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JANNE RÄSÄNEN

**THE ROLE OF NORTHERN TREE SPECIES IN THE CAPTURE
OF ATMOSPHERIC FINE PARTICLES**

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ABSTRACT

A massive load of particulate matter (PM) in the atmosphere originates from both natural and anthropogenic sources. Although anthropogenic sources, e.g. combustion for energy supply, road transport and industrial processes, contribute only 10% of global PM emissions, they dominate in the urban atmosphere. A high concentration of PM in the air can be seen as a haziness that is mostly caused by particles of the accumulation mode (0.05-2.5 μm), which have a relatively long lifetime in the atmosphere. These particulates worsen air quality and are associated with causing diverse cardio-pulmonary health problems.

Fine particles are removed from the atmosphere by several deposition mechanisms that depend on the particle size. Trees are three-dimensional objects that can capture PM from the atmosphere. In total, one third of the land area is covered by forests, of which boreal forests comprises 30%. Current knowledge on the particle removal efficiency of trees is rather limited and also controversial. A considerable mass of particles is removed from the atmosphere by trees and other vegetation, thus potentially reducing human health problems. On the other hand, trees can prevent the dilution of air pollutants, thus providing higher particle concentrations at the vicinity of the sources. Particle capture efficiencies of different tree species are related to leaf surface structure and leaf functional properties, but also depend on environmental variables.

In this thesis, four sets of wind tunnel studies were conducted in order to determine the particle capture efficiencies of five different tree species which grow in the boreal city environments. Two different study sets were designed to determine the effects of different soil drought on the particle capture efficiency of Norway spruce (*Picea abies*) saplings exposed to artificial 0.7 μm (geometric mean diameter, geometric standard deviation 3.0) NaCl particles propelled by air at a velocity of 3 m s⁻¹. Both previous and current year needles and different branch orientations were studied. The same experimental design was used to determine the fine particle capture efficiencies and deposition velocities of silver birch (*Betula pendula*), pubescent birch (*Betula pubescens*), common lime (*Tilia × vulgaris*) and Scots pine (*Pinus sylvestris*) saplings. The effects of stomatal conductance and anatomical

characteristics of the leaves and needles on particle capture were also analysed. The last study aimed to determine the the particle capture efficiencies of silver birch and pubescent birch when exposed to inert 0.27 μm (geometric mean diameter, geometric standard deviation 1.6) titanium dioxide (TiO_2) particles at air velocities of 1 m s^{-1} , 3 m s^{-1} and 6 m s^{-1} . The effects of air velocity and inert particle deposition on the biogenic volatile organic compound (BVOCs) emissions of the trees were also analysed. In addition, the particle penetration to the intercellular space of the silver birch leaves was studied by transmission electron microscopy.

The results of this thesis support earlier findings that coniferous species have higher capture efficiencies for particles of geometrical mean diameter of 0.7 (geometrical standard deviation 3.0) μm in size. The smaller unit size of conifer leaves compared to those of broadleaved species increased shoot complexity and increased particle deposition on conifers. Drought treatment also increased particle deposition on coniferous species, but not on broadleaved species. Moreover, particle capture of broadleaved species was increased by a rough leaf surface. Current year needles of Norway spruce under drought collected fewer particles than previous year needles. Branch orientation within the wind tunnels did not effect rates of particle deposition.

Increasing air velocity decreased deposition of TiO_2 particles of a geometric mean diameter of 0.27 μm (geometric standard deviation 1.6) on silver birch and pubescent birch leaves. These fine particles also penetrated through the stomata into the leaves. Short term fine particle exposure did not effect biogenic volatile organic compound (BVOC) emissions of silver birch or pubescent birch. Air velocity, however, altered BVOC emissions of the birches.

In conclusion, trees' water availability and leaf characteristics can have an impact on particle capture efficiency that should be considered in further studies and in estimation of pollution mitigation in a changing climate. Coniferous tree species could be favored for urban tree planting because of their more efficient particle collection capacities compared to broadleaved species. Among the broadleaved species, those with hairier leaves showed more efficient particle capture and are thus recommended for urban tree planting. The finding that solid fine particles can penetrate into inner parts of the leaves through stomata should be further studied, since nanomaterials are becoming more prevalent in built environments.

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CAB Thesaurus: air pollutants; particles; removal; deposition; pollution control; trees; urban environment; Picea abies; Betula pendula; Betula pubescens; Tilia europaea; Pinus sylvestris; experimental design; wind tunnels; volatile compounds; leaves; leaf area; stomata; water availability; drought

Yleinen suomalainen asiasanasto: ilmansaasteet; hiukkaset; pienhiukkaset; poisto; saostuminen; puut; kaupunkiympäristö; boreaalinen vyöhyke; kuusi; koivu; rauduskoivu; hieskoivu; puistolehmus; haittavat orgaaniset yhdisteet; lehti; vesi; kuivuus

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Janne Räsänen

ABBREVIATIONS

BVOC	Biogenic volatile organic compound
EDS	Energy dispersive spectroscopy
LAI	Leaf area index (m ² of leaf surface area – envelope area, single-sided, or double-side area, as defined in the pertinent text – per m ² of ground-surface area]
LRTAP	Long-range transboundary air pollution
PM	Particulate matter
PM _{0.1}	Particulate matter less than 0.1 μm in diameter
PM _{2.5}	Particulate matter less than 2.5 μm in diameter
PM ₁₀	Particulate matter less than 10 μm in diameter
RH	Relative humidity
SLA	Specific leaf area
TiO ₂	Titanium dioxide

ACRONYMS

CCSP	U.S. Climate Change Science Program
FAO	Food and Agriculture Organization of the United Nations
IPCC	Intergovernmental Panel on Climate Change
JRC	Joint Research Centre, European Commission's science and knowledge service
WHO	World Health Organization

SYMBOLS

C_p	Particle capture efficiency [dimensionless]
D_p	Particle diameter [aerodynamic, optical, mobility, as defined in the pertinent text]
D_{pg}	Geometric particle diameter
σ_g	Geometric standard deviation
V_g	Deposition velocity [cm s ⁻¹]

LIST OF ORIGINAL PUBLICATIONS

This thesis is based on data presented in the following articles, referred to by their chapter numbers.

- Chapter 2 Räsänen J V, Yli-Pirilä P, Holopainen T, Joutsensaari J, Pasanen P and Kivimäenpää M. 2012. Soil drought increases atmospheric fine particle capture efficiency of Norway spruce. *Boreal Environmental Research*, 17: 21-30.
- Chapter 3 Räsänen J V, Holopainen T, Joutsensaari J, Ndam C, Pasanen P, Rinna A 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.
- Chapter 4 Räsänen J V, Holopainen T, Joutsensaari J, Pasanen P and Kivimäenpää M. 2014. Particle capture efficiency of different-aged needles of Norway spruce under moderate and severe drought. *Canadian Journal of Forest Research*, 44: 831-835.
- Chapter 5 Räsänen J V, Leskinen J T T, Holopainen T, Joutsensaari J, Pasanen P and Kivimäenpää M. 2017. Titanium dioxide (TiO₂) fine particle capture and BVOC emissions of *Betula pendula* and *Betula pubescens* at different wind speeds. *Atmospheric Environment*, 152: 345-353.

AUTHOR'S CONTRIBUTION

- Chapter 2 Janne Räsänen was the main author of this article, he contributed to study design, construction of the wind tunnel, obtained plant material, collected research data and performed laboratory and scanning electron microscope analyses. Pasi Yli-Pirilä measured and analysed particle mass size distribution and contributed to reporting the results of the particle measurements. Toini Holopainen, Jorma Joutsensaari, Pertti Pasanen and Minna Kivimäenpää contributed to the study design and writing of the article.
- Chapter 3 Janne Räsänen was the main author of this article, he contributed to study design, obtained plant material and performed analyses by ion chromatography, scanning electron microscopy, porometer measurements of stomatal conductance, particle measurements and contact angle measurements. Collins Ndam performed wind tunnel exposures and analysed leaf areas. Åsmund Rinnan performed PCA analyses and built the PLS model. Toini Holopainen, Jorma Joutsensaari, Pertti Pasanen and Minna Kivimäenpää contributed to study design and writing of the article.
- Chapter 4 Janne Räsänen was the main author of this article, he contributed to study design, obtained plant material, collected research data and performed analyses by ion chromatography, scanning electron microscopy, porometer measurements of stomatal conductance and particle measurements. Toini Holopainen, Jorma Joutsensaari, Pertti Pasanen and Minna Kivimäenpää contributed to study design and writing of the article.
- Chapter 5 Janne Räsänen was the main author of this article, he contributed to study design, obtained plant material, collected research data and performed analyses of BVOC quantification by gas-chromatography-mass spectrometry, stomatal conductance and particle measurements. Jari T. T. Leskinen and Janne Räsänen performed scanning electron microscope analyses and Jari T. T. Leskinen provided expertise in EDS analyses. Minna Kivimäenpää performed analyses by transmission electron microscopy. Toini Holopainen, Jorma Joutsensaari, Pertti Pasanen and Minna Kivimäenpää contributed to study design and writing of the article.

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1 INTRODUCTION

1.1 BACKGROUND

Atmospheric fine particles originating from both natural and anthropogenic sources worsen living conditions and can even cause premature deaths. Residential combustion, energy supply systems, road traffic and long-range transportation produce most of the fine particles in Finland and the same trend is observed in other industrial countries (Karvosenoja et al. 2008). Fine particle pollution has been long associated with many adverse health effects (Dockery et al. 1993). Two major characteristics, size and chemistry, define the health effects of particulate matter. Fine particles smaller than 2.5 μm can easily penetrate into the respiratory system and even into the cardiopulmonary system, thus causing premature deaths, asthma symptoms and increased risk of respiratory infection (Schwartz, Dockery & Neas 1996, Pope III, Dockery 2006, Sturm 2012). Furthermore, the chemical characteristics of particles originating from burning processes, industry, and traffic are connected to major adverse health effects that are caused by the particles (Tsai, Apte & Daisey 2000). The most effective way to control fine particle emission is to control the emission sources or develop the burning process and filtering techniques to produce less particle emissions. Use of renewable energy sources can also affect particle emissions. In addition, the surrounding environment reduces particle pollution by absorbing it on surfaces. Forests or single trees are potential three-dimensional filters that could improve air quality by reducing particle concentration. Typically forests consist of several tree species of different ages with various forms. The particle capture efficiency (C_p) and deposition velocity (V_g) of different tree species can be compared in controlled laboratory experiments. This thesis provides results of particle capture efficiency tests for five different tree species and describes the leaf characteristics that affect the capture efficiency of particles.

1.2 ATMOSPHERIC FINE PARTICLES

1.2.1 Particle types, main sources and behavior

Aerosol is a suspension of fine solid or liquid particulate matter (PM) in a carrier gas, typically in air, with a particle size from 0.002 μm to over 100 μm . Particles are often classified according to their size, which is expressed as PM_x , where the subscript x refers to the upper diameter of the particles in μm (i.e. $\text{PM}_{2.5}$ meaning particles of diameter less than 2.5 μm). The finest particles occur in nucleation mode ($<0.05 \mu\text{m}$) and are born in the atmosphere from gaseous compounds via chemical reactions and condensation processes. Particle mass in nucleation mode is very low and particles grow by vapor condensation and through coagulation with other particles (Seinfeld, Pandis 2006). High concentrations of PM in air can be seen as a haziness, which is mostly caused by light interactions with particles of the accumulation mode (0.05-2.5 μm), which have relatively long lifetimes in the atmosphere (Hinds 1999, Seinfeld, Pandis 2006). Particulate matter borne by larger particles (with aerodynamic diameter greater than about 2.5 μm) settles relatively fast and close to the source due to gravitation, and is also subjected to significantly more efficient impaction mechanisms than smaller particles (Seinfeld, Pandis 2006).

Particles are also classified into two groups depending on their origin (Hinds 1999). Primary particles are released directly to the atmosphere from burning processes, biogenic sources (i.e. fragments of organisms, dust) or sea spray. Secondary particles are formed when gases undergo conversion to particles (e.g. when SO_2 and NO_x are oxidized in the atmosphere). Plant emitted biogenic volatile organic compounds (BVOC) can also act as precursors for secondary particles (Atkinson 2000).

Most of the fine particles originate from natural sources such as volcanoes, wildfires, dust storms, sea spray and vegetation (CCSP 2009). However, 10% of the total particle load originates from anthropogenic sources, which dominate in urban atmospheres (CCSP 2009). The main sources of a total of 1200 Gg of anthropogenic $\text{PM}_{2.5}$ in the European Union (year 2014) are commercial, institutional and household sources (56%), road transport (13%) and industrial processes (10%) (LRTAP Convention 2016). The typical annual mean $\text{PM}_{2.5}$ concentration in urban areas of Finland is less than 10 $\mu\text{g m}^{-3}$, with most EU cities reaching values from 10 $\mu\text{g m}^{-3}$ to 25 $\mu\text{g m}^{-3}$ (EEA 2016a). In heavily polluted urban areas, such as Nanjing in China, $\text{PM}_{2.5}$ can be over 480 $\mu\text{g m}^{-3}$ (Wang et al. 2002).

Ideally, the process of biomass burning would yield mainly CO_2 and water. However, there is poor control of the burning process in small scale burning units e.g. residential combustion, and thus, the emissions of particulate matter per unit of energy produced is higher than in power stations (Tissari 2008). The total PM mass in residential combustion depends on the fireplace, source material and its physical characteristics (e.g. moisture), but typical $\text{PM}_{2.5}$ emissions have been measured from about 33 to over 500 mg per produced MJ of energy (Savolahti et al. 2016). Particulate

matter consists of organic and inorganic materials, which are dominated by organic carbon (50%) and elemental carbon (12%) (Fine, Cass & Simoneit 2004, Schmidl et al. 2008). Other substances in particles include metals and ionic species (Schmidl et al. 2008).

Fine particles are removed from the atmosphere by several deposition mechanisms. Particles encounter gravitation, and thus sediment to surfaces over a certain time period. Particles larger than $0.5\ \mu\text{m}$ move in air streams, but due to their inertia they easily collide with obstacles and undergo impaction (Figure 1). Smaller particles are more inert against gravitational settling and impaction and are mainly deposited by random movements referred to as Brownian motion, where particles near to an obstacle can adhere to its surface (Figure 1). Particles can also intercept with surfaces when they are very near to them, or in some special cases electrostatic forces can attract particles to surfaces. Once particles hit the surface they attach tightly by adhesive forces (Hinds 1999). However, bounce-off can detach particles from surface thus having effect on net capture efficiency. In addition, growth of hygroscopic particles due to air humidity has effect on particle capture (Litschke, Wilhelm 2008). Particles can also be removed from the atmosphere by wet deposition via precipitation. Particles in the size range of $0.1\text{-}2.5\ \mu\text{m}$ are referred to as the accumulation mode due to the deposition mechanism having only a weak influence on particle removal (Hinds 1999, Sturm 2012). Overall, deposition velocities are higher in forest area than in grasslands (Pryor et al. 2008).

Measuring the actual size of fine particles is difficult due to their very small size and various shapes. Furthermore, different characteristics can be defined for atmospheric particles. The aerodynamic diameter of a measured particle is defined as that equal to a spherical particle with a density of $1000\ \text{kg m}^{-3}$ and the same settling velocity as the measured particle (Hinds 1999). Diameter can also be optically measured for particles larger than $50\text{-}100\ \text{nm}$, but the various sizes and shapes of the particles can affect the measurements. Many continuous particle size analyzers use a so called mobility diameter, which is obtained from measuring movements of charged particles in an electric field. The mobility diameter can be measured for particles in the range of $2.5\text{-}900\ \text{nm}$. Different fine particle diameters are not directly comparable, but they can be mathematically converted (Hinds 1999).

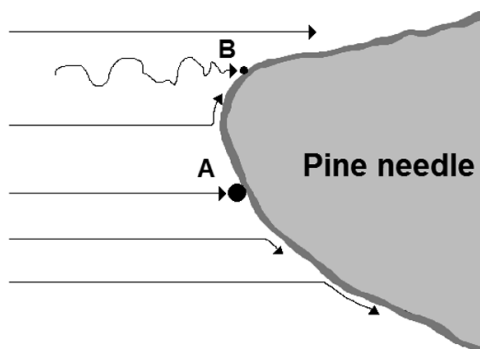


Figure 1. Edge of pine needle cross-section removing particles from the atmosphere by several deposition mechanisms. Larger particles typically impact with an obstacle (A) and smaller particles deposit by random movements called Brownian motion (B). Figure drawn based on deposition mechanisms introduced in Hinds (1999).

1.2.2 Particles and air quality

Transport of particles in the atmosphere starts the moment they are formed. PM_{2.5} has an approximately 1-6 days lifetime in the atmosphere, during which it can move up to 3000 km from the original emission location (WHO 2007). Several air pollution episodes in the past decades raised the issue of the effects of PM on human health (Anderson 2009). The most harmful effects of PM have been linked to PM_{2.5} (Schwartz, Dockery & Neas 1996) and later to PM₁ (Oberdörster, Oberdörster & Oberdörster 2005). The majority of adverse health issues linked to PM relate to cardiopulmonary problems, especially as a result of long term exposures (Pope III, Dockery 2006).

Wind speed has a major effect on particle deposition rate to surfaces, such as plant leaf surfaces, but whether deposition rate increases or decreases with increasing wind speed depends on the particle size. Particles larger than 0.8 µm are deposited more with increasing wind speed (Belot, Gauthier 1975, Beckett, Freer-Smith & Taylor 2000). Deposition rate of PM_{0.1} tends to decrease with increasing wind speed (Lin, Khlystov 2012). Deposition rate of larger particles can also decrease from theoretical values in strong wind because particles are not adhered to the leaf surface and often bounce off (Beckett, Freer-Smith & Taylor 2000). Air humidity can increase the size of hygroscopic particles (Stock et al. 2011), which can lead to higher deposition rate by increased inertia. Increasing temperature can increase formation of ozone and non-volatile secondary particulates, but their overall effect on new particle formation also depends on concentrations of other compounds such as sulphuric acid, nitric acid and ammonium nitrate (Aw, Kleeman 2003).

Solar radiation that is absorbed by the Earth and this warms our climate, whereas scattered or reflected solar radiation cools it down. This difference between absorbed and scattered solar radiation is expressed as radiative forcing ($W m^{-2}$) where negative values represent cooling of the climate and positive values represent warming of the climate. The total anthropogenic radiative forcing during the years 1750-2011 has been estimated to be $2.3 W m^{-2}$ (IPCC 2014), whereas for aerosols it was estimated to be $-0.4 W m^{-2}$ (Stocker et al. 2013). The cooling effect of aerosols is due to the fact that most of the aerosols scatter radiation, although the role of particles from biomass burning, secondary organic aerosols and minerals are still unclear. Only black carbon from fossil fuel and biofuel burning absorbs solar radiation and thus has a clear warming effect (IPCC 2014). Furthermore, atmospheric fine particles can also act as cloud condensation nuclei providing more clouds and thus increasing reflectance of solar radiation (albedo), which scatters radiation (Spracklen et al. 2008).

1.2.3 Effects of particulates on vegetation

Particulate pollution comes into contact with vegetation through the processes of wet and dry deposition, which can be substantial in magnitude due to natural particle

sources, e.g. sand storms and vegetation originated pollen, and anthropogenic sources, e.g. motorways and industrial processes (CCSP 2009). Deposition of nitrogen and sulphur containing particles can lead to soil acidification, thus reducing nutrient uptake and plant growth (Grantz, Garner & Johnson 2003). However, greater growth of forest stands have also been measured with increased nitrogen deposition (Solberg et al. 2009). Growth reduction of trees was observed in the vicinity of a rock quarry, where a heavy particle load on the forest soil increased alkalinity (Farahat, Linderholm & Lechowicz 2016). Particle deposition to leaf surfaces can cause visible symptoms due to photochemical reactions or can directly cover the leaf surface and block the stomata, thus reducing photosynthesis (Davies, Unam 1999, Grantz, Garner & Johnson 2003, Farahat, Linderholm & Lechowicz 2016). Most of the adverse effects of particles on vegetation occur at high levels of exposure, i.e. when vegetation is near to the emission source or when the particles have high toxicity, scenarios that are considered more as local problems (Grantz, Garner & Johnson 2003). In other words, vegetation and especially trees can collect and be exposed to remarkable amounts of particles, especially in the urban atmosphere, without suffering from serious injury. Therefore, they can be used efficiently in particulate pollution abatement.

Vegetation constitutively emits BVOCs from numerous tissues ranging from roots to the branches and leaves of the canopy (Loreto, Schnitzler 2010). The total global BVOC emissions have been estimated at 1000 Tg, of which half consists of isoprene (Guenther et al. 2012). Other greatly emitted BVOCs include various monoterpenes, methanol, acetone and ethanol (Guenther et al. 2012). In total, 1700 different compounds have been identified from plant emissions (Dudareva, Pichersky 2006). The BVOC composition and emission rates of vegetation can be altered by environmental conditions (Holopainen, Gershenson 2010). BVOCs are essential in communication between plants and other organisms and for defense against biotic and abiotic stresses (Loreto, Schnitzler 2010). Trees are important emitters of BVOCs, which can act as precursors for secondary organic aerosols that can have a cooling effect on the climate (Spracklen et al. 2008) and for ground level ozone that is harmful to plants and humans (Atkinson 2000, Guenther et al. 2012). Increasing amounts of secondary organic matter can lead to formation of new particles and cloud-condensation-nuclei and thus affect climate (Kavouras, Mihalopoulos & Stephanou 1998). The potential effects of particle exposure and deposition on leaf surfaces on BVOC emissions are not well understood.

1.3 FUNCTION OF TREES IN FINE PARTICLE CAPTURE

1.3.1 Forests and urban green areas

Approximately one third of the total land area of the world is covered by forest (FAO 2016). The largest forest areas are in Russia, Brazil and Canada, which comprise up to 40% of the total forest area (Figure 2). Finland has over 20 million hectares of forest and is part of the boreal forest region (taiga), which comprises 30% of the world's forest area (FAO & JRC 2012, Metla 2014). Forests have an important role in carbon sequestration from the atmosphere by photosynthesis, and current models of climate change mitigation suggest increasing afforestation and decreasing deforestation to improve global carbon storage (IPCC 2014). Forest loss in the 1990s was 0.18%, but despite the increasing demands of a growing human population the forest loss for the period 2010-2015 has declined to a level of 0.08%, which is 3.3 million ha per year (FAO 2016). Deforestation is greatest in South America and Africa, but the planted forest area is increasing worldwide by about 3.6 million ha per year (FAO 2016). Overall, the highest values for forest area per capita are in the boreal region, where it is over 6 ha per person. The lowest values are in the temperate forest area with less than 0.3 ha per person (FAO 2016).

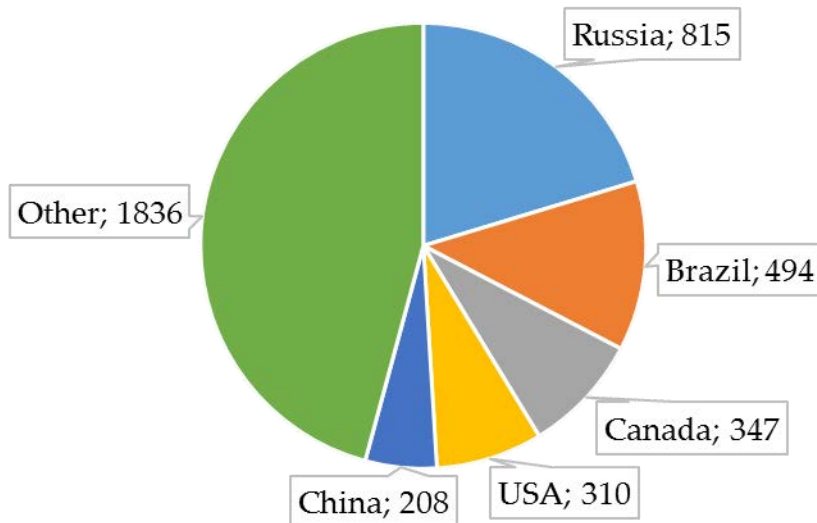


Figure 2. Forest land area (millions of ha) of the five most forested countries. The Other – segment includes all remaining countries. Data collected from FAO, 2016.

Geographical location and climate, as well as native and planted tree species, create the basis for the structure of urban forests. Nowak et al. (1996) reported tree cover ranging from 1% to 55% in U.S. cities with a population density over 386 people km⁻². The most forested cities were those formed inside forested areas (Nowak et al. 1996). More recent statewide examination in the U.S. has revealed Connecticut as having

the most (66.5%) tree covered urban areas and Montana and Wyoming the least (9%) tree covered urban areas (Nowak, Greenfield 2012). In Europe the urban tree cover in cities has been estimated to range from 1.5% to 62%, with an average cover of 30% (see Konijnendijk 2003). European forest resources have also been plotted on a map of total tree cover, with deciduous and coniferous species separated (EEA 2016b). The urban forest area in Finnish cities has not been the subject of much research and the exact values of percent cover in city areas is missing. Individual cities might have plans for maintaining urban forests and knowledge of the forested area, but a national compilation is unavailable.

Forests provide several benefits including ecosystem services (e.g. products such as wood and fiber), regulatory (e.g. effects on climate), cultural value (e.g. aesthetics) and health improvement (e.g. urban green areas) (Pataki et al. 2011, Tyrväinen et al. 2014). One important role of trees is removing particulate pollution (Beckett, Freer-Smith & Taylor 1998). Particle removal of PM_{2.5} that was annually 4.7 to 64.5 tonnes also has a monetary value worth millions of dollars in big cities, yet overall air quality improvement is at a rather low percentage (0.05% to 0.24%) of the total aerosol concentration (McPherson, Nowak & Rowntree 1994, Nowak et al. 2013). Pataki et al. (2011) questioned the pollution abatement capacity of trees and pointed out the lack of experimental studies in this field. In fact, the efficiency of trees at preventing sea fog particles of 20 µm in size from entering inland was identified decades ago (Hori 1953). Since then the focus has turned to finer particles and comparing particle removal by different tree species (Freer-Smith, Beckett & Taylor 2005). Setälä et al. (2013) reported significant effects of forest on PM₁₀ and PM_{2.5} removal in boreal forests of Southern Finland. Furthermore, PM concentrations near roads (carrying 8000 to 100 000 vehicles day⁻¹) were significantly lower in forested than open areas (Yli-Pelkonen, Setälä & Viippola 2017). In Italy, an urban wooded park with a total tree covered area of 116 ha, 42 ha comprising conifers and 74 ha comprising broadleaved trees, was measured and modelled to remove 12% of PM₁₀ and 2.6% of PM_{2.5} (Silli, Salvatori & Manes 2015). In Beijing, China, which has higher background values of PM_{2.5}, the planted forest area sustained higher particle levels than open areas during the day time, but removed particles during the night time (Xuhui, Xinxiao & Zhenming 2015). Particle levels could be higher in the day due to higher BVOC emissions that participate in the formation of new particles (Xuhui, Xinxiao & Zhenming 2015).

Forest structure and species composition are also important factors that affect particle capture efficiency (C_p). Particles are deposited 50%-60% more frequently at forest edges than the inner part of the forest (Reinap et al. 2012). PM_{2.5} accumulation on different tree species can vary by over 20-fold (Saebo et al. 2012). A denser tree canopy increases particle deposition rate (Xuhui, Xinxiao & Zhenming 2015). On the other hand, increasing the amount of avenue trees has been shown to increase particle pollution as a result of wind flow changes (Gromke, Blocken 2015).

1.3.2 Significance of canopy and leaf characteristics and environmental conditions

There is an undefined number of different tree species in the world that can be divided into broadleaved and coniferous species. The National Forest Inventory for 2012 distinguished four coniferous and 27 broadleaved indigenous tree species in Finland (Metla 2011). Scots pine (*Pinus sylvestris*) was the main species with 67% coverage, spruce (*Picea abies*) 22%, silver birch (*Betula pendula*) and pubescent birch (*Betula pubescens*) together 10% (Metla 2011). Single trees form a complex structure that changes during growth. Leaf shape and size have been shown to be important factors in particle capture (Beckett, Freer-Smith & Taylor 2000, Leonard, McArthur & Hochuli 2016). Leaf area measurements of trees – especially for coniferous species – can be laborious, yet are needed in many analyses (e.g. calculating particle capture efficiency and measuring stomatal conductance). For broadleaved species, the total leaf area is two times the projected leaf area. For coniferous species, there have been several different methods used to analyse needle area. Flower-Ellis and Ollson (1993) introduced a method that can be used to calculate the total area of Scots pine needles from the needle length. A method to determine the total needle area of Norway spruce was introduced by Sellin (2000), which uses minor needle width of the cross-section and projected needle area in calculations. Field studies often determine the leaf area of a canopy using a leaf area index (LAI), which represents the amount of one-sided leaf area per unit of ground surface area ($\text{m}^2 \text{m}^{-2}$) (Davies et al. 2016). The method for LAI measurement and analysis needs to be improved to overcome challenges in measurement accuracy for different types of forest e.g. broadleaved and coniferous forest (Wang et al. 2004).

Increasing complexity in the structure of trees seems to increase $\text{PM}_{2.5}$ deposition rate in the canopy (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Reinap et al. 2009). However, strong wind can bend foliage, which can adapt to be more streamlined and thus decreases impaction of particles and reduces the total particle deposition to below the theoretical maximum (Reinap et al. 2009). However, branch orientation did not affect $\text{PM}_{0.1}$ deposition rate on leaf surfaces in a wind tunnel study with pine (*Pinus taeda*) or juniper (*Juniperus chinensis*) (Lin, Khlystov 2012).

In general, coniferous species have been recorded to have far higher C_p values than most of the broadleaved species, which was linked to a more complex structure of conifers (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004). Single leaf size and posture together with branch orientation defines the complexity of the single tree form. Single leaf area is smallest for needles of coniferous species. Specific leaf area (SLA) is typically expressed as leaf area per unit dry weight (Sellin 2000). SLA for coniferous species has been reported to vary from $50 \text{ cm}^2 \text{ g}^{-1}$ to $79 \text{ cm}^2 \text{ g}^{-1}$ and for broadleaved species from $125 \text{ cm}^2 \text{ g}^{-1}$ to $386 \text{ cm}^2 \text{ g}^{-1}$ (Liu, Jin & Qi 2012). However, rather than SLA the actual area of a single leaf would better describe the obstacle that is the tree that particles face on deposition. Johansson

(1999) calculated that the average Norway spruce of age 17 to 54 years has 2.4 million needles yielding a 21.5 kg dry mass. Therefore, according to Liu et al. (2012), a single needle has a surface area of less than 1 cm². Typical broad leaved species such as birch can have two-sided leaf areas of over 20 cm² (Lihavainen et al. 2016). The difference is emphasized by the LAI values, which were reported for conifers to be in the range of 3 m² m⁻² to over 10 m² m⁻² and for broadleaved species to be from 4 m² m⁻² to 7 m² m⁻² (Bréda 2003, Gower, Norman 1991).

Leaf surface structure can also have an effect on particle deposition rate. For example, hairiness has been shown to increase particle deposition rate on leaf surfaces (Wedding et al. 1975, Little 1977). The stomata regions of coniferous needles are covered by a structural wax layer that has been shown to increase particle deposition rate compared to more smooth stomata surfaces (Burkhardt, Peters & Crossley 1995). Needle ageing can erode the wax layer (Grodzińska-Jurczak 1998), thus potentially having an effect on particle capture. Soil water availability has a major influence on tree physiology due to effects on the functioning of stomata. For example, soil drought induces a closure of stomata and thus lowers transpiration (Cornic 2000, Reynolds-Henne et al. 2010). Transpiration from the stomata can potentially prevent particle deposition by diffusiophoresis or increase deposition by cooling the leaf surface (Burkhardt, Peters & Crossley 1995, Hinds 1999). Rai et al. (2010) showed that PM₁₀ particles are more likely to be deposited near to stomata than other parts of the leaf. The overall effects of stoma function (i.e. transpiration), stoma density and stoma size on particle deposition on leaf surfaces needs to be clarified. Hydrophilic fine particles can also penetrate inside the leaf via stomata in a liquid solution (Eichert et al. 2008). However, penetration of solid and dry particles through stomata have not been described. In addition, other factors that can have an effect on plant physiology (i.e. water availability, age) and thus on particle deposition on leaf surfaces are not well known.

1.4 STUDY METHODS FROM LABORATORY EXPOSURES TO FIELD SURVEYS

There have been several estimates of the particle removal capacities of forest and trees of structured urban areas (McPherson, Novak & Rowntree 1994, Nowak, Crane & Stevens 2006, Nowak et al. 2013). These estimates require accurate measurements of the particle removal efficiencies of different tree species. The removal efficiencies of trees can be measured in the laboratory or in field surveys.

However such comparisons allow deposition mechanisms to be investigated and can provide confirmation that deposition models are accommodating all the mechanisms operating in the field and thus providing good estimates of total deposition. C_p and V_g values to modeled forest canopy, single trees, their parts and single leaves have usually been measured using laboratory experiments (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Ould-Dada 2002, Reinap et al. 2009, Lin, Khlystov 2012, Lin, Khlystov & Katul 2014). Laboratory experiments can be justified for particular research objectives, e.g. to investigate the effects of wind speed, particle material and species selection on C_p and V_g (Burkhardt, Peters & Crossley 1995, Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Reinap et al. 2009, Lin, Khlystov 2012, Lin, Khlystov & Katul 2014). Laboratory experiments can provide accurate species specific or single leaf specific data for particle deposition rate under controlled conditions. However, laboratory experiments are often short term trials and have restrictions compared to outdoor measurements (Ould-Dada 2002).

Field experiments have been conducted in urban green areas (Freer-Smith, Beckett & Taylor 2005) and natural forest sites (Ruijgrok, Tieben & Eisinga 1997). In natural systems the variables affecting deposition cannot be controlled, but prevailing conditions can be observed and recorded during studies (Yli-Pelkonen, Setälä & Viippola 2017). Field measurements provide information on the C_p and V_g of trees under real life circumstances. Since particle size, wind speed and surface humidity etc. have major roles in particle deposition rate on trees, the comparison between field studies and laboratory experiments can be challenging (Erisman, Draaijers 2003, Freer-Smith, Beckett & Taylor 2005, Yli-Pelkonen, Setälä & Viippola 2017).

1.5 OBJECTIVES OF THE THESIS

Several studies have shown the importance of trees in removing particles from the atmosphere, both in natural and laboratory systems (Hori 1953, Beckett, Freer-Smith & Taylor 2000, Freer-Smith, Beckett & Taylor 2005). Research has mainly focused on southern European species and a few Asian species, thus species typical of boreal forests are less studied (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Reinap et al. 2009). This thesis is aimed at adding knowledge on particle capture by five common tree species occurring in boreal forest and urban green areas.

Most wind tunnel studies have focused on the effects of wind speed or particle size on particle capture by trees (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, Beckett & Taylor 2005, Reinap et al. 2009), but there are also a few studies showing the importance of anatomical characteristics on particle capture (Burkhardt, Peters & Crossley 1995). This thesis aims to find out the effects of species specific leaf characteristics of common boreal tree species on particle capture. The effects of age related changes in needle structure on particle capture by coniferous tree species was also examined.

Soil water availability has a major influence on tree physiology, which results in closure of stomata during drought (Cornic 2000, Reynolds-Henne et al. 2010). This thesis aims to clarify the role of stomatal function on particle capture by trees.

It is well known that particles deposit on leaf surfaces, but the potential for particles to penetrate into leaf inner structure is not well understood. Eichert et al. (2008) showed that hydrophilic particles placed on leaf surfaces in liquid solution can penetrate inside the leaf via stomata. This thesis aims to clarify the role of inner parts of leaves in the capture of inert dry and solid fine particles.

BVOC emissions from vegetation can be increased in response to several external factors (Holopainen, Gershenson 2010). Stronger wind can cause damage that can lead to higher BVOC emissions by trees (Mochizuki et al. 2011, Bourtsoukidis, Bonn & Noe 2014), but movement of needles by mild wind did not increase BVOC emissions (Juuti, Arey & Atkinson 1990). However, the effects of wind on BVOC emissions of trees have received little attention. The potential for fine particle deposition on leaf surfaces to alter BVOC emissions of trees, either alone or in interaction with increasing wind speed, is not known and was one of the aims of this thesis.

The main objectives were formulated into six hypotheses that are shown in Table 1.

Table 1. The main topics and hypotheses of this thesis, which focused on determining the leaf structural and environmental characteristics that affect particle deposition rate on leaves of selected tree species. Particle penetration into leaves and the effects of wind speed and particles on BVOC emissions of the trees were also investigated.

Topic	Hypothesis
1) C_p of five boreal tree species	Coniferous species capturing more particles than broadleaved species (Chapters 2, 3, 4,)
2) Leaf characteristics and effects of needle ageing on C_p	Leaf properties that have an effect on C_p can be quantified. Ageing decreases particle deposition on needle surfaces of coniferous species due to them having a simpler wax structure (Chapter 2, 3, 4)
3) Effects of soil drought on C_p	Drought increases particle deposition on leaf surfaces by lowering stomatal conductance (Chapters 2, 3, 4)
4) Particle penetration into leaves	Fine particles can penetrate inside the leaf via stomata (Chapter 5)
5) Effects of wind speed on C_p of $PM_{0.5}$	Wind speed decreases C_p of $PM_{0.5}$ (Chapter 5)
6) BVOC emission after particle exposure at different wind speeds	Particle exposure alone, or in interaction with wind, alters BVOC emissions (Chapter 5)

1.6 OVERVIEW OF THE STUDIES

This thesis is based on controlled wind tunnel (Figure 3) studies conducted in laboratory conditions. Five different tree species were exposed to artificially produced fine particles. Particle concentration in the air was measured during the exposure and particles deposited on the leaves were determined after the trial. Four original research papers are represented in the following chapters. A short description of each experiment is shown in Table 2.

In chapter 2, two-year-old Norway spruce (*Picea abies*) saplings were installed (Figure 4) one at a time into the illuminated part (Figure 5) of the wind tunnel with a 3 m s^{-1} air flow. NaCl fine particles were mixed in the air flow and propelled towards the saplings. Particle concentrations in the air and on the surfaces of needles of different generations were measured to determine particle capture by the trees. Half of the saplings were well watered, whereas the other half was drought treated (Figure 6) to determine if the physiological state of the tree had an effect on particle capture. Furthermore, the effect of branch position on particle capture was studied at different sides of the airstream.

In chapter 3, particle capture efficiency and deposition velocity were determined for silver birch (*Betula pendula*), pubescent birch (*Betula pubescens*), common lime (*Tilia × vulgaris*) and Scots pine (*Pinus sylvestris*) saplings. In addition, several physiological and anatomical characteristics of the leaves and needles were analysed in order to determine their effects on particle capture. The effects of reduced soil water availability on particle capture efficiency was also studied with these four species.

The earlier finding that particle capture by Norway spruce increased with lower soil water content was further studied in chapter 4. Two-year-old Norway spruce saplings were divided into three groups; controls, short-term exposure to moderate drought and long-term exposure to severe drought. The effects of drought on needle stomatal conductance and anatomy were studied and their effects on particle capture efficiency were observed.

In chapter 5, the efficiency of silver birch and pubescent birch at capturing inert titanium dioxide particles was measured. In addition, the penetration of particles to the intercellular space of the leaves was studied. Three different wind speeds were tested during particle exposure to find out the possible effects of wind speed on particle capture efficiency and BVOC emission of the birches.

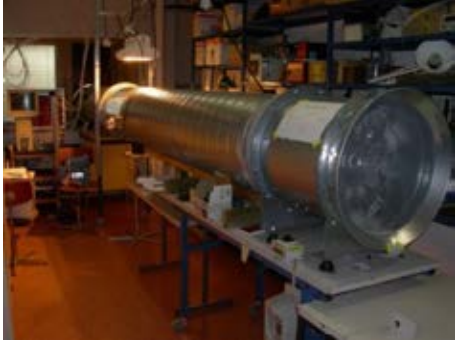


Figure 3. The wind tunnel system. The total length of the tunnel was 6 m and the diameter was 0.5 m.

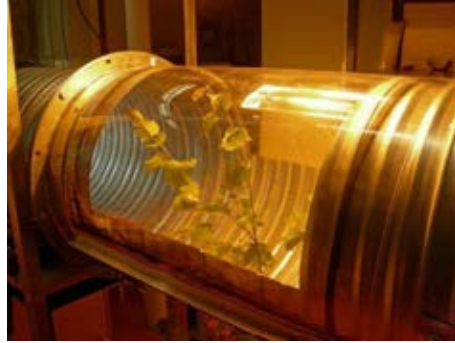


Figure 4. An adjustable input system located in mid-section of the tunnel allowed tall broadleaved saplings to be used in the experiments.

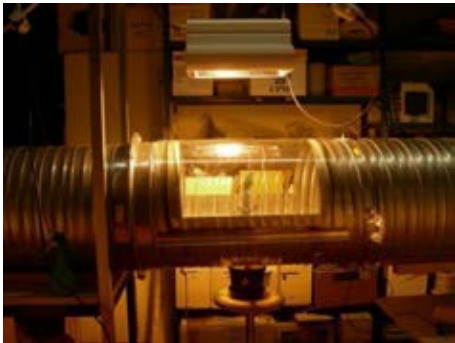


Figure 5. The exposed sapling was illuminated with a greenhouse light yielding $450 \mu\text{mol m}^{-2} \text{s}^{-1}$ at mid-canopy.



Figure 6. During the experiment drought treated and control saplings were maintained in growth chambers that had controlled growing conditions.

Table 2. A short description of the studies conducted to determine particle deposition on tree foliage. More details can be found in the cited chapters. GMD is the geometric mean diameter of the particles with σ_g geometric standard deviation.

Studied species	Main variables	Particles	GMD (μm)	σ_g	Mass concentration ($\mu\text{g m}^{-3}$)	Wind speed (m s^{-1})	Chapter
<i>Picea abies</i>	Surface deposition	NaCl	0.7	3.0	933	3	2
	Needle age						
	Substrate watering						
	Stomatal function						
	Branch position						
<i>Pinus sylvestris</i>	Surface deposition	NaCl	0.7	3.0	944	3	3
	Substrate watering						
	Leaf characteristics						
	Species differences						
<i>Picea abies</i>	Surface deposition	NaCl	0.7	3.0	1013	3	4
	Extended substrate watering						
	Needle age						
<i>Betula pendula</i>	Surface deposition	TiO ₂	0.27	1.6	207	1	5
	Particle penetration						
<i>Betula pubescens</i>	Air velocity					3	
	BVOC emission					6	

6 GENERAL DISCUSSION

6.1 SUMMARY OF THE MAIN RESULTS

Reduced soil moisture tended to increase particle capture by coniferous species due to reduced transpiration (i.e. decreased stomatal conductance and openness), especially in interaction with erosion of the stomatal wax layer. Soil moisture did not have an effect on particle capture by broadleaved species. Overall, coniferous species captured more particles than broadleaved species due to their small leaf unit area (shoot complexity). Among the broadleaved species, particle capture was increased by a rough leaf surface. Deposition velocity of finer, D_{pg} $0.27 \mu\text{m}$ ($\sigma_g = 1.6$), particles was greater at lower wind speeds. These inert particles also penetrated through stomata into the inner parts of the birch leaves. Wind speed altered BVOC emissions of the two studied birch species, but inert particle exposure did not affect the BVOC emissions. The main findings of the studies in this thesis are summarized in Table 3, which can be compared to hypotheses contained in Table 1.

Table 3. The key findings of the studies from each chapter in this thesis.

Chapter 2	<ul style="list-style-type: none">• Soil drought increased particle deposition on needle surfaces of Norway spruce• Particles were deposited equally on current and previous year needles• Branch orientation did not have an effect on particle deposition
Chapter 3	<ul style="list-style-type: none">• Small leaf unit area increased particle deposition• Leaf properties (e.g. hairiness, low wettability and low stomatal density) increased particle deposition• Moderate drought slightly increased particle deposition on Scots pine, but not on broadleaved species
Chapter 4	<ul style="list-style-type: none">• Previous year needles collected more particles than current year needles• Soil drought increased particle deposition on previous year needles
Chapter 5	<ul style="list-style-type: none">• Similar deposition values for <i>Betula pendula</i> and <i>Betula pubescens</i>• Increasing wind speed decreased particle deposition• Particles can penetrate into leaf tissue• Wind speed affected BVOC emission rates and profiles, but particulate pollution did not affect them

6.2 EFFECTS OF THE TREE SPECIES AND LEAF SURFACE CHARACTERISTICS ON PARTICLE DEPOSITION

Our results showed more efficient particle capture on coniferous species than on broadleaved species, which is in line with other studies (Table 4) conducted in the field (Freer-Smith, Beckett & Taylor 2005, Saebo et al. 2012) and wind tunnels (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Hwang, Yook & Ahn 2011). Scots pine had the highest particle capture efficiency in our studies (Table 4), which was also higher than those of earlier studied coniferous species such as Douglas fir and Corsican pine (Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004). Of the tested broad leaved species, pubescent birch tended to have the highest particle capture efficiency, even exceeding that of coniferous Norway spruce (Table 4). Results of Sæbø et al. (2012) showed silver birch to be the most effective broadleaved species at collecting particles, which was particularly evident for PM_{0.2}. However, it should be noted that C_p of birches was nearly 30 times lower for particles of 0.27 µm than for particles of 0.7 µm (Table 4). Particle deposition rate in the canopy is a delicate process that is strongly modified by several parameters, e.g. increasing wind speed (Beckett, Freer-Smith & Taylor 2000), particle size (Hinds 1999) and humidity of hygroscopic particles (Litschke, Wilhelm 2008). Therefore, care is needed when results of different studies conducted in variable settings are compared.

The surface areas of single leaves were 20-40 times smaller in Scots pine than in broadleaved species, which increased particle deposition rate on Scots pine (Chapter 3). It is known that small single leaf areas increase the complexity of canopy structure, which in turn increases particle deposition rate (Beckett, Freer-Smith & Taylor 2000). On the contrary, Norway spruce had similar C_p values to those of birches, even though they had even smaller needles than Scot pine (Table 4). The smaller old needles of Norway spruce captured more particles than the larger sized current year needles, but the difference in size was too small to explain the difference in C_p (Chapter 4).

Leaf hairiness and surface wettability increased particle deposition rate on broadleaved species (Chapter 3). Hairiness has been suggested to increase particle deposition rate on other broadleaved trees and vegetation, although this has not been confirmed by quantitative methods (Little 1977, Beckett, Freer-Smith & Taylor 2000, Baraldi et al. 2011). Glandular trichomes did not increase particle deposition rate on leaf surfaces (Chapter 3). Surface wettability decreases with increasing water repelling forces such as a more waxy leaf surface. Burkhardt et al. (1995) observed greater particle deposition rate on more waxy coniferous species. Our two experiments with Norway spruce gave somewhat controversial results because the first study showed a higher overall C_p on needles with a well preserved stomatal wax layer (Chapter 2), whereas the second study showed a higher particle capture efficiency on older needles with less structured waxes (Chapter 4). However,

although the structured wax layer was different it is difficult to analyze possible differences in the wettability of the surface due to the contact angle not having been measured in that experiment. It is also possible that low stomatal conductance had a stronger effect on C_p of less waxy needles, thus hydrophilic particles could have been deposited more efficiently on the hydrophilic surfaces of the needles.

Table 4. Particle capture efficiencies ($C_p\%$) of the tree species tested in wind tunnels with a 3 m s^{-1} wind speed and an average aerodynamic particle diameter (D_{pg}) of $PM_{2.5}$. Mean values \pm SE are calculated both for deciduous ($n=16$) and coniferous ($n=6$) tree species. This table is modified from Table 2 in Chapter 3 and updated with the latest data.

	C_p %	D_{pg}	σ_g	Reference
Deciduous	0.043 ± 0.011			
<i>Acer campestre</i>	0.01	1.3 μm	0.07	Beckett et al. (2000)
<i>Acer pseudo-platanus</i>	0.01	0.8 μm	0.18	Freer-Smith et al. (2004)
<i>Alnus glutinosa</i>	0.02	0.8 μm	0.18	Freer-Smith et al. (2004)
<i>Betula pendula</i>	0.043	0.7 μm	3.0	Chapter 3
	0.0015	0.27 μm	1.6	Chapter 5
<i>Betula pubescens</i>	0.083	0.7 μm	3.0	Chapter 3
	0.0029	0.27 μm	1.6	Chapter 5
<i>Cupressocyparis leylandii</i>	0.13	1.3 μm	0.07	Beckett et al. (2000)
<i>Eucalyptus globulus</i>	0.003	0.8 μm	0.18	Freer-Smith et al. (2004)
<i>Ficus nitida</i>	0.01	0.8 μm	0.18	Freer-Smith et al. (2004)
<i>Fraxinus excelsior</i>	0.03	0.8 μm	0.18	Freer-Smith et al. (2004)
<i>Populus deltoids x trichocarpa</i>	0.02	1.3 μm	0.07	Beckett et al. (2000)
<i>Quercus petraea</i>	0.14	0.8 μm	0.18	Freer-Smith et al. (2004)
<i>Quercus sessiliflora</i>	0.06	2.0 μm	1.1	Belot and Gauthier (1975)
<i>Sorbus intermedia</i>	0.07	1.3 μm	0.07	Beckett et al. (2000)
<i>Tilia x vulgaris</i>	0.047	0.7 μm	3.0	Chapter 3
Coniferous	0.13 ± 0.028			
<i>Picea abies</i>	0.06	0.7 μm	3.0	Chapter 2
	0.024	0.7 μm	3.0	Chapter 4
<i>Pinus nigra</i>	0.15	1.3 μm	0.07	Beckett et al. (2000)
<i>Pinus sylvestris</i>	0.21	0.7 μm	3.0	Chapter 3
	0.15	2.0 μm	1.1	Belot and Gauthier (1975)
<i>Pseudotsuga menziesii</i>	0.16	0.8 μm	0.18	Freer-Smith et al. (2004)

6.3 EFFECTS OF SOIL DROUGHT AND AIR VELOCITY ON PARTICLE DEPOSITION

Soil drought increased particle deposition rate on Norway spruce needles (Chapters 2, 4) and a similar trend was observed with Scots pine needles (Chapter 3). Reduction in stomatal conductance, determined for both drought treated Norway spruce and Scots pine (Chapters 2-4), can increase particle deposition rate by lower diffusionphoresis (Hinds 1999). Deposited particles can also increase the surface roughness, which can further increase particle deposition rate (Burkhardt, Peters & Crossley 1995). Previous year needles had greater degradation of the stomatal wax structure (Chapter 4), which increases surface hydrophilicity (Neinhuis, Barthlott 1998) and can thus increase the deposition rate of hydrophilic particles. Particle deposition rate to broadleaved species remained similar despite the drought treatment. This was noted for both of the studied birch species, even when the drought treatment reduced the single leaf areas, which was expected to increase C_p (Chapter 3). Rai et al. (2010) showed that particles were more likely to be deposited on the stoma area than on other parts of the leaf of the annual plants studied. In our study, the density of stomata in pubescent birch leaves was less than half that of lime leaves, but pubescent birch still captured more particles (Chapter 3). This emphasizes the significance of leaf characteristics other than stomatal density in explaining species differences in particle capture efficiency. Overall, soil drought did not have an effect on the C_p of broadleaved species, but increased the C_p of coniferous species.

Increasing air velocity affects particle deposition rate, but the direction of the effect is related to particle size so that deposition by diffusion ($D_p < 0.5 \mu\text{m}$) is decreased and deposition by impaction ($D_p > 0.5 \mu\text{m}$) is increased (Hinds 1999). The strongest increase in C_p of PM_{10} was found for small leaved species i.e. *Pinus nigra* (0.13% to 0.38%), *Cupressocyparis leylandii* (0.08% to 0.25%) and *Sorbus intermedia* (0.04% to 0.13%) with particle deposition tripled as wind speed increases from 1 m s^{-1} to 3 m s^{-1} (Beckett, Freer-Smith & Taylor 2000). This was also the case for *Acer pseudo-platanus* with C_p two times higher at a wind speed of 6 m s^{-1} (0.033%) than 3 m s^{-1} (0.014%) (Freer-Smith, El-Khatib & Taylor 2004). However, C_p of PM_{10} on coniferous *Pseudotsuga menziesii* almost twice as efficient at a wind speed of 3 m s^{-1} (0.42%) than 6 m s^{-1} (0.27%) (Freer-Smith, El-Khatib & Taylor 2004). Deposition rate of fine $\text{PM}_{0.1}$ particles to leaf surfaces was decreased with increasing wind speed (Lin, Khlystov 2012), which was also seen in our study with $0.27 \mu\text{m}$ particles (Chapter 5). Particle deposition rate of PM_{10} on leaf surfaces depends on several factors, such as surface characteristics and particle size, but the effects of increasing air velocity seems to be predicted by theory of aerosol filtration by porous media, with the deposition rate of smaller ($0.27 \mu\text{m}$) particles (Chapter 5) decreased, and the deposition rate of larger ($0.8 \mu\text{m}$) increased (Freer-Smith, El-Khatib & Taylor 2004).

6.4 PARTICLE INTAKE INTO LEAVES

This was the first study to show that solid and dry particles can penetrate through the stomata into the intercellular space of silver birch leaves (Chapter 5). This was the case even with very low C_p values on leaf surfaces (Table 4). The penetration was possible to demonstrate in our study material by using inert TiO_2 particles (Chapter 5).

There has been great interest in determining the pathways of solute particles inside the leaf tissue. In particular, many agricultural sprays, such as lipophilic pesticides and herbicides and ionic foliar fertilizers, need to pass the leaf cuticle for proper function (Schreiber 2005). Nanoparticles can also penetrate stomata in liquid solution, which can be a faster route to the inner tissue than via the cuticle (Eichert et al. 2008). On a rainy day this could form a pathway for particles deposited on leaf surfaces to enter the plant via the stomata. Our studied broadleaved tree species had substantial variation in the size and density of the stomata (Chapter 3), which is an important factor if stomata can control the size of the particles entering the leaf (Eichert et al. 2008).

A higher particle load can also clog the stomata (Hirano, Kiyota & Aiga 1995). Rai et al. (2010) showed that over 75% of the stomata on the upper leaf surface of annual plant species were occluded after spraying with urban dust. Coniferous species typically have a stomatal wax layer that can prevent particle intake, but may increase particle deposition rate on needle surfaces (Burkhardt, Peters & Crossley 1995). Air pollution can damage these stomatal wax layers in conifers, thus opening the route for particle uptake. Increasing use of nanomaterials and the possibility for pathogens to enter leaves via stomata increases the importance of further research on both dry deposition uptake and liquid solution particle uptake.

6.5 BVOC EMISSIONS AND PARTICLE DEPOSITION

Plants produce a large variety of BVOCs whose emission rates and composition differ between species and even among individuals of the same species (Hakola et al. 2001, Bäck et al. 2012, Kivimäenpää et al. 2013). High variation in BVOC composition and emission rates can complicate analyses of factors that affect BVOC emissions. Nevertheless, wind clearly had an effect on BVOC emissions of the tested birch species (Chapter 5). A similar effect of wind has been observed in a hemiboreal forest with Norway spruce, silver birch and black alder as dominant tree species, where a wind speed greater than 2 m s^{-1} doubled the emission of sesquiterpenes compared to values measured at a wind speed of 1 m s^{-1} (Bourtsoukidis, Bonn & Noe 2014). Wind has also been shown to increase monoterpene emissions of Japanese cypress (*Chamaecyparis obtusa*) (Mochizuki et al. 2011), while monoterpene emissions of Monterey pine (*Pinus radiata*) remained similar irrespective of wind speed (Juuti, Arey & Atkinson 1990).

The silver birch and pubescent birch used in our studies (Chapter 5) differed in their leaf properties (Chapter 3). Silver birch had more glandular trichomes (which are terpenoid storage structures) (Biswas et al. 2009, Schollert et al. 2017), and had the highest BVOC emissions at a medium wind velocity that could have mechanically damaged the trichomes (Chapter 5). The highest wind velocity may have caused a stronger BVOC release, but because the emission measurements were done after the exposure and storage structures may have been emptied, it was not detectable. Emissions of a few prevailing green leaf volatile (GLV) compounds increased for both species with increased air velocity (Chapter 5), which may be due to mechanical stress caused by increased membrane damage (Vuorinen, Nerg & Holopainen 2004, Mithöfer, Wanner & Boland 2005).

A short term exposure of birches to fine particles did not have an effect on BVOC emission (Chapter 5). Therefore, it is likely that deposition rate of inert fine particles with a low mass loading to leaf surfaces does not affect BVOC emissions. However, long term or heavy deposition of particles can cover leaf surfaces and have a consequent effect on plant physiology (i.e. photosynthesis and function of stomata) (Hirano, Kiyota & Aiga 1995, Rai, Kulshreshtha & Srivastava 2010), which may have a further effect on BVOC emissions (Holopainen 2011). In addition, the direct influence of certain PM pollution (i.e. salts) that can cause symptoms on leaf surfaces through chemical effects (see Burkhardt 2010) could have the potential to affect BVOC emissions.

6.6 MITIGATION OF PARTICLE POLLUTION BY TREES

Many of the anthropogenic emissions of fine particulate matter can be efficiently controlled by filtering or by adjusting the burning process and by using renewable (e.g. solar) energy sources. Nevertheless, particle concentrations originating from several anthropogenic and natural sources may have adverse effects on human health and the surrounding nature. Particles are deposited on surfaces at a certain rate that depends on the particle size and environmental conditions. Trees are three-dimensional objects that hinder a particle's journey in the atmosphere and thus improve air quality. The size range of fine particles is very broad, from nanometers to hundreds of micrometers, and particles across that size range have totally different deposition mechanisms. In general, the focus of studies on particle capture by trees has changed from coarse particles (Hori 1953) to PM₁₀ (Belot, Gauthier 1975) and further to PM_{2.5} or PM₁ (Chapters 2-5, Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Reinap et al. 2009), which are considered to cause most of the adverse effects on human health (Pope III, Dockery 2006). Furthermore, increasing use of nanomaterials (PM_{0.1}) has increased interest in researching the deposition rate of these very fine particles on trees (Lin, Khlystov 2012).

Pope et al. (2009) used regression models incorporating changes in fine particle pollution concentrations during the 1980s and 1990s and human life expectancy to show that decreasing PM_{2.5} in the air by 10 µg m⁻³ would yield 223 more living days per resident. Several wind tunnel studies have shown a rather low C_p% of single trees (Table 4) and similar values (0.05% to 0.24%) have been modeled for air quality improvement by urban forest trees in a study of PM_{2.5} in ten U.S. cities (Nowak et al. 2013). Whitlow et al. (2014) calculated, by using the maximum air quality improvement values of Nowak et al. (2013), that doubling the number of trees in New York City would yield only slightly more than five more living days per resident. Pataki et al (2011) reviewed the ecosystem services provided by urban nature and proposed a low potential for trees to improve air quality. Moreover, it was suggested that the overall understanding of the topic is highly uncertain (Pataki et al. 2011). Several researchers in the field of urban ecology have expressed their concern that the potential for air quality improvement by trees capturing fine particles has been exaggerated from the study results (Nowak et al. 2013, Whitlow et al. 2014). However, urban trees produce numerous other advantages, e.g. aesthetics, cooling effects, shelter for urban animals, etc. that are favourable and justify increasing the amount of urban trees (Pataki et al. 2011). The urban growth environment can set special needs for species based on, e.g. air and soil pollution, demands for trimming, water availability and low amount of leaf littering or pollen production.

There are also studies showing that single trees can capture particles from the atmosphere, thus reducing their concentration in the air (Freer-Smith, Beckett & Taylor 2005, Saebo et al. 2012). Maher et al. (2013) showed a remarkable decrease (>50%) in indoor PM₁₀ levels after placing young birches (*Betula utilis* 'Doorenbos') in a single line in front of houses. On the forest scale, a reduction of 23% to 40% in the

PM₁₀ concentration compared to open spaces has been measured; however, the reduction did not correlate with canopy closure, number and size of the trees or the extent of ground vegetation in a mostly deciduous northern forest (Setälä et al. 2013, Yli-Pelkonen, Setälä & Viippola 2017). In this context, particle removal efficiency of trees could be optimised by selecting tree species such as evergreen conifers or deciduous species with complex leaf structures that are known to be efficient particle collectors (Table 4).

Overall, trees capture fine particles and the deposition rate on leaf surfaces depends on the species characteristics and environmental factors that have effects on tree physiology. Results from outdoor measurements and wind tunnel studies indicate that coniferous species capture more particles than broadleaved species (Chapter 3, Beckett, Freer-Smith & Taylor 2000, Freer-Smith, El-Khatib & Taylor 2004, Saebo et al. 2012). Particle deposition rate on broadleaved species having more surface structures i.e. hairs was greater than on those having more plain structure (Chapter 3). Single tree lines can affect air circulation in the vicinity of roads and can thus affect particle concentrations at road sides and inside buildings (Maher et al. 2013, Gromke, Blocken 2015). Substantial particle removal by single trees is not likely due to quite low values for particle capture by single trees being recorded, both in earlier and in the present studies (Table 4.). At the forest-scale particle removal in total mass can be substantial, although single trees only capture a small share of particles passing them (Freer-Smith, Beckett & Taylor 2005, Saebo et al. 2012, Setälä et al. 2013, Yli-Pelkonen, Setälä & Viippola 2017).

6.7 METHODOLOGICAL CONSIDERATIONS

The wind tunnel built for this study was made of a cylindrical air duct equipped with an axial fan. In the first test trials the air flow was spiraling in the duct. Wind conditions were improved by inserting a honeycomb structure after the fan to settle the wind into a more laminar flow. Fine particles were produced by atomizing NaCl water solution with an aerosol generator (Chapters 2-4). Inert TiO₂ particles were produced from powder by a powder disperser (Chapter 5). Particles produced from liquid solution should be dry before deposition since wetness enhances deposition rate and distort results (Joutsensaari et al. 2001). In our experiments, the relative humidity (RH) was measured in each run and in the different experiments it was 11% (Chapter 2), 48% (Chapter 3) and 49% (Chapter 4). This is one important factor that may affect the results of the different studies. The laboratory air came from the building air ventilation system, thus the RH was not adjustable and changed during the year, but it remained constant during each study set. In addition, the RH of the growth chamber where the saplings were maintained was adjusted to be near to the RH of the wind tunnel (excluding Chapter 2) to avoid the influence of changing RH.

Fine particles (PM_{2.5}) were produced at a level of 1000 µg m⁻³ (Chapters 2-4), which is higher than those measured in southern Finland (12 µg m⁻³) or in urban areas like in Nanjing, China (480 µg m⁻³) (Pakkanen et al. 2001, Wang et al. 2002). In short term exposures, the particle load has to be rather high for analytical purposes. With the particle concentrations used, the surfaces of leaves and needles were rarely covered after a two hour exposure period. However, collision of particles together (coagulation) might be more probable with a higher particle concentration in the air, potentially leading to a lower particle number concentration but having no effect on particle mass concentration. For the different deposition mechanisms of particles, size is also an issue when selecting measurement devices. When the particle number concentration is measured, the fraction of finer particles is favored, while when measuring mass concentration the fraction of larger particles dominates the measurements (Janhäll 2015).

Wind tunnels have constant wind flow and our study showed a significant reduction in stomatal conductance after 10 minutes of wind exposure. However, the effects of lowered stomatal function due to reduced soil water availability was connected to particle capture by the trees. Thus, it would be of interest to further study the effects of stomata on particle capture in chambers introduced by Hwang et al. (2011), where more turbulent air circulates around the tested plant and probably enables higher stomatal conductance than in the constant flow of a wind tunnel.

Fine particles were produced in two different ways, as discussed above. NaCl was chosen for inexpensive production and analytical costs, which enabled the completion of many replicates and the studying of more tree species. Atomizing NaCl water solution was more stable for particle production than production of TiO₂ particles by brushing the powder, which needed more adjustment during each experimental run. NaCl was analysed by ion chromatography from washings of the

leaves, but the analysis of TiO₂ was more laborious involving a scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS). However, the use of inert TiO₂ particles enabled us to determine that solid particles can penetrate through the stoma and into the inner part of the leaf, which would have been difficult with NaCl particles because both elements occur in plant cells and particles are water soluble.

BVOCs emitted from the trees were collected after a two hour run in the wind tunnel. Half of the trees were exposed to TiO₂ particles and the other half were used as controls. The air volume that passed through the tree during the experiment was 1400 m³, 4200 m³, 8500 m³ for wind speeds of 1 m s⁻¹, 3 m s⁻¹ and 6 m s⁻¹, respectively. The limit of detection of the analytical devices that we had was not sufficient for instant measurements of BVOCs. For a lower volumes of air in the wind tunnel a proton-transfer-reaction mass spectrometer (PTR-MS) could be used due to it having a very low detection limit, i.e. a 10⁻¹² share of the volume. By comparison, a PTR-MS has been successfully used for in situ measurements of BVOCs in a spruce forest (Bourtsoukidis, Bonn & Noe 2014). Many BVOCs are emitted instantly after stress, therefore occurring within the two hour exposure period used in our experiments (Chapter 5). As our measured BVOCs were collected at the end of the exposure, they were more likely reflecting the past effect of wind induced stress on BVOC emissions.

Overall, studies conducted in laboratory conditions are often criticized for a lack of direct reference to a natural environment. Air flow in a wind tunnel is typically moving constantly in one direction, thus the penetration of wind and the load of particles on the surface of vegetation placed in the tunnel can be high (Janhäll 2015). In nature, wind faces barriers such as buildings, trees and other shapes that have effects on wind flow (Janhäll 2015). This can be seen as a higher particle deposition rate at forest edges compared to the inner parts (Reinap et al. 2012). Street trees can also prevent air flow from street canyons, thus reducing the dilution of pollutants in the air and leading to higher pollution concentrations in the street canyons (Gromke, Blocken 2015). However, systems in nature are complicated, variable and unpredictable, which makes study arrangements and interpretation of results challenging. The strength of the wind tunnel studies is the possibility to control variables, thus allowing certain particle capture mechanisms to be more accurately studied than in natural system (Chapters 2-5, Beckett, Freer-Smith & Taylor 2000, Reinap et al. 2009).

6.8 CONCLUSIONS

Trees are living organisms that can capture fine particles from the air. According to this thesis, particle capture depends on environmental conditions that have effects on tree physiology and growth. The effects of soil drought were especially evident for coniferous species, which captured more particles when soil moisture was low. The effect of stomata remains controversial since results of this thesis suggest a low density of stomata to increase particle capture, but with particles frequently found near to stomata. Particle deposition on leaf surfaces with increasing air velocity seems to follow aerosol theory, with deposition rate of the lower range of PM_{10} decreasing and deposition rate of the higher range of PM_{10} and larger particles increasing. Small leaf size and greater surface roughness increases particle deposition rate on leaf surfaces.

Particle penetration to the inside of the leaf through stomata has earlier been demonstrated for liquid solutions, but this thesis provides new evidence of penetration by inert dry particles through the stomata to the inside of the leaf. Coniferous species typically have a wax layer that prevents particle penetration through the stoma, but environmental conditions can degrade this wax layer thus enabling particles to reach these stomata. Particle uptake should be further studied to determine the intensity and the effects of particle penetration through stomata of different species.

Short term deposition of inert particles on leaf surfaces did not affect BVOC emissions of birches. Air velocity had an effect on BVOCs emitted from birches, which could be due to mechanical damage of BVOC storage structures i.e. glandular trichomes. Overall, trees emit high amounts of BVOCs that participate in the production of secondary organic aerosols and thus affect climate and cause health issues.

Urban trees have several different functions in aesthetics, culture, animal shelter, temperature control and many more. If particle pollution abatement is the main priority of species selection, coniferous species should be favored. Occasional drought in paved urban areas could even increase particle capture efficiency of conifers. Long-term particle measurements in urban wooded areas and control measurements in treeless areas could confirm the findings of these wind tunnel studies.

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This thesis aims to find out the effects of species specific leaf characteristics of common boreal tree species on particle capture. The effects of age related changes in needle structure on particle capture by coniferous tree species was also examined. In addition, the effects of soil water availability on particle capture and particle penetration into leaf inner structures were studied. Urban trees have several different functions in aesthetics, culture, animal shelter, temperature control and many more. Results of this thesis increases knowledge of particle removal of boreal trees and can be used in urban planning.



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