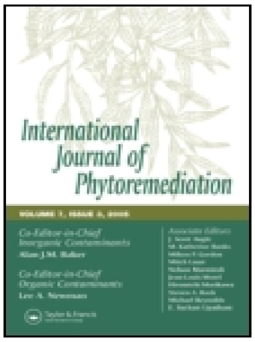

International Journal of Phytoremediation

ISSN: 1522-6514 (Print) 1549-7879 (Online) Journal homepage: https://www.tandfonline.com/loi/bijp20

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To cite this article: Kajetan Dzierżanowski , Robert Popek , Helena Gawrońska , Arne Sæbø & Stanislaw W. Gawroński (2011) Deposition of Particulate Matter of Different Size Fractions on Leaf Surfaces and in Waxes of Urban Forest Species, International Journal of Phytoremediation, 13:10, 1037-1046, DOI: 10.1080/15226514.2011.552929

To link to this article: https://doi.org/10.1080/15226514.2011.552929

Accepted author version posted online: 01 Jun 2011.
Published online: 12 Jul 2011.

International Journal of Phytoremediation, 13:1037–1046, 2011

ISSN: 1522-6514 print / 1549-7879 online
DOI: 10.1080/15226514.2011.552929

DEPOSITION OF PARTICULATE MATTER OF DIFFERENT SIZE FRACTIONS ON LEAF SURFACES AND IN WAXES OF URBAN FOREST SPECIES

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Particulate matter (PM) is an air contaminant in urban and industrial areas that often exceeds limit values, creating serious problems due to its harmful effects on health. Planting trees and shrubs as air filters is a way to improve air quality in these areas. However, further knowledge on species effectiveness in air purification is essential. This study compared four species of tree (**Acer campestre** ***L.,*** **Fraxinus excelsior** ***L.,*** **Platanus × hispanica** ***Mill. ex Muenchh. 'Acerifolia',*** **Tilia cordata** ***Mill.), three species of shrub (*****Forsythia × intermedia** ***Zabel,*** **Physocarpus opulifolius** ***(L.)*** **Maxim., Spiraea japonica** ***L.), and one climber species (*****Hedera helix** ***L.) that are commonly cultivated along streets in Poland to capture fine, coarse and larger particles from air. Separate gravimetric analyses were performed to quantify PM deposited on surfaces and trapped in waxes. Significant differences were found between the plant species tested. The distribution of different particle size fractions differed between and within species and also between leaf surfaces and in waxes.***

KEY WORDS: air pollutants, phytoremediation, coarse particles, fine particles, wax, urban forest

INTRODUCTION

Particulate matter (PM), a common air contaminant, is a mixture of solid and liquid substances of organic and inorganic character suspended in air. Particles vary in terms of origin, chemical composition and size. The size is described as aerodynamic diameter and ranges from 0.001 to 100 μm (AQEG 2005). Particles are often defined as coarse (2.5–10 μm), fine (0.1–2.5 μm), and ultra fine ($\leq$0.1 μm) (Beckett, Freer-Smith, and Taylor 1998 and references therein).

Particulate matter is emitted by anthropogenic sources: vehicle exhausts, road dust, coal burning, industrial processing, cement, and fertiliser production. However, under some circumstances natural sources such as volcanic eruptions, forest fires, sandstorms, and soil

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and rock erosion also release substantial amounts of PM to the atmosphere. PM may also derive from nucleation, condensation or coagulation of nitrogen oxides, sulphur dioxide, ammonia, and volatile organic compounds present in the air as gaseous pollutants. These are called secondary particles and are generally smaller than 2.5 μm ($PM_{2.5}$) (USEPA 2004). Particles, especially $PM_{2.5}$, often contain highly toxic polycyclic aromatic hydrocarbons (PAH), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs) and heavy metals, making them more hazardous and carcinogenic (Caricchia et al. 1999; Jouraeva et al. 2002; USEPA 2004; Ariola et al. 2006; Yu et al. 2006). Particulate matter containing some organic pollutants may be of a lipophilic nature and thus able to penetrate the wax layer covering leaves and young twigs. There is a lack of data in the literature concerning the relative distribution of deposited particulate matter between plant surfaces and the wax layer.

Air quality standards in the European Union, introduced with European Council Directive 1999/30/EC, distinguish particles with diameter of 10 μm and smaller (PM_{10}) as being of most importance with regard to public health, with limit values established as an annual average of 40 μg m^{-3} and a daily average of 50 μg m^{-3} not to be exceeded more than 35 times a year (EC 1999). According to the EU Directive, by 2010 measures should be in place to lower the annual average value to 20 μg m^{-3}.

In urban and industrial areas, particles from vehicle exhausts, road dust and production processes often exceed limit values, especially in hotspots, i.e., sites with particularly high levels of pollution. This carries a risk of harmful effects on human health, a risk that increases with exposure to small particles, which are able to penetrate deeper into the lungs, even to the alveolar regions (Dockery et al. 1993; Kampa and Castanas 2008). Children seem to be especially vulnerable to air pollution (Salvi 2007). Short-term exposure to PM in inhaled air may cause an increase in cardiopulmonary disease, while long-term exposure can lead to chronic disease and reduced life expectancy as a result of cardiopulmonary mortality and lung cancer (WHO 2005). EEA Report 2 (2007) estimated that the life of the average European is nine months shorter because of exposure to PM_{10}. The areas most affected are BeNeLux, Poland, the Czech Republic, Hungary, the Po Valley in Italy, and southern Spain. In some of these areas life expectancy may be reduced by up to three years (EEA 2007).

Plants play an important role in filtering ambient air by adsorbing particulate matter onto leaf surfaces. Trees, with their large total leaf area, are considered the most effective type of vegetation for this purpose (McDonald et al. 2007). The structure of tree crowns leads to turbulent air movements, which increase PM deposition on leaves (Fowler et al. 1989). Some species-specific features of leaves may enhance this air filtration process, e.g., trichomes (Smith and Staskawicz 1977) and the chemical composition and structures of epicuticular waxes (Jouraeva et al. 2002; Kaupp et al. 2000). For example, leaves of broad-leaved species, which have rough surfaces, are more effective in capturing PM than those with smooth surfaces (Beckett et al. 2000). In addition, needles of coniferous trees, which produce a thicker epicuticular wax layer, are more effective in PM accumulation than broad-leaved species (Beckett et al. 1998). Moreover, evergreen conifers have the potential for accumulating toxic pollutants throughout the year. On the other hand, since most of these plants keep their needles for several years, there is no possibility of recycling PM accumulated on needles every year, as is the case for deciduous species. In addition, conifers are in general less tolerant to high traffic-related pollution, especially if salt is used for road de-icing during winter, and they are often not recommended for roadside plantings. Therefore, evergreen conifers may not be as useful as deciduous leafy species, in spite of their high efficiency in PM scavenging (Beckett et al. 2000).

Some particles that are captured can later be washed off from the leaves with rain and deposited in soil/ground, where natural processes decompose the organic components of PM, while the inorganic components are accumulated in soil and the soil solution.

Although relatively much is known about the mechanisms of particulate deposition on vegetation (Farmer 2002), less is known about the differences between species in PM accumulation. Such information is important, especially in view of the large number of tree and shrub species and cultivars being used in urban areas. Choice of species and planting design could have a major influence on the PM filtering performance of urban vegetation. Furthermore, little is known about the capture of particles larger than PM_{10} by plants.

The aim of this study was to determine the quantity of particulate matter deposition on foliage of eight plant species commonly cultivated in urban areas of Poland in terms of: (1) particle size fractions and (2) particles accumulated on leaf surfaces and those trapped in waxes.

MATERIALS AND METHODS

Plant Material

Four tree species, three shrub species and one climber commonly grown along busy urban streets and roads in Poland were selected for this study. All plants had already been growing in the selected locations for several years and were in good condition, healthy and free from pests. Field maple (*Acer campestre* L.) is considered one of the most tolerant trees for urban conditions. European ash (*Fraxinus excelsior* L.), London plane (*Platanus* × *hispanica* Mill. ex Muenchh. ‘Acerifolia’), small-leaved lime (*Tilia cordata* Mill.) and border forsythia (*Forsythia* × *intermedia* Zabel) are very popular in Polish cities. Common ivy (*Hedera helix* L.) is a much used evergreen climber. Common ninebark (*Physocarpus opulifolius* (L.) Maxim.) and Japanese spiraea (*Spiraea japonica* L.) are becoming increasingly popular due to their ornamental value and tolerance to the urban environment.

Study Sites and Sample Collection

The plants studied were growing at highly polluted sites in the city centre of Warsaw, Poland (along KEN avenue and the streets Rodowicza “Anody,” Rosola, and Solidarnosci). For each species, leaves were harvested from four plants (replicates) in two growing seasons ($n = 8$). Tree species were examined in 2007 and 2008 and the other species in 2008 and 2009. In order to obtain sufficient material to determine the fine fraction of PM and still avoid filter blockage by particles during filtration, the leaf area per sample ranged between 300 and 400 cm^2. This leaf area was found to be suitable for washing off particles in the rinse liquids used (water, chloroform). Leaf samples of all species were collected in October, at the end of the growing season, from the traffic-exposed side of the plant at 0.6–2.0 m height above ground level, depending on plant structure. Samples of leaves were placed in paper bags, labelled, transported to the laboratory and kept at ambient temperature until analysis. In both seasons samples were collected from the same individuals and from the same parts of the plants.

Quantitative Analysis of PM and Epicuticular Waxes

The filters used for the analysis were first dried for 30 minutes at 60°C in a KCW-100 drying chamber (PREMED, Poland) and then left in the weighing room to stabilise the humidity of hygroscopic paper filters before weighing. After a further 30 minutes, the filters were pre-weighed on a XS105DU balance (Mettler-Toledo International Inc., Switzerland). In order to avoid electrostatic charges on the filters, they were passed through a deioniser gate (HAUG, Switzerland) before weighing.

Every sample of leaves was placed in a glass container with 250 mL of water and agitated for 60 seconds in order to wash off particles from leaf surfaces. These represent particles that can be washed off the leaves during rainfall (for simplicity, in this paper termed 'surface PM'). The water was then filtered using a metal sieve (Haver and Boecker, Germany) with mesh diameter 100 μm in order to eliminate particles larger than 100 μm. The water was next filtered using a 47 mm glass filter funnel with stopper support assembly (PALL Corp., USA) connected to a vacuum pump (KNF Neuberger, Inc., USA), first on pre-weighed paper filters Type 91 (retention 10 μm) and next on Type 42 filters (retention 2.5 μm) and finally on PTFE membrane filters (retention 0.2 μm) (all Whatman, UK). In order to reduce surface tension on the PTFE membrane, a few droplets of isopropyl alcohol were placed on the membrane before filtration. Thus, three fractions of particulate matter were collected on filters: (1) Large: 10-100 μm, (2) Coarse: 2.5–10 μm, and (3) Fine: 0.2–2.5 μm. Initially (i.e., for tree species), only the two larger fractions of PM were assayed, but later, as we improved our methodology, the fine particulate matter fraction was also determined (on shrubs and the climber). Filters were then dried and post-weighed with the same procedure as in pre-weighing to calculate the mass of PM in each fraction of every sample.

After rinsing with water, each sample of leaves was washed with 150 ml of chloroform for 40 seconds in order to dissolve the epicuticular wax layer from leaf tissues and to wash out particles trapped in waxes (termed 'in-wax PM'). The filtration procedure was the same as for water-rinsed particles, with the exception that isopropyl alcohol was not used.

The times used for filter drying, cooling, and leaf rinsing with water and chloroform were selected on the basis of preliminary tests.

The mass of washed-off waxes dissolved in chloroform was assayed for every leaf sample in pre-weighed beakers after chloroform evaporation. Total area of the leaf sample was measured using Image Analysis System (Skye Instruments Ltd, UK) and SkyeLeaf software, which allowed the amount of PM and waxes to be expressed as μg cm^{-2} leaf area. Although particulate matter was washed off from both the adaxial and abaxial surfaces of the leaves, the amount of PM was expressed per one surface of leaf area (as measured by image analysis), as is used for calculation of LAI values and in other physiological studies.

Statistical Analysis

Data were subjected to one-way analysis of variance using StatGraphics Plus 4.1 software (StatPoint Technologies, Inc., USA). Significance of differences between mean values was tested using Tukey's honestly significant difference test (HSD) at $\alpha = 0.05$. Values presented on bar charts are means ± SE, n = 8, while values on dot charts are individuals with trend line and correlation coefficient (r). The latter two were calculated using Microsoft Excel (Microsoft Corp., USA)

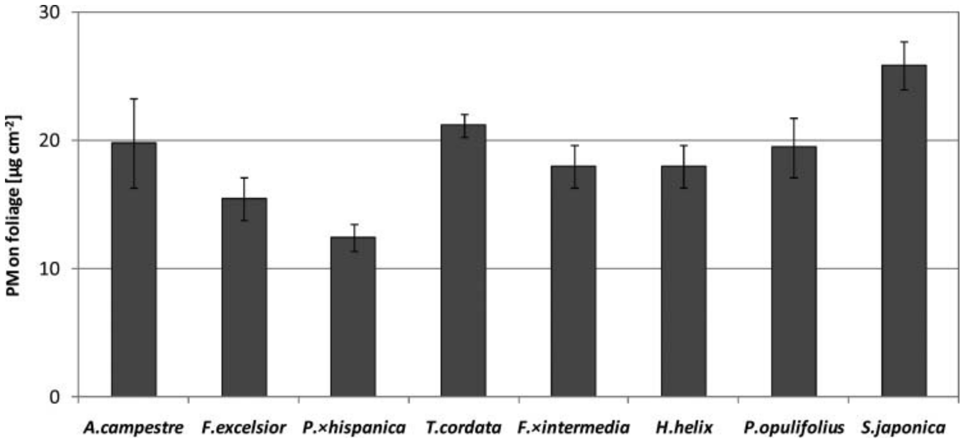

Figure 1 Total mass of PM (μg cm^{-2}) accumulated on leaves of the eight broad-leaved plant species examined. Data are mean $\pm$ SE, n $=$ 8 (two growing seasons with four replicates in each).

RESULTS AND DISCUSSION

Data on the total PM accumulated on the leaves of the eight plant species studied are presented in Figure 1. All tested species captured some particles on their leaves, but the quantity of PM differed significantly between the species. *Spiraea japonica*, a small shrub with dense leaves, was found to be the most effective in PM accumulation, while large, branchy trees of *Pl.* $\times$ *hispanica* were least effective, with over twofold difference between these contrasting species. Similarly, significant differences in the quantity of particulate matter deposited on leaves were reported for five tree species by Beckett et al. (2000). Even greater genotypic differences than those recorded in the present work have been found in an ongoing three-year study with 22 tree and 23 shrub species cultivated at clean sites under near identical growing conditions in plant nursery (authors' study, in preparation).

Since the objective of the present study was to evaluate the air purifying properties of urban plant species and their potential to decrease the health hazards in polluted cities, we opted to distinguish between three size fractions of PM. The reason for this is that they have different health impacts. The amounts of these three size fractions in surface PM and in-wax PM are presented in Figure 2 (a, b, and c).

Large particles (10–100 μm in diameter) appeared mainly on the leaf surfaces (Figure 2a). This means that most of this fraction can easily be washed off during rain events or dislodged by wind and thus these large particles may ultimately be deposited on the ground. *Hedera helix* accumulated the highest quantity of large particles (surface PM), but the difference was significant only between *H. helix* and *For.* $\times$ *intermedia*. For all species there was less mass of particles of this size deposited in epicuticular waxes, but there were large differences between the species. *Tilia cordata*, *S. japonica,* and *A. campestre* had about fourfold more large particles in waxes than *H. helix* (Figure 2a).

Smaller particles (diameter below 10 μm) are considered to be more harmful for human health, with fine ones being more dangerous than coarse (Dockery et al. 1993). Therefore selection of plants for urban air purification should be based on their ability to collect these PM fractions. The coarse fraction (2.5–10 μm) mainly appeared as surface PM, but *A. campestre*, *Fr. Excelsior,* and *Pl.* $\times$ *hispanica* accumulated more of these particles in waxes (Figure 2b). *Spiraea japonica* was most effective in accumulating this fraction,

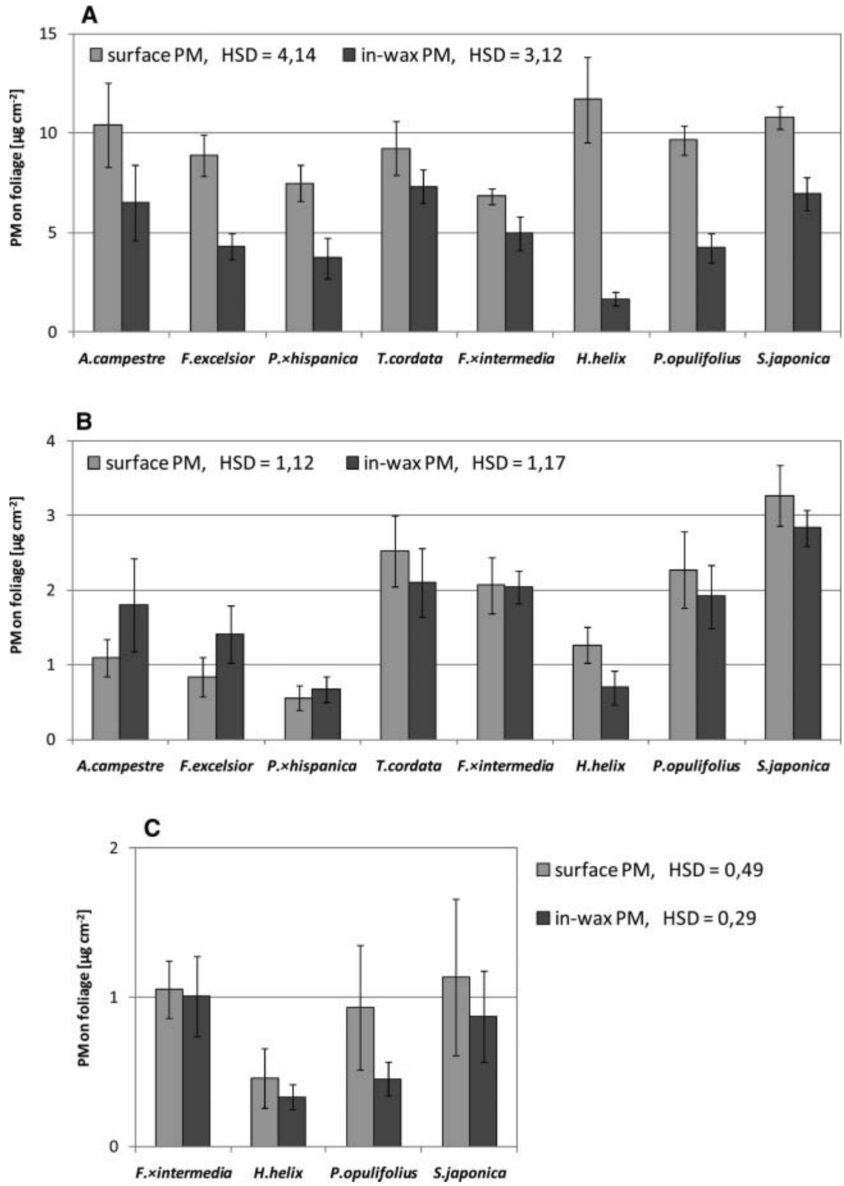

Figure 2 Mass (μg cm^{-2}) of surface PM and in-wax PM with particle diameter (A) 10–100 μm, (B) 2.5–10 μm, and (C) 0.2–2.5 μm accumulated on leaves of the eight plant species. Data are mean ± SE, n = 8 (two growing seasons with four replicates in each).

with a total of about 50% more coarse particles than *T. cordata*, *For.* × *intermedia* and *Ph. opulifolius*, and between twofold and almost fivefold more than the remaining species (Figure 2b).

The filtration method and conditions for determination of fine particles were refined over the study period, so this fraction was measured only for the shrubs and climber (Figure 2c). *Forsythia* × *intermedia* and *S. japonica* were more effective in capturing fine particles on foliage than the two other species. *Ph. opulifolius* accumulated the same amount of surface PM as these two species, but only half as much in-wax PM. *Hedera helix* appeared to have the smallest amount of fine PM deposited, both on surfaces and in the wax layer (Figure 2c).

The results showed that the examined species accumulated large quantities of particulate matter. The largest quantity of PM occurred in the form of large particles, i.e., 10–100 μm diameter followed by significantly smaller amount of coarse ones (2.5–10 μm). The quantity of fine PM (0.2–2.5 μm) was the smallest.

Similarly, Beckett et al. (2000) found more coarse PM than fine deposited on the leaves of five tree species. However, Freer-Smith et al. (2005), in a similar study, reported that when leaves of two conifer species were harvested for analysis in winter time, more fine PM than coarse was found on needles. In contrast to our data, Ottelé et al. (2010) reported particles $\geq$ 10 μm to be rather rare in visual counts of PM on ESEM photographs of *H. helix* leaves. They found the greatest number of particles with the smallest diameter (0.5–1.0 μm), then between twofold and eightfold fewer particles with diameter 1.0–5.5 μm and very few with diameter 5.5–10 μm. However, in our preliminary microscopic studies we observed particles of all fractions studied here, with the large particles (10–100 μm) being present quite frequently. It should be borne in mind, however, that comparisons of data collected using different methods may be of little relevance, for example due to lack of a direct relationship between the number of particles in a given size fraction and their mass.

In all plant species examined in the present study, particles of each size fraction were found in the epicuticular wax layer. However, the quantity of large particles was always smaller in the wax layer than on the surface. For the smaller PM fractions, this difference was much less evident. Moreover, there were species in which the total amount of in-wax PM was either nearly the same or even greater than surface PM (Figure 2a, b, and c). To our knowledge, this is the first time such data have been reported. In a study still in progress with 22 tree and 23 shrub species, the results so far show that in all these species, cultivated in plant nurseries under nearly identical growing conditions, PM accumulates both as surface PM and in-wax PM (authors' study, in preparation).

The total quantity of waxes on the washed leaf areas was determined in order to examine possible relationships between particulate matter deposition in the wax layer and the amount of waxes (Figure 3). Leaves of *For.* $\times$ *intermedia* had between twofold and eightfold more waxes than other species. However, no significant relationship was found between mass of waxes and total PM, surface PM, in-wax PM, PM with diameter 10–100 μm (both on surface and in waxes), coarse PM (both on surface and in waxes) and fine surface PM. A moderate correlation ($r = 0.79$) was observed only between wax amount and quantity of fine in-wax particles when analysis was performed on the total data of the four species tested (3 shrubs and a climber) (Figure 4a).

No relationship was found between amount of waxes and quantity of PM deposited on the leaves. Similar findings were reported by Jouraeva et al. (2002) in a study with *Tilia* $\times$ *euchlora* and *Pyrus calleryana*. The potential of wax in trapping particles may thus depend more on the chemical composition and structure of the epicuticular wax layer, which are species-specific traits (Post-Beittenmiller 1996; Kaupp et al. 2000; Jouraeva et al. 2002), than on wax quantity. Since these features should not differ within a taxon, the relationship between wax amount and PM deposition of every fraction was also analysed for each species separately. A significant correlation ($r = 0.93$) was found only between the quantity of wax and deposited coarse in-wax PM in leaves of *T. cordata* (Figure 4b).

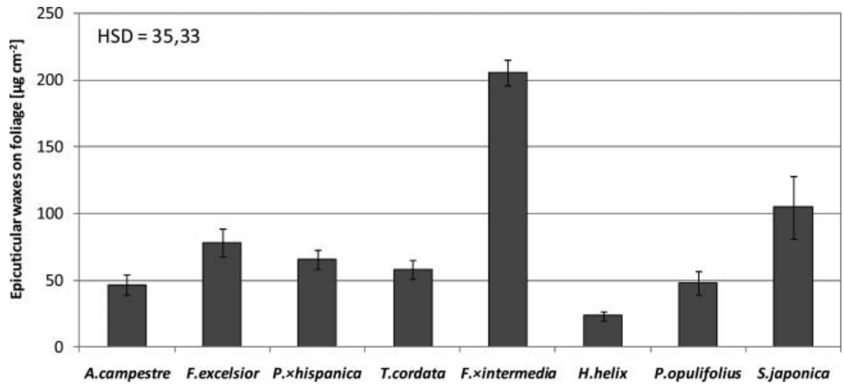

Figure 3 Mass (μg cm^{-2}) of epicuticular waxes extracted from leaves of the eight species. Data are mean ± SE, n = 8 (two growing seasons with four replicates in each).

The other species demonstrated only weak or no relationships. There were even some cases where increasing amount of waxes was weakly correlated with decreasing quantity of particulate matter.

Plants scavenging particulate matter will also remove the associated PAHs and heavy metals from the air. Therefore, plants are important for decreasing the levels of such pollutants in the air in urban areas. Research conducted in Beijing (China), where air pollution is very high, showed that trees in the city centre removed 772 tons of PM_{10} during one year (Yang et al. 2005). In similar studies in Chicago (USA) urban trees, which occupy 11% of city area, removed about 234 tons of PM_{10} (Nowak 1994). In the whole USA, urban trees and shrubs remove about 215 kilotons of PM_{10} every year (Nowak et al. 2006). In studies in UK cities, McDonald et al. (2007) found that planting trees on one quarter of the available urban area was able to reduce the PM_{10} concentration by between 2 and 10%. This would have a direct positive effect on human health, in addition to all the other well-known positive effects of vegetation on the urban environment and people's well-being. In fact, in the case of the open air, there is no other purification option except the use of plants. This environmental biotechnology is called phytoremediation. Air

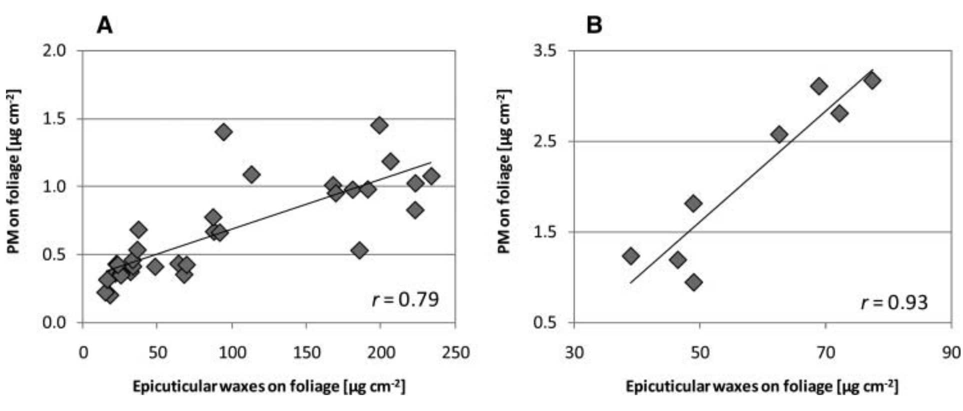

Figure 4 Relationship between amount of waxes on foliage (μg cm^{-2}) and (A) mass of in-wax fine particles (μg cm^{-2}) for all eight species; and (B) in-wax coarse particles (μg cm^{-2}) for small-leaved lime.

phytoremediation, as a solution for air quality improvements, is relatively cheap, easy to introduce and environmental friendly technology. However, it should be borne in mind that leaves with significant loads of PM deposited during the growing period in heavily polluted areas should be collected after natural autumn defoliation and disposed of in a controlled manner.

Knowledge about the efficiency of plant species and cultivars in filtering and channelling polluted air and their tolerance to urban environments is essential in devising measures to improve air quality. Landscape architects and planners need such knowledge in order to design appropriate vegetation for given urban sites, hotspots in particular. The role of plants as an urban 'green liver' (Burken 2003) can then be exploited, in addition to their other well-documented benefits (Brack 2002).

CONCLUSIONS

- Plants of all species tested accumulated particulate matter (PM) of large (10–100 μm), coarse (2.5–10 μm), and fine (0.2–2.5 μm) fraction sizes. The PM was deposited on leaf surfaces and trapped in waxes.
- The largest quantity of deposited PM observed consisted of large particles, while there were smaller quantities of coarse and fine particles.
- The quantities of in-wax PM depended on species and particle size fraction.
- The eight species examined differed significantly in their ability to capture particulate matter. *Spiraea japonica* was most while *Platanus* × *hispanica* least effective.
- Plants of all species examined contributed to particulate matter scavenging and thus improved air quality.

ACKNOWLEDGMENTS

This study was supported by a grant from Norway through the Norwegian Financial Mechanism, # PNRF-193-AI-1/07 granted to S.W. Gawroński and A. Sæbø.

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