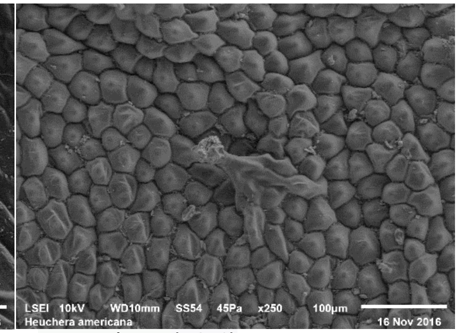
Appendix 1**: Sample Scanning Electron Microscope images of leaf micromorphology (micromorphology of the leaves which were not included in the main text are given here)**

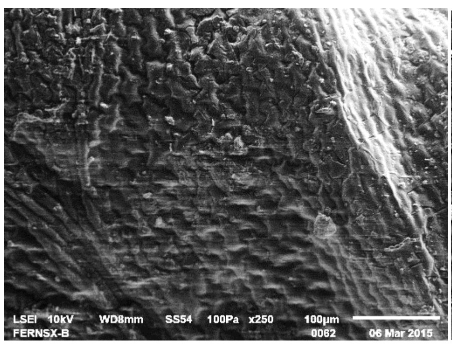
Micrographs taken visualising the adaxial surfaces of the leaves

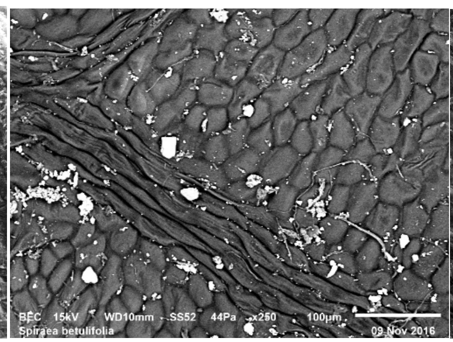

A. gramineus (x 250)

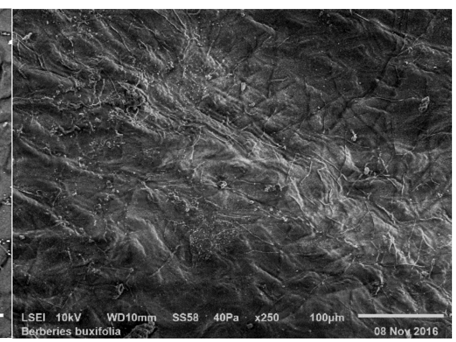

S. japonica (x 250)

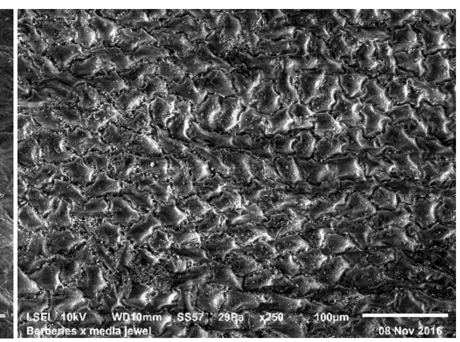

L. kamtschatica (x 250)


H. americana (x 250)

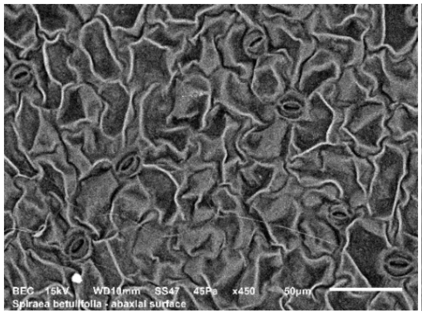

B. spicant (x 250)


S. betulifolia (x 250)

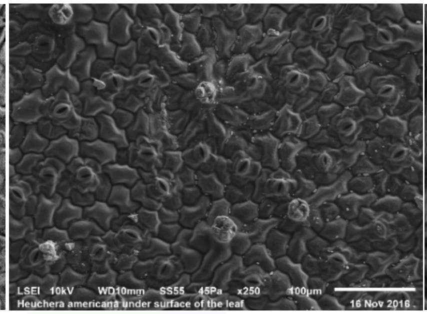

B. buxifolia (x 250)


B. ×media (x 250)

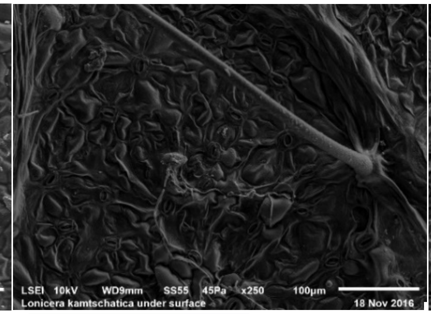
Micrographs taken visualising the abaxial surfaces of the leaves



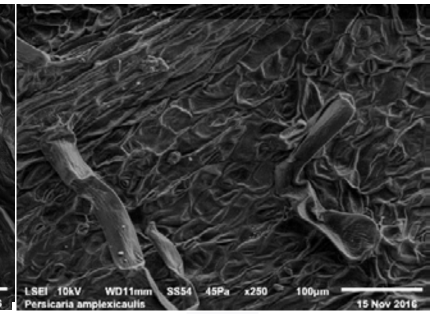
S. betulifolia (x 450)



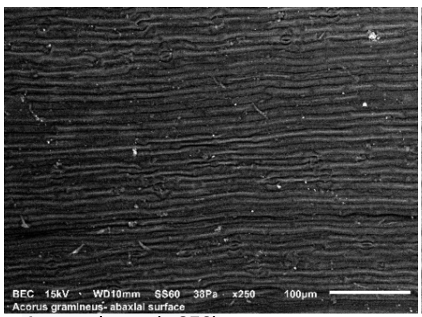
H. americana (x 250)



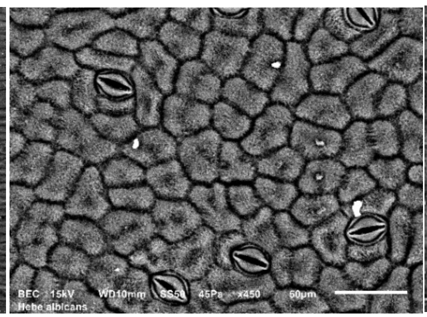
L. kamtschatica (x 250)



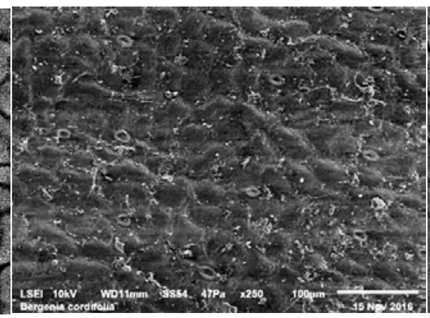
P. amplexicaulis (x 250)



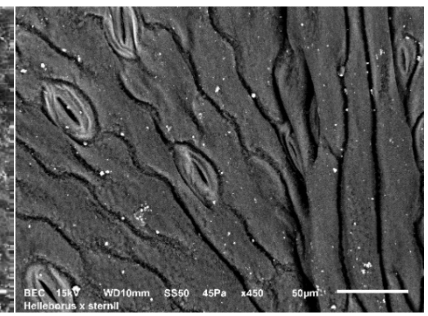
A. gramineus (x 250)



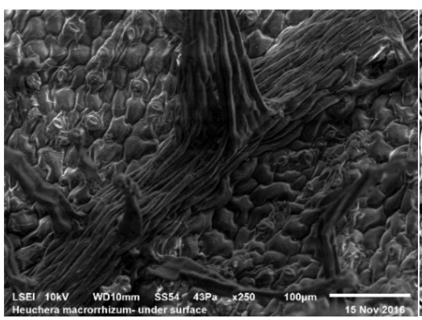
H.albicans (x450)



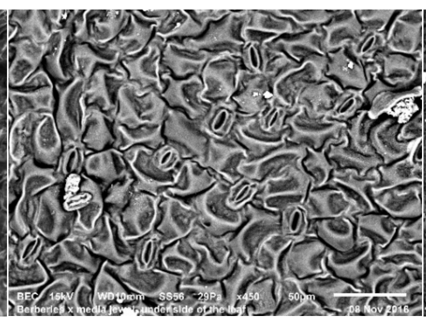
B. cordifolia (x 250)



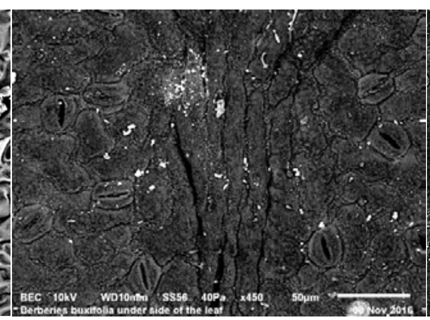
H. × sternii (x 450)



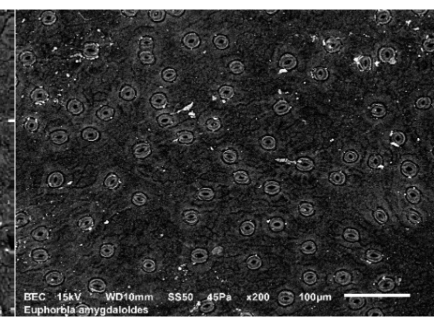
H. villosa (x 250)



B. ×media (x 450)



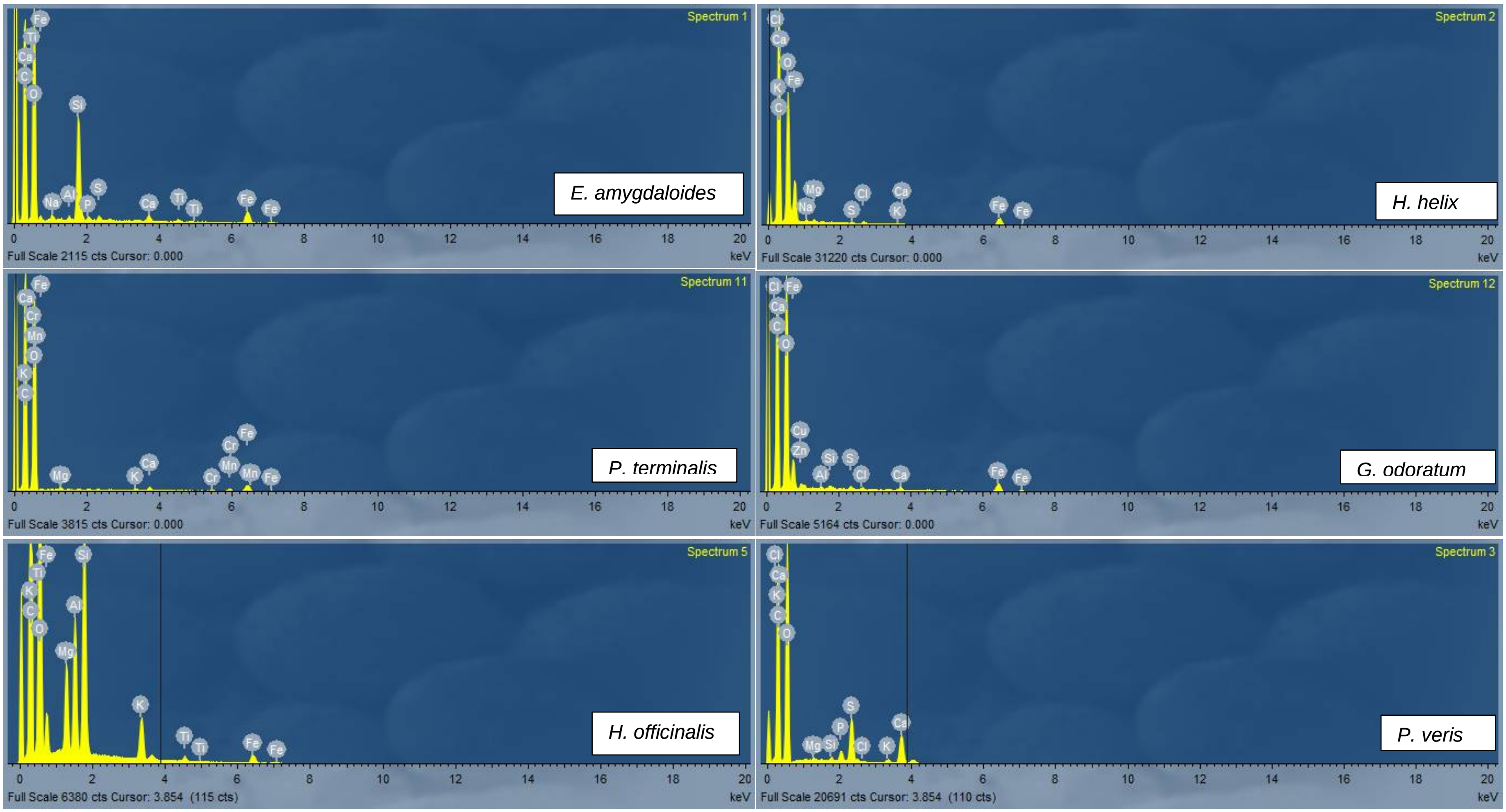
B. buxifolia (x 450)

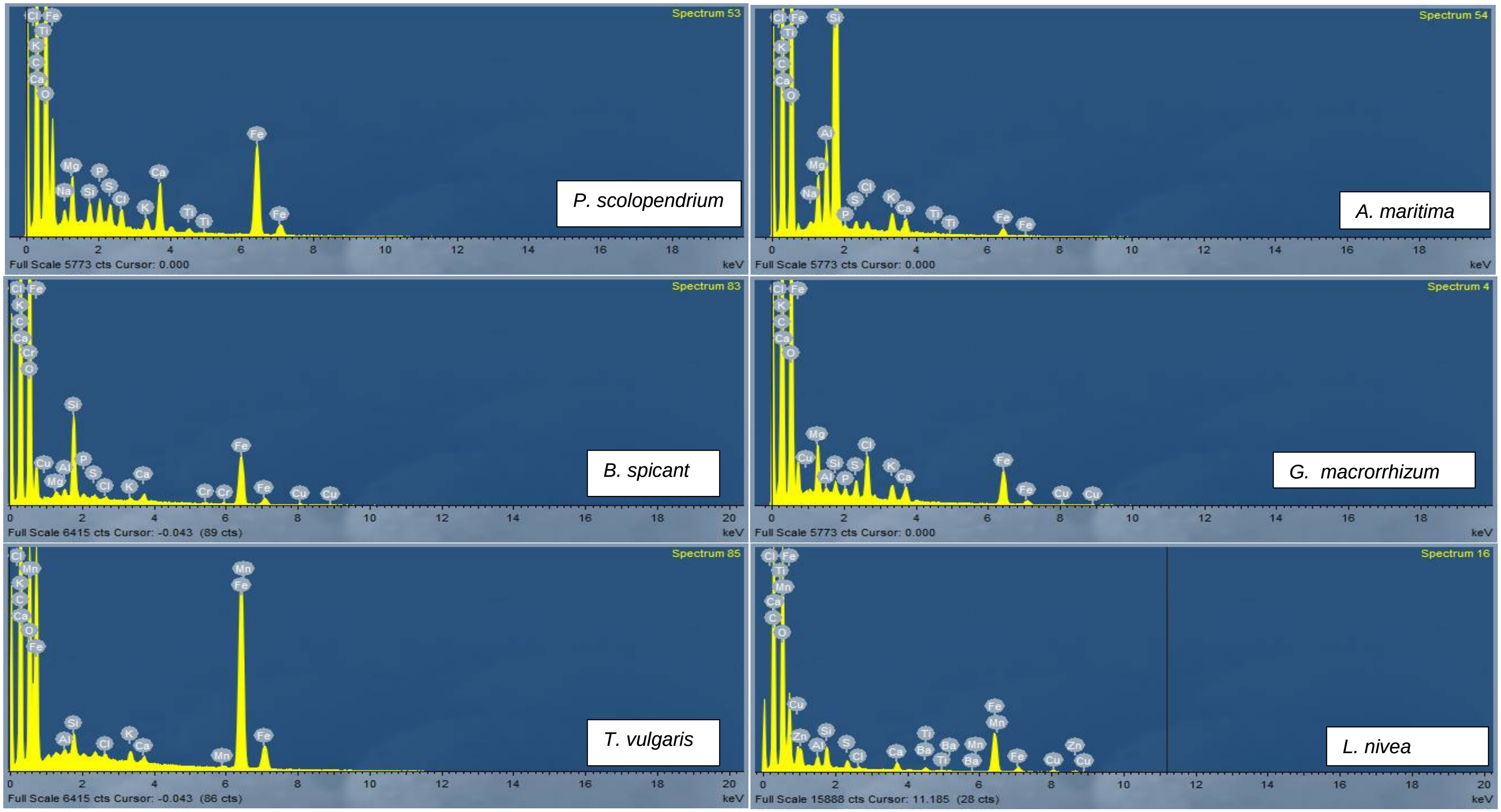


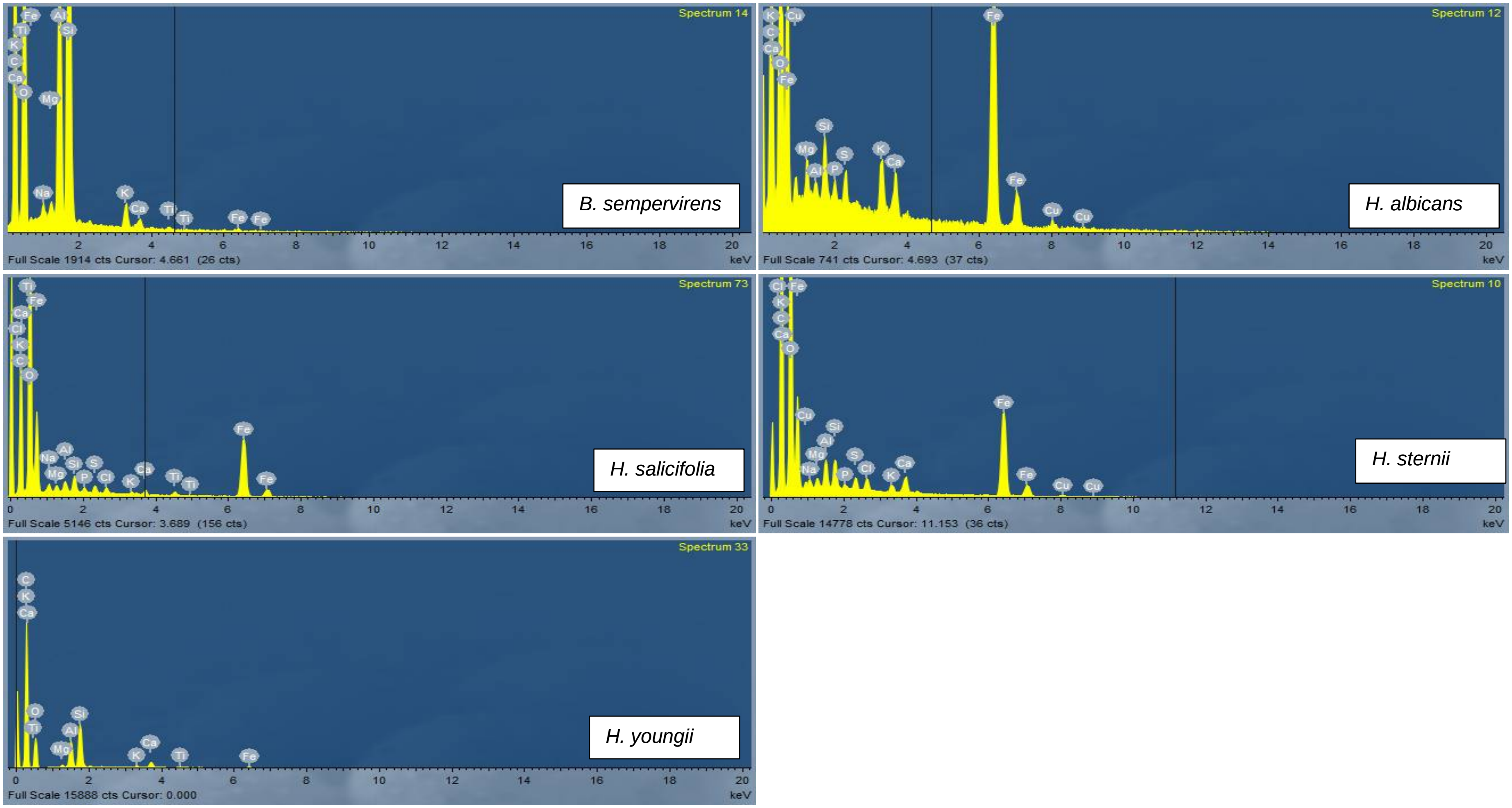
E. amygdaloide (x 200)

Appendix 2: Sample EDX spectra of elemental compositions of PM captured on leaves of species grown on living wall studied in this thesis.

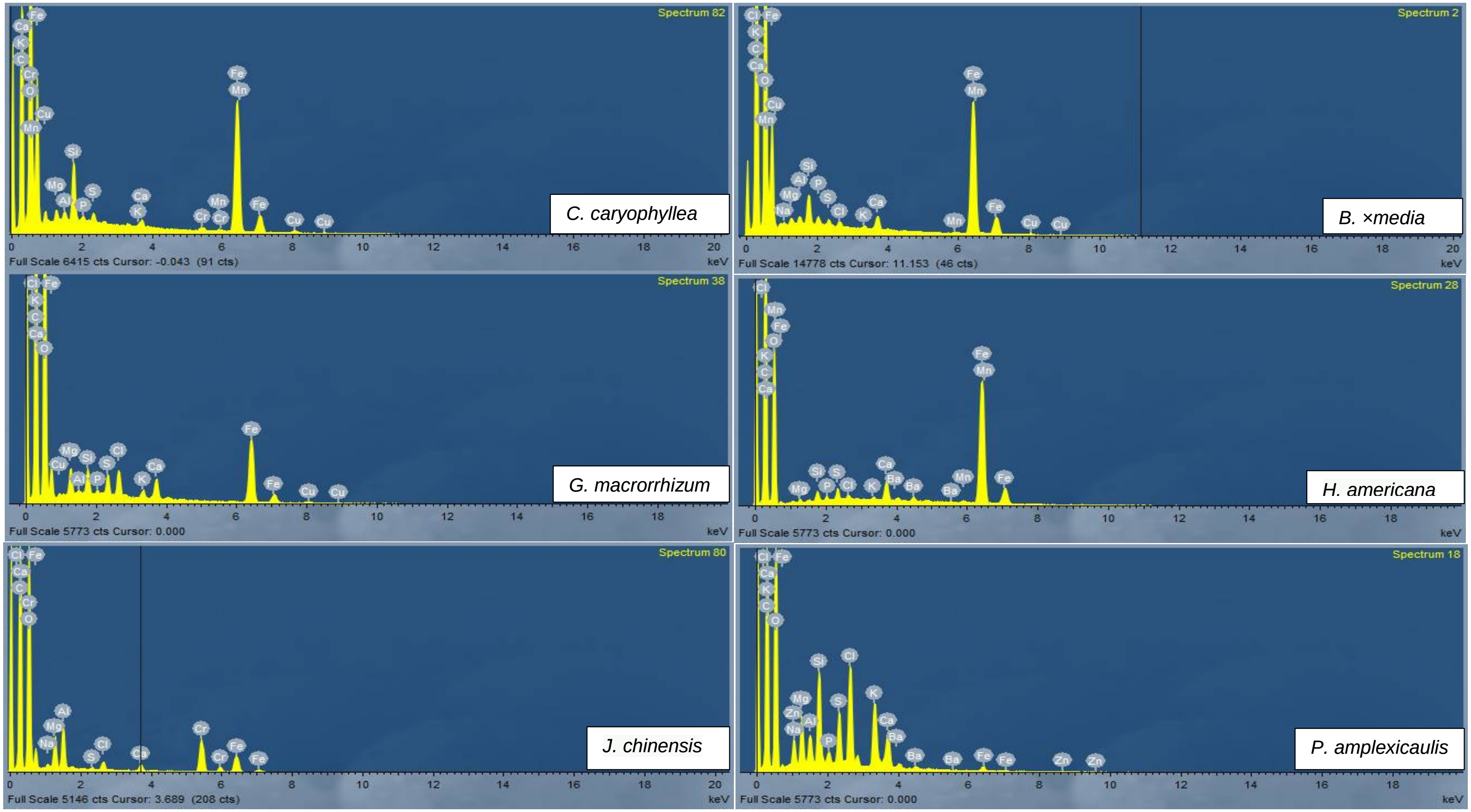
Species grown on the living wall located adjacent to New Street station, Birmingham, UK

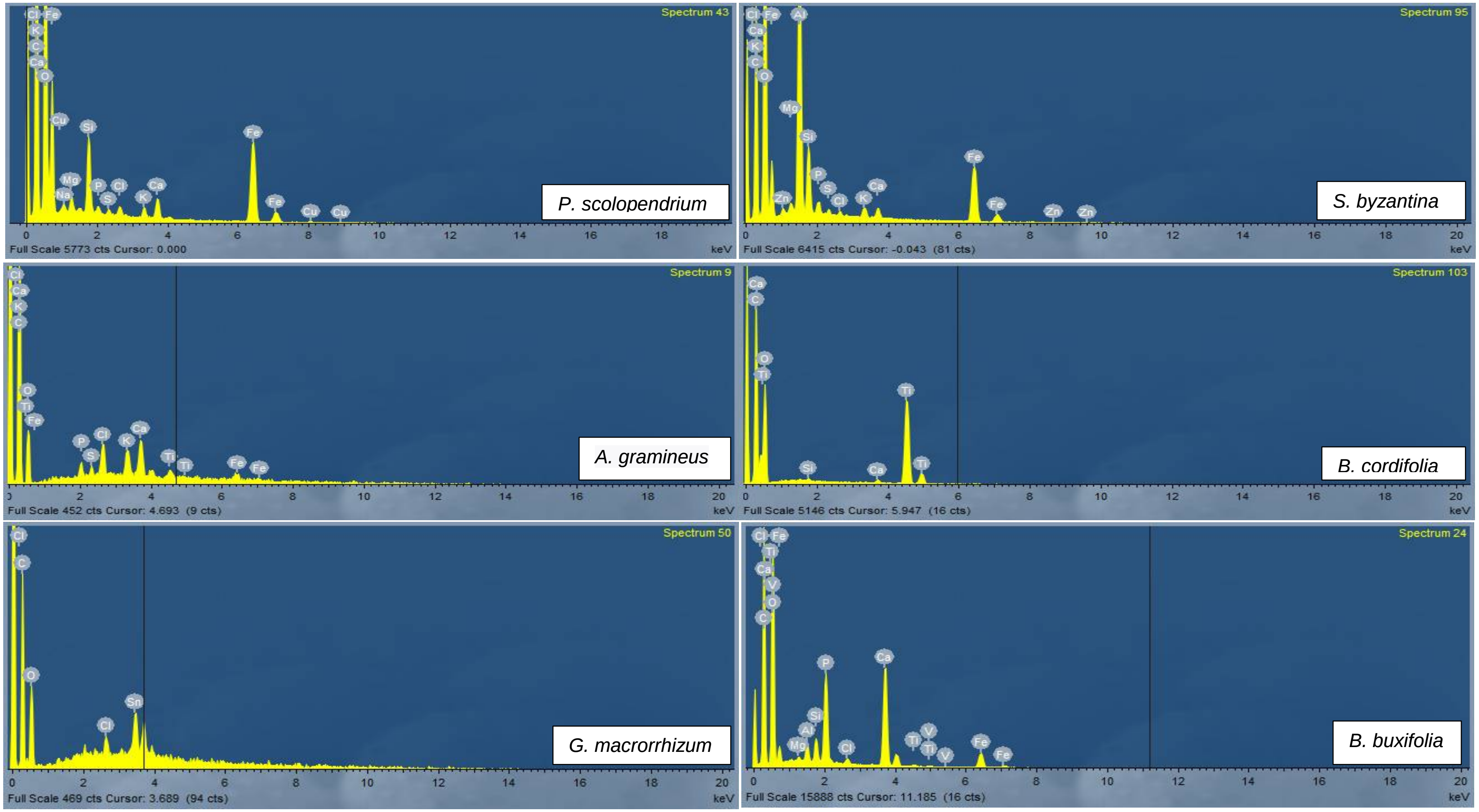


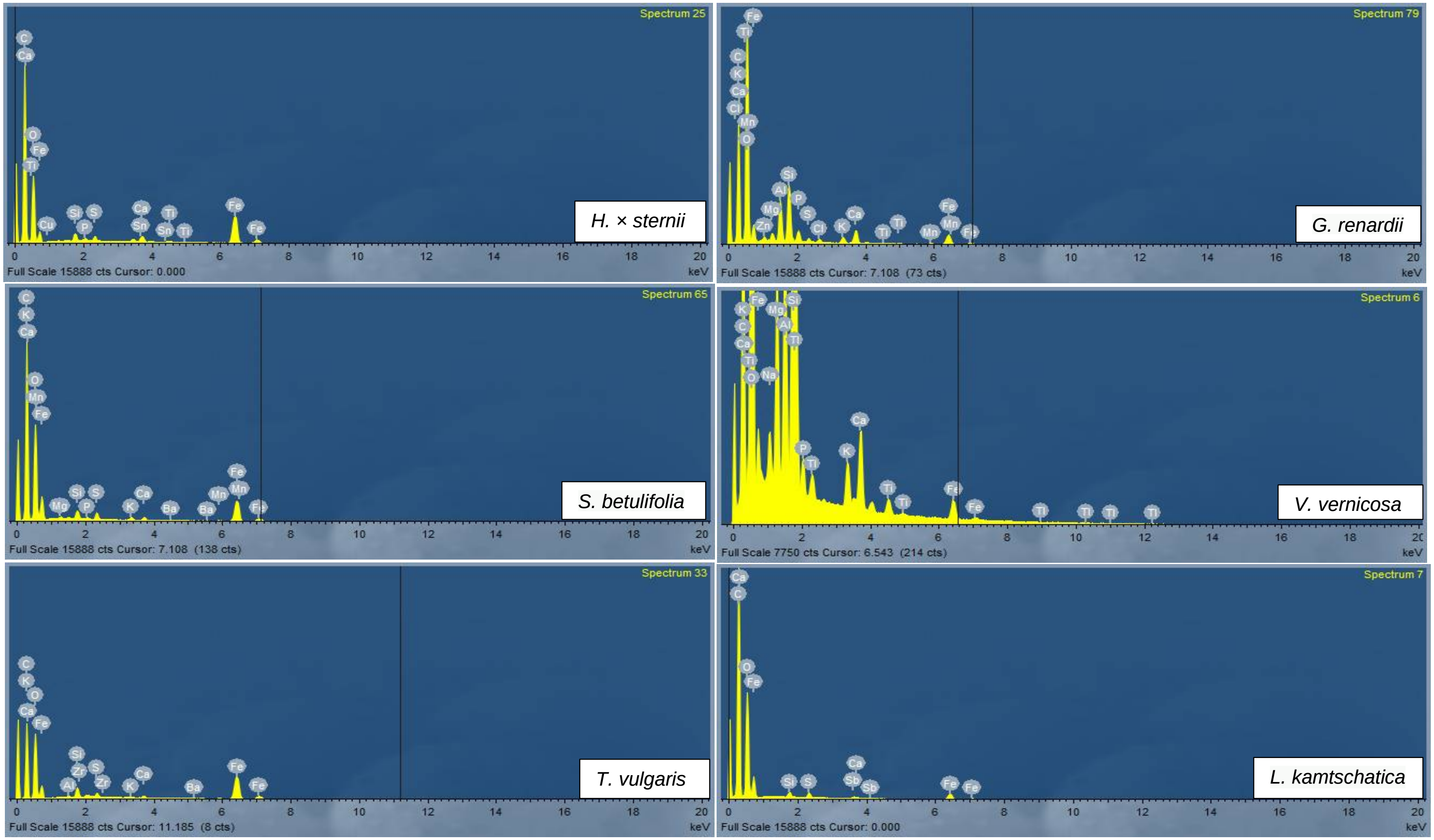

Spectrum 53
P. scolopendrium
Full Scale 5773 cts Cursor: 0.000
keV
Spectrum 54
A. maritima
Full Scale 5773 cts Cursor: 0.000
keV
Spectrum 83
B. spicant
Full Scale 6415 cts Cursor: -0.043 (89 cts)
keV
Spectrum 4
G. macrorrhizum
Full Scale 5773 cts Cursor: 0.000
keV
Spectrum 85
T. vulgaris
Full Scale 6415 cts Cursor: -0.043 (86 cts)
keV
Spectrum 16
L. nivea
Full Scale 15888 cts Cursor: 11.185 (28 cts)
keV

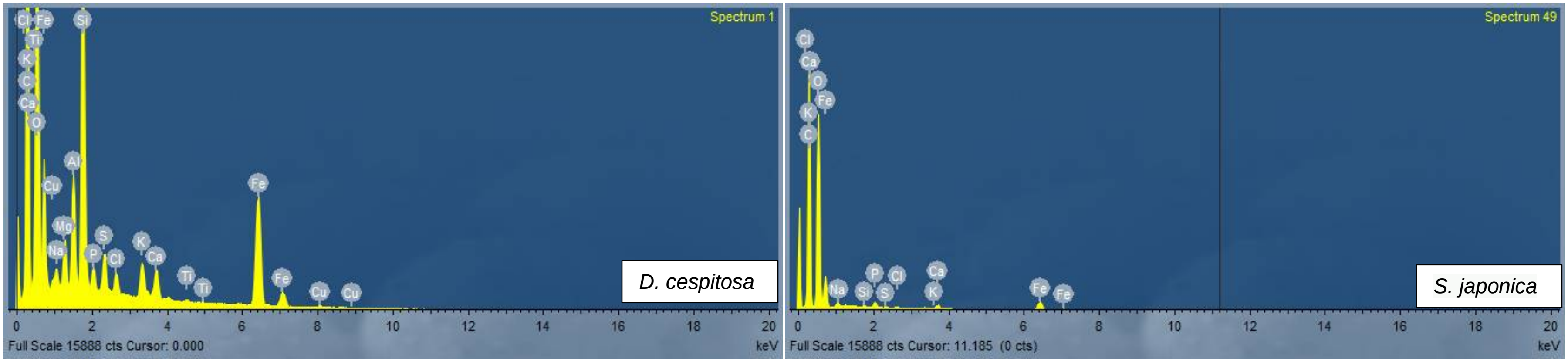

Spectrum 14
B. sempervirens
Full Scale 1914 cts Cursor: 4.661 (26 cts)
keV
Spectrum 12
H. albicans
Full Scale 741 cts Cursor: 4.693 (37 cts)
keV
Spectrum 73
H. salicifolia
Full Scale 5146 cts Cursor: 3.689 (156 cts)
keV
Spectrum 10
H. sternii
Full Scale 14778 cts Cursor: 11.153 (36 cts)
keV
Spectrum 33
H. youngii
Full Scale 15888 cts Cursor: 0.000
keV

Species used in the experimentally manipulable living wall located along Leek Road, Stoke-on-Trent, UK




Spectrum 43
P. scolopendrium
Full Scale 5773 cts Cursor: 0.000
keV
Spectrum 95
S. byzantina
Full Scale 6415 cts Cursor: -0.043 (81 cts)
keV
Spectrum 9
A. gramineus
Full Scale 452 cts Cursor: 4.693 (9 cts)
keV
Spectrum 103
B. cordifolia
Full Scale 5146 cts Cursor: 5.947 (16 cts)
keV
Spectrum 50
G. macrorrhizum
Full Scale 469 cts Cursor: 3.689 (94 cts)
keV
Spectrum 24
B. buxifolia
Full Scale 15888 cts Cursor: 11.185 (16 cts)
keV


Spectrum 25
H. × sternii
Full Scale 15888 cts Cursor: 0.000
Spectrum 79
G. renardii
Full Scale 15888 cts Cursor: 7.108 (73 cts)
Spectrum 65
S. betulifolia
Full Scale 15888 cts Cursor: 7.108 (138 cts)
Spectrum 6
V. vernicosa
Full Scale 7750 cts Cursor: 6.543 (214 cts)
Spectrum 33
T. vulgaris
Full Scale 15888 cts Cursor: 11.185 (8 cts)
Spectrum 7
L. kamtschatica
Full Scale 15888 cts Cursor: 0.000


Spectrum 1
D. cespitosa
Full Scale 15888 cts Cursor: 0.000
keV
Spectrum 49
S. japonica
Full Scale 15888 cts Cursor: 11.185 (0 cts)
keV

Appendix 3: Results of GLMM in the analysis of relationship between leaf micromorphology and PM density on the leaves of different species.

Results of GLMM in the analysis of relationship between leaf micromorphology and PM densitie s on the leaves of plant species on the experimental living wall located along Leek Road, Stoke-on-Trent (n=10). (Chapter 5: Section 5.1.4.2)

Character	Density of PM on the adaxial surfaces of the leaves			Density of PM on the abaxial surfaces of the leaves		
PM size fraction	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
Hair/trichomes	t= 0.49 p=0.62	t= 0.37 p=0.71	t= 3.27 p=0.001	t= 0.06 p=0.95	t= 0.76 p=0.45	t= 0.42 p=0.66
Grooves	t= 1.73 p=0.08	t= 1.39 p=0.16	t= 0.97 p=0.33	t= 0.36 p=0.71	t= 1.34 p=0.18	t= 0.96 p=0.34
Ridges	t= 1.20 p=0.23	t= 0.62 p=0.53	t= 0.25 p= 0.80	t= 0.29 p= 0.76	t= 2.99 p= 0.003	t= 0.04 p= 0.97
Stomata	t= 1.49 p=0.13	t= 2.13 p=0.03	t= 5.11 p=0.0000	t= 0.48 p= 0.63	t= 0.91 p=0.36	t= 2.39 p=0.017

Results of GLMM in the analysis of relationship between leaf micromorphology and PM densities on the leaves of *Rubus* species used in an existing living wall located on Staffordshire University campus, Stoke-on-Trent (n=10). (Chapter 5: Section 5.3.2.2)

Character	Density of PM on the adaxial surfaces of the leaves			Density of PM on the abaxial surfaces of the leaves		
PM size fraction	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
Hair/trichomes	t= 1.17 p=0.24	t=0.88 p=0.38	t= 0.91 p=0.36	t= 4.35 p=0.0001	t= 1.21 p=0.23	t= 0.24 p=0.22
Grooves	t= 3.65 p=0.0005	t= 5.52 p=0.0000	t= 0.55 p=0.12	NA	NA	NA
Ridges	t= 1.98 p=0.052	t= 3.77 p=0.0004	t= 0.88 p= 0.38	NA	NA	NA
Stomata	NA	NA	NA	t= 0.57 p= 0.57	t= 1.56 p=0.12	t=1.65 p=0.10
Elongated trichomes	NA	NA	NA	t= -6.17 p= 0.0000	t= -2.42 p=0.018	t= 2.09 p=0.052

Appendix 4: Results of the statistical analyses of baseline PM accumulation on leaves of species used in different experiments

Results of one-way Anova in the analysis of baseline PM densities on the leaves of plant species on the experimental living wall located along Leek Road, Stoke-on-Trent (n=20). (Chapter 5: Section 5.1.3.2)

Adaxial surfaces of the leaves			Abaxial surfaces of the leaves		
PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
F= 2.93 p=0.00005	F= 3.42 p=0.00003	F= 2.91 p=0.00006	F= 1.23 p= 0.24	F= 1.82 p=0.02	F= 2.19 p=0.004

Results of Student's t-test in the analysis of baseline PM densities on the leaves of *B. sempervirens* plants used in random and cluster planting designs of the living wall located along Leek Road, Stoke-on-Trent (n=20) (Chapter 5: Section 5.2.3)

Plant height	Tall plants			Short plants		
PM size fraction	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
Prior to replicate 1	t= 1.83 p=0.07	t= 1.60 p=0.11	t= 0.89 p=0.37	t= 0.74 p=0.46	t= 0.81 p=0.43	t= 0.63 p=0.53
Prior to replicate 2	t= 0.12 p=0.89	t= 1.16 p=0.25	t= 0.97 p=0.38	t= 0.12 p=0.89	t= 0.96 p=0.32	t= 0.18 p=0.79
Prior to replicate 3	t= 0.006 p=0.99	t= 0.78 p=0.44	t= 1.30 p= 0.19	t= 0.97 p= 0.41	t= 1.72 p= 0.09	t= 0.07 p= 0.93
Prior to replicate 4	t= 0.92 p=0.36	t= 0.53 p=0.59	t= 0.87 p=0.39	t= 1.69 p= 0.10	t= 1.09 p=0.26	t= 0.05 p=0.96

Results of one-way Anova in the analysis of baseline PM densities on the leaves of *Rubus* species used in an existing living wall located on Staffordshire University campus, Stoke-on-Trent (n=20). (Chapter 5: Section 5.3.2.1)

Adaxial surfaces of the leaves			Abaxial surfaces of the leaves		
PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
F= 1.82 p= 0.1	F= 2.04 p=0.06	F= 1.8 p= 0.10	F= 0.56 p= 0.76	F= 0.78 p= 0.58	F= 1.34 p= 0.24

Appendix 5: Results of paired t-tests[a] in the analysis of PM wash-off due to 16mm.hr^{-1} and 41 mm.hr^{-1} of rainfall on the leaves of four evergreen species grown in a living wall and a green screen located on the Staffordshire University campus along Leek Rd, Stoke-on-Trent, UK

Variation in PM wash-off due to rainfall on the adaxial surfaces of the leaves using a paired t-test															
Duration (min)	*Heuchera villosa* (16 mm.hr^{-1})			*Helleborus × sternii* (16 mm.hr^{-1})			*Bergenia cordifolia* (16mm.hr^{-1})			*Hedera helix* (16 mm.hr^{-1})			*Hedera helix* (41 mm.hr^{-1})		
	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
10	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	P<0.001	P<0.001	P<0.001
	t = 5.46	t = 5.22	t = 9.35	t = 5.28	t = 12.3	t = 6.39	t = 6.16	t = 6.77	t = 6.75	t = 5.17	t = 7.21	t = 5.93	t = 6.19	t = 3.98	t = 11.71
20	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 5.82	t = 4.33	t = 5.69	t = 8.86	t = 8.93	t = 14.8	t = 6.56	t = 5.41	t = 9.25	t = 6.37	t = 6.49	t = 6.54	t = 6.71	t = 4.91	t = 5.41
30	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 7.3	t = 9.14	t = 5.41	t = 7.36	t = 7.81	t = 8.14	t = 4.01	t = 5.23	t = 14.8	t = 7.72	t = 5.51	t = 5.8	t = 4.25	t = 5.17	t = 5.17
40	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 10.38	t = 13.78	t = 15.35	t = 13.74	t = 9.51	t = 13.91	t = 9.21	t = 11.8	t = 17.53	t = 14.52	t = 6.07	t = 12.16	t = 7.49	t = 9.01	t = 6.14
50	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 7.215	t = 8.04	t = 12.13	t = 8.72	t = 19.03	t = 59.4	t = 9.25	t = 9.14	t = 13.1	t = 8.53	t = 8.43	t = 8.21	t = 6.71	t = 8.59	t = 9.32
60	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 6.39	t = 12.17	t = 30.93	t = 27.37	t = 32.25	t = 20.04	t = 9.25	t = 9.14	t = 13.05	t = 15.78	t = 16.59	t = 44.42	t = 9.44	t = 8.95	t = 10.44
Variation in PM wash-off due to rainfall on the abaxial surfaces of the leaves using paired t-test															
	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
10	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 4.93	t = 7.75	t = 6.71
20	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 5.31	t = 5.17	t = 5.17
30	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 6.38	t = 7.48	t = 4.98
40	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 7.39	t = 2.83	t = 6.30
50	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 11.12	t = 7.75	t = 9.61
60	P=0.13	P=0.52	P=0.51	P=0.35	P=0.49	P=0.95	P=0.48	P=0.73	P=0.51	P=0.89	P=0.14	P=0.19	P<0.001	P<0.001	P<0.001
	t = 1.54	t = 0.66	t = 0.66	t = 0.95	t = 0.69	t = 0.05	t = 0.71	t = 0.34	t = 0.66	t = 0.13	t = 1.57	t = 1.36	t = 7.99	t = 6.94	t = 4.49

[a] in all cases n=20 for each species

Appendix 6: Sample images of leaves of species grown on living walls studied in this thesis.

a) Species grown on the living wall located adjacent to New Street station, Birmingham, UK

Armeria maritima

Blechnum spicant

Buxus sempervirens

Euphorbia amygdaloides

Gallium odoratum

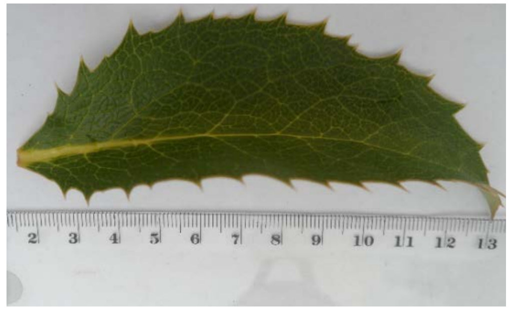
Geranium macrorrhizum

Hebe albicans

Hebe salicifolia

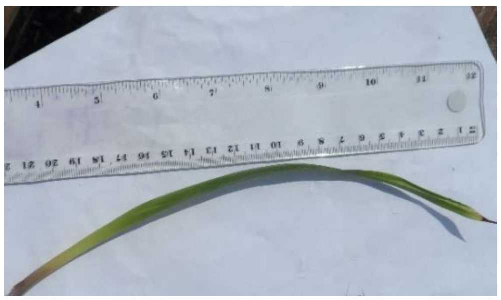
Hebe x *youngii*

Hedera helix



Helleborus x *sternii*

Hyssopus officinalis



Luzula nivea

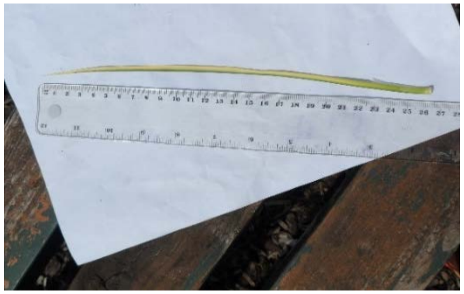
Pachysandra terminalis

Phyllitis scolopendrium

Primula verisg

Thymus vulgaris

b) Species used in the experimentally manipulable living wall located along Leek Road, Stoke-on-Trent, UK *(the ones given under New Street wall were excluded)*



Acorus gramineus

Berberis *buxifolia*

Bergenia cordifolia

Berberis × media

Carex caryophyllea

Deschampsia cespitosa

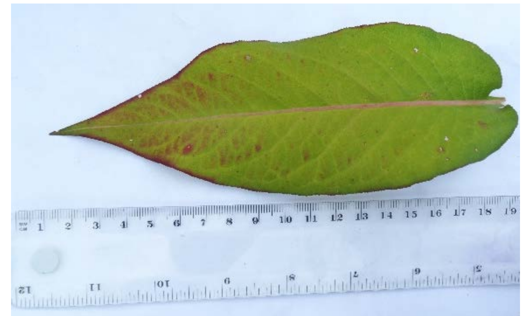
Geranium renardii

Heuchera Americana

Heuchera villosa

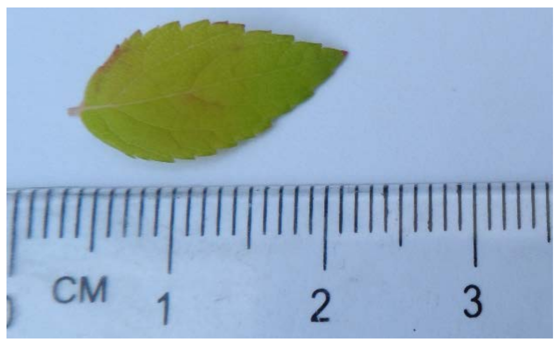
Juniperus chinensis

Lonicera kamtschatica



Persicaria amplexicaulis

Spiraea betulifolia



Spirea japonica

Stachys byzantina