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Quantifying Particulate Matter Accumulated on Leaves by 17 Species of Urban Trees in Beijing, China

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Abstract

Airborne particulate matter (PM) has become a serious environmental problem and harms human health worldwide. Trees can effectively remove particles from the atmosphere and improve the air quality. In this study, a washing and weigh method was used to quantify accumulation of water-soluble ions and insoluble PM on the leaf surfaces and within the wax of the leaves for 17 urban plant species (including 4 shrubs and 13 trees). The deposited PM was determined in three size-fractions: fine (0.2–2.5 μm), coarse (2.5–10 μm), and large (> 10 μm). Significant differences in the accumulation of PM were detected among various species. The leaves of *Platycladus orientalis* and *Pinus armandi* were the most effective in capturing PM. Across the species, 65% and 35% of PM on average deposited on the leaf surface and in the wax, respectively. The greatest PM accumulation by mass on leaves was in the largest PM size fraction, while accumulation of coarse and fine particles size fractions was smaller. Water-soluble ions accumulated on leaf surfaces contributed 28% to the total PM mass on average. This study demonstrated that leaves of woody plants accumulate PM differently, and the most effective plant species should be selected in urban areas for attenuating ambient PM.

Key words Particulate matter; Urban trees; Leaf deposition; Wax; Water-soluble ions.

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Introduction

Airborne particulate matter (PM), consisting of particles with aerodynamic diameters in the range of 0.001-100 μm, is a major atmospheric pollutant (Pope III and Dockery 2006; WHO 2006). Anthropogenic sources contributing to PM include vehicle exhausts, road dust, domestic and large-scale coal burning, and cement and other industrial processes (Bosco et al. 2005; Dzierzanowski et al. 2011). Due to rapid urbanization and industrialization, the Beijing-Tianjin-Hebei region is undergoing serious PM pollution. For example, the annual average $PM_{2.5}$ (PM with diameter <2.5 μm) concentration in Beijing was 86 μg m^{-3} in 2014 (Chen et al. 2015), which was about 2.5 times higher than the WHO Air Quality Guideline value of 35 μg m^{-3} (WHO 2006). Positive associations have been found between $PM_{2.5}$ and health impacts (e.g. lower respiratory infections, trachea bronchus, lung cancers and cerebrovascular disease) (Lelieveld et al. 2015; Liu et al. 2016). It has been estimated that $PM_{2.5}$ pollution in urban areas of China led to 763,595 premature deaths in 2013 (Song et al. 2016). Beijing was worst affected with estimates of 5.2, 9.0, 2.3, and 1.6 thousand premature mortalities from ischemic heart disease (IHD), cerebrovascular disease, chronic obstructive pulmonary disease (COPD) and lung cancer (LC), respectively (Liu et al. 2016).

Numerous studies have identified that urban greening filters accumulate atmospheric particles more effectively than other land surfaces (Chen et al. 2016a; Janhäll 2015; Tallis et al. 2011;

Thithanhthao et al. 2015). Enhanced the deposition flux to the surface is the benefit for reducing pollutant concentrations near the ground and thus exposure of people in urban areas. This is one of the recognized ecosystem services of urban vegetation (Salmond et al. 2016). Compared with other urban surfaces, vegetation enhances deposition of particles because of the finely divided structure of many leaves, especially conifers. They have a larger collecting surface per unit ground area, and reduce the laminar boundary layer that limits the particles uptake. Using the i-tree model, Nowak et al. (2013) modeled $PM_{2.5}$ removal by trees in ten American cities and estimated that annual masses of particles removal ranged from 4.7 t in Syracuse to 64.5 t in Atlanta. Schaubroeck et al. (2014) used the canopy interception and PM removal multilayered model (CIPAM) to estimate PM accumulated on a forestry canopy. For a case study in 2010 they estimated that a Scots pine stand in Belgium accumulated about 31 kg $PM_{2.5}$ ha^{-1} yr^{-1}. The plain afforestation project in Beijing has been estimated to decrease annual $PM_{2.5}$ concentrations by 0.57 μg m^{-3}, or 2% of the target set by the “Beijing Clean Air Action Plan (2013-2017)” (Chen et al. 2014).

At a leaf scale, particles deposited on leaf surface and ultrafine PM (particles with diameter < 100 nm) may enter the leaf stomata. Previous studies have examined the effectiveness of urban trees around the world for accumulation of $PM_{2.5}$ and PM_{10} (Chen et al. 2016b; Sgrigna et al. 2015) and demonstrated that PM accumulated on leaves differed significantly among plant species (Beckett et al. 2000; Przybysz et al. 2014). Conifer species have greater potential in capturing PM than broad-leaved species and evergreen conifers accumulate PM throughout the year (Sæbø et al. 2012). However, broad-leaved species with large amounts of pubescence and rougher surfaces also accumulate greater masses of particles (Mo et al. 2015). Particles mainly deposit on the adaxial leaf surfaces, and accumulated masses about six times higher than on abaxial leaf surfaces (Wang et al. 2006). The importance of leaf microstructure characteristics on the effectiveness of capturing PM was evidenced by using scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) analysis (Yan et al. 2016).

Given limited greening space in urban Beijing, the most effective plant species in removing PM should be selected for urban greening. Although many studies have focused on quantifying the amount of PM deposited on trees (Yin et al. 2011; Chen et al. 2015; Wang et al. 2015; Leonard et al. 2016), there is still a knowledge gap in terms of accumulation of PM in epicuticular waxes between different species. Xu et al. (2017) used an artificial rainfall simulation system to investigate the influence of rainfall duration and intensity on PM removal from four broad-leaved species, and they detected that final wash off rates in the rainfall events was only 51-70% of initial deposition. In other words, most PM captured by leaves is stored temporarily, since deposited particles on leaf surfaces may be subsequently resuspended from the leaves to the air by wind and rain (Schaubroeck et al. 2014). However, PM accumulation in the wax fraction is very important because these particles are trapped permanently (Song et al. 2015). Most studies collect leaves only once at the beginning or in the middle of the season (Binze et al. 2014; Cheng et al. 2016; Mo et al. 2015). However, the characteristics of leaf structure are different during the growing season, which may impact their ability to accumulate PM. The leaf-washing methodology is the most common experimental approach. A large proportion of PM is water-soluble ions, which comprised approximately 40% of $PM_{2.5}$ in Beijing (Han et al. 2016). However, the rinse and weigh method may underestimate the mass of particles on leaves. Therefore, water solutions ions need to be quantified in order to accurately assess the effectiveness of plant species in capturing PM. Analyzing water-soluble ions in deposited PM on leaf surface allows to identify pollution source of PM. The aim of this study was to investigate the effectiveness of 17 plant species in accumulating PM during the growth season in Beijing for the year 2014. Two key features of this study were investigation of: 1) particles deposited onto the leaf surface and into the wax as a function of three particles size fractions; and (2) the accumulation of water-soluble ions by the leaf surface.

Material and methods

Study Area

All of the plant materials were collected from the campus of Beijing Forestry University (BFU) (40°0'29.64"N, 116°20'46.04"E), Beijing City, China. The BFU campus is located in Haidian

District outside the Fourth Ring Road, which is the transition from urban to suburban areas. There are no polluting industries or power plants within 5 km space range. All plant species were located within 600 m each other, in the center of the BFU campus and 50 m away from surrounding streets. It is a reasonable assumption that all species were exposed to the same background PM concentrations. The average background PM_{10} and $PM_{2.5}$ concentrations were 139 and 84 μg m^{-3} from April to September 2014, respectively, in Olympic Sport Center which was the nearest air quality monitoring station (about 4.4 km apart) set by the Ministry of Environmental Protection of China (http://113.108.142.147:20035/emcpublish/).

Plant material and sample collection

Vegetation samples were collected from 17 plant species (listed in Table 1). All are common plants in northern China and widely used in urban areas. For each species three similar individuals were used as replicates. Leaves were collected on 16 April, 20 May, 30 June, 27 July, 11 September and 14 October 2014. Criteria for the weather on a sampling day were sunny, wind speed less than 5 m s^{-1}, and 9-10 days since precipitation amount of the last rain event >5 mm. Leaves were assumed to be washed clean by rainfall, so the average mass of daily deposited PM was measured for the same exposure days. The study of Liu et al. (2013) investigated the dust-retaining capacity of the four urban species in Guangzhou, and their results showed that the amounts of PM deposited on leaf surface may approach saturation around 24 days. In the present study, the average mass of daily deposited PM was measured for the same exposure days without saturation, and leaves were assumed to be fully washed by rainfall. Intact leaves were in good growth conditions, namely, with little or no disease and/or pests. They were sampled from four directions in the canopy with the same height for each species. Between 200 and 300 cm^2 of leaf area (about 5 - 30 leaves) were cut off with scissors and stored in plastic bags. Great care was taken to ensure that particles did not dislodge from the leaves. The samples were then transported to the laboratory and stored at 4°C freezer prior to analysis. Although the detailed collection method of leaves samples has described in our previous studies (Chen et al. 2016b; Mo et al. 2015), it is repeated here for the reader's convenience.

Quantitative analysis

PM was washed and quantified from the leaf surfaces and in leaf waxes using the methods described by Dzierzanowski et al. (2011). The methods for rinsing water-soluble ions from the leaves were as described by Freer-Smith et al. (2005). Leaves were first washed with 250 mL of deionized water. The rinse water was passed through a 100 μm metal sieve to remove particles >100 μm and then filtered sequentially through 10 μm, 2.5 μm and 0.2 μm filters (EMD Millipore Corp., Billerica, Massachusetts, USA) to separate the PM into the following three particles size fractions: fine (0.2–2.5 μm), coarse (2.5–10 μm), and large (> 10 μm). The aqueous sample after filtering was stored in the freezer (−18°C) prior to determination of the water-soluble ions (see below). Before filtering, a deionizer gate (AP-BC2451, AP&T, Shanghai, China) was be use to avoid an electrostatic charge on the filters. Before and after analysis, filters were dried in an oven for 30 minutes at 50ºC and stabilized in an 80 cm × 80 cm × 80 cm polytetrafluoroethylene (PTFE) balance box at 25ºC and 40% relative humidity for 24 hours, which was controlled by a balance and a humidity controller (WHD48-11, ACREL Co., Ltd., Jiangsu, China). Filters were weighed using a BT125D balance (Sartorius, precision: 10 μg). PM accumulated in waxes was also measured similarly except that the deionized water was replaced with chloroform in order to dissolve the wax on the leaves. Also since nitrocellulose filters are damaged by chloroform, PTFE filters were used in the filtering for in-wax PM.

Each washed broad-leaved sample was scanned using a scanner (HP Scanjet 4850, China Hewlett-Packard Co., Ltd., Beijing, China), then the area was measured by Image J (1.50i, National Institutes of Health, Bethesda, US). For needle leaves, leaf area was measured by measuring the water displacement leaf volume and converting to leaf area using the following equation:

$$S = 2L(1+\frac{\pi}{\mathrm{n}})\sqrt{\frac{\mathrm{n}V}{\pi L}}$$

where S represents leaf area; L is the average length of the leaves; V is the needle volume, and n is the number of needle leaves.

To analyze the concentrations of water-soluble inorganic ions, the volumes of post-filtering aqueous extracts were measured (V_{water}) and stored at -18 °C until analysis within four weeks. Three anions (Cl^-, SO_4^{2-}, NO_3^-) and five cations (NH_4^+, Ca^{2+}, $Na^{+,}$ Mg^{2+}, K^+), were determined using a Dionex model ICS-120 ion chromatograph equipped with a conductivity detector (ASRS-ULTRA) following the method of Lun et al. (2003) and Tang et al. (2016). The mass of these ions obtained in each filtrate was calculated by multiplying V_{water} by the ion concentration, and was expressed in per unit area per leaf.

Statistical analysis

One-way analysis of variance (ANOVA) was used to test for significant differences in PM accumulation between species. Post-Hoc analysis (Duncan's test) was performed when multiple comparisons among species were necessary. Spearman's correlation analysis was used to assess the linear correlations between surface PM, in-wax PM and water-soluble ions. All statistical analyses were carried out using SPSS 17.0 (SPSS 17.0 for Windows, SPSS Inc., IL, USA).

Results and discussion

PM accumulation on leaves

Fig. 1 shows the mass of accumulated surface and in-wax PM on the 17 plant species. The PM accumulation on leaves varied significantly between plant species. Several studies have also identified that different height of trees may affect the capacity for accumulating PM

(Dzierżanowski et al. 2011; Sæbø et al. 2012; Mo et al. 2015). In this study, we divided the species into shrubs and trees (Table 1). The average PM accumulation for four shrubs was 49 µg cm^{-2}. *E. japonicus* showed the highest PM accumulation (56 µg cm^{-2}) of the four shrubs. Although the difference was not significant. Xie et al. (2014) and Wang et al. (2006) reported that E. *japonicas* captured larger amounts of PM than other shrubs such as *Buxus sinica*, *Syringa oblata* and *Lagerstroemia indica.* The leaf surface structure of *E. japonicus* is unique to those shrubs. Tomentose pubescence distributes on the abaxial leaf surface, which can efficiently capture and accumulate PM (Mo et al. 2015).

PM accumulation on *P. orientalis* and *P. armandi* were significantly higher than for other trees. Accumulations on *F. pennsylvanica, P. tomentosa, A. altissima* and *S. japonica* were significantly lower than for other trees (Fig. 1). The average PM accumulation on *P. orientalis* and *P. armandi* (156 µg cm^{-2}) was 4.5 times greater than the average accumulation on the latter four species (28.4 µg cm^{-2}). The PM accumulation on *R. typhina* (76 µg cm^{-2}) was second only to *P. orientalis* and *P. armandi.* Trees can be divided into conifer or broad-leaved species. *P. tabulaeformis, P. orientalis* and *P. armandi* are three common evergreen conifer species, which accumulated more PM on leaves than other species except *R. typhina*. The PM accumulation difference between the adaxial and abaxial broad-leaved surfaces is due to wind turbulence. Wang et al (2006) investigated 11 broad-leaved species such as *P. tomentosa*, *S. Japonica* and

Ailanthus altissima using a scanning electron microscope (SEM) in Beijing urban area. They found that on average only 17% of PM accumulated on the abaxial surface. However, the needles of conifer species can accumulate PM over the entire leaf surface (Ottelé et al. 2010). The leaves of conifer species also have unique microstructure, such as mucus oils, a thicker epicuticular wax layer and a grooved ridge protuberance, which can help leaves to accumulate large particles (Sabin et al. 2006). Previous studies showed that the leaves of *Platycladus* were more rough than *Pinus* (Wang et al. 2007). This structure retained more particles on leaves of *P. orientalis.* The average PM accumulation of broad-leaved trees was less than broad-leaved shrubs. The meta-analysis results showed that PM leaf deposition on shrubs was significantly higher than that of trees (Cai et al. 2017).

Across all species, PM was found both on leaf surface and in waxes. Fig. 1 shows that particles distribution between surface and waxes is similar for all shrub species, with PM mass in waxes about 4 times lower than on surface. The average surface and in-wax PM deposition on the shrub leaves were 39 and 10 μg cm^{-2}. This corresponds to 21% of PM deposition in waxes, on average. There were significant differences between tree species in the accumulation of PM on leaf surfaces and in waxes. The lowest and highest surface PM deposition were found in *P. tomentosa* (12.5 μg cm^{-2}) and *P. armandi* (56.9 μg cm^{-2}). The in-wax PM accumulation of *P. orientalis* (101.9 μg cm^{-2}) and *P. armandi* (96.6 μg cm^{-2}) was significantly higher than other tree species. A positive relationship between different PM fractions accumulation and the quantity of leaf waxes was detected (Sæbø et al. 2012). Popek et al (2013) analyzed all tested species and also detected a positive correlation between the amount of waxes and course PM. But the model had a low partial fit (r=0.54). By analyzing for each species separately, a significant correlation was found in leaves of *Tilia cordata* (Dzieranowski et al. 2011) and *Corylus colurna* (Popek et al. 2013). In some cases, only weak, no relationship or negative correlation between mass of in-wax PM and mass of wax were reported across some plant species (Jouraeva et al. 2002; Dzieranowski et al. 2011). The relationship showed different results among species. Song (2015) found that the waxes in conifer species was a factor of about 2.5 times higher than broadleaves. *P. orientalis* and *P. armandi*, of which the waxes can accumulate large fraction of the PM. The composition and structure of the waxes may significantly affect the capacity of leaves in accumulating PM (Kaupp et al. 2000; Jouraeva et al. 200; Bukhardt et al. 2010). On the other hand, *P. tabulaeformis, P. orientalis* and *P. armandi* which accumulated greater in-wax PM than other plant species are evergreen conifer species. The leaf growth cycle of these evergreen conifer species of more than 12 mouths is considerably longer than for deciduous species. Song et al (2015) investigated the mass of PM deposited on five evergreen species in Beijing. The PM accumulated on the leaf surface were range from 72.31 to 231.84 μg cm^{-2}. Cai et al. (2017) reviewed 150 studies and used a meta-analysis and also found that the weekly PM leaf deposition of conifer species was significantly higher than broad leaves, by approximately 31.9%. These results showed that evergreen conifer species had better performance in accumulating PM both on the leaf surface and in the surface wax, which was accounted for by the structural characteristics and habits of conifer species.

In our study we found significant positive correlation between in-wax PM accumulation and surface PM accumulation ($R^2 = 0.43$, $p = 0.002$). Based on collection and analysis of existing raw data of 56 plant species in 3 different urban areas (Popek et al. 2013; A. Przybysz et al. 2014; Mo et al. 2015; and Song et al. 2015), a positive correlation relationship between total amount of surface PM and in-wax PM accumulated on foliage was also detected (R^2=0.64, P<0.0001, In-wax PM=0.24×Surface PM+2.35). The leaf surface PM deposition amounts was 3.57 times higher that of in-wax. Actually, PM in the wax layer can account for a significant amount, about 22% in the present study. The epicuticular wax layer and releasing PM were dissolved by chloroform, which has environmental health concern. In the previous studies, PM accumulated on leaf surface was assessed (Freer-Smith et al. 2005; Chen et al. 2015). Lack of the examination of PM encapsulated in the wax layer may lead to an underestimation of the present results. However, a significant correlation was found between surface and in-wax PM, which can improve our knowledge on this issue. Because leaves with greater amounts of surface PM may also accumulate high mass of PM in wax layer, and these species should be attractive options for urban greening.

The average mass of surface PM accumulation in trees (34 μg cm^{-2}) was lower than that in shrubs (39 μg cm^{-2}). But trees have the higher average of in-wax PM accumulation (28 μg cm^{-2}) relative to shrubs (10 μg cm^{-2}) but not statistical significant. Contributions of in-wax depositions to leaf total (surface plus in-wax) depositions were > 50% for *P. tomentosa* (54%), *P. orientalis* (65%) and *P. armandi* (63%). In the remaining 10 tree species, the contributions of in-wax to total insoluble PM were on average 32%. In Norway and Poland, *Fagus silvatica* and *Stephanandra incisa* also accumulated about 25% and 28% of PM in the waxes and on the surface, respectively. *Betula pendula* accumulated 82.6% of PM in the wax fraction (Sæbø et al. 2012), which is significantly higher than the value of coniferous species obtained in this study. Popek et al. (2013) reported that more PM was deposited on the leaf surface. In their study, the waxes in 5 of 39 woody species accumulated greater than 50% of insoluble PM (range 53%-63%), consistent with our findings.

This study (Fig. 6 and 7) showed that PM leaf accumulation varied greatly among months, which was different between leaf surface and waxes. Leaf surface PM retention amounts in trees and shrubs were significantly higher in April and May, respectively, compared with those in other months. The lowest amounts were measured in September. Masses of PM accumulated on surfaces of trees and shrubs showed inconsistent relations among different months. Atmospheric PM concentration, meteorological condition and leaf characteristics varied among months, which may influence PM leaf deposition. Basis on analysis of our dataset, monthly ambient average concentrations of PM_{10} and $PM_{2.5}$ did not correlate with deposited amounts of total surface particle. However, with increasing atmospheric PM concentrations, particles accumulated on leaf surface raised slightly. By quantitatively analyzing in four districts of the city in Italy, leaf surface PM deposition didn't correlate with local atmospheric PM_{10} concentration (G. Sgrigna et al. 2015). On the other hand, a meta-analysis result showed that PM accumulation is generally highest in winter compared to other seasons and thought this variable is affected by atmospheric PM concentration (Cai et al. 2017). In this study, monthly data was converted to seasons and showed highest total PM deposited was in spring for trees and shrubs. Leaf PM depositions were all most equally between summer and autumn. But background PM concentrations in summer were lower than spring and autumn. On the other hand, monthly change of PM accumulation in waxes is increased firstly and then decreased during growing season, which in trees and shrubs were highest in July and September, respectively. Vegetable characteristics and ambient PM concentrations show the interaction in leaf PM deposition. In winter, most studies focused on evergreen species such as coniferous plants, which show high capacity to capture particles. So, the conclusion that PM leaf deposition is highest in winter is one-sided. Across all growing months, trees show higher PM accumulation in wax than shrubs. The PM accumulation on leaves varied significantly among months. Leaves only once at the begin or the middle of the seasons to assess the capacity of PM capturing may underestimate or overestimate it.

Overall, results suggested that leaf surface accumulated more PM than waxes. Although for most species there was lower mass of PM deposited in epicuticular waxes. However, PM accumulation in the wax fraction is very important because these particles are trapped permanently (Song et al. 2015).

Different Size-fraction Particles

Different size fractions of airborne PM have different chemical composition and posed adverse effects on human health (Luo et al. 2011; Li et al. 2017). For example, particulate matters were responsible for respiratory, cardiovascular and others (Cai et al. 2017), especially fine particles (Englert et al. 2004). To assess the capacity of plant leaves to capture different size fractions, the present study separated quantification of PM into: fine (0.2–2.5 μm), coarse (2.5–10 μm), and large (> 10 μm). The masses in each of these size fractions accumulated on leaf surface and in waxes are presented in Fig. 2 and 3, respectively.

PM of different size fractions accumulated on leaf surface and in waxes differed among the species. Non-significant differences were observed among the different size-fraction surface PM accumulations on shrubs (Fig. 2). The averages of the three size fractions for surface PM accumulations on shrubs were 31.5 µg cm^{-2} (81%), 4.7 µg cm^{-2} (12%) and 2.5 µg cm^{-2} (7%) for large, coarse and fine PM, respectively.

Across all trees, *P. orientalis* (44.4 µg cm^{-2}) and *P. armandi* (43.7 µg cm^{-2}) showed the greatest large particles fraction accumulation on the leaf surface. Large particles fraction accumulation on *P. tomentosa* (9.8 µg cm^{-2}), *A. altissima* (15.4 µg cm^{-2}) and *S. japonica* (12.7 µg cm^{-2}) leaves were significantly lower than the greatest accumulators. The average surface accumulation of large particles fraction was 25.0 µg cm^{-2}, which accounted for 75% of the total insoluble PM.

The average coarse fraction PM accumulated on leaf surfaces (5.1 µg cm^{-2}) comprised 15% of the total insoluble PM. The greatest accumulation of coarse PM was found on *P. armandi* leaves (8.9 µg cm^{-2}), which was weakly significant difference from accumulation by other trees. The species with the lowest surface accumulation of the coarse fraction was on P. *tomentosa* (1.8 µg cm^{-2}).

The greatest accumulation of fine fraction PM on the leaf surface was on *R. typhina* (9.30 µg cm^{-2}), which was significantly greater than for other trees. *P. occidentalis* (2.0 µg cm^{-2}), *F. pennsylvanica* (1.2 µg cm^{-2}), *P. tomentosa* (1.0 µg cm^{-2}), *G. biloba* (2.1 µg cm^{-2}) and *S. japonica* (1.6µg cm^{-2}) had considerably lower accumulation of fine fraction PM. The average fine PM fraction surface accumulation was 3.5 µg cm^{-2}, or 10% of the total insoluble PM deposition.

These present results showed that shrubs accumulated more mass of large size fraction PM on leaf surface than most of the trees, except that *P. orientalis* and *P. amandi* which showed slightly higher accumulation than that of shrubs. However, trees were more efficient than shrubs in accumulating coarse and fine PM on the leaf surface.

There were no significant differences between the three particles size fraction accumulations in the wax of shrub leaves. On average, the PM accumulation in wax on the shrub leaves was 6.8 µg cm^{-2} (65%) for the large particles size fraction, 2.3 µg cm^{-2} (21%) for the coarse size fraction and 1.5 µg cm^{-2} (14%) for the fine particles size fractions. For the trees, *P. orientalis* (84.1 µg cm^{-2}) and *P. armandi* (85.3 µg cm^{-2}) showed significantly greater accumulation of the large particles size fraction in wax. The average accumulation of the large particles size fraction in wax on trees was 22.0 µg cm^{-2}, which was 77% of the total accumulated PM. The highest coarse particles accumulation in wax was for *P. orientalis* (10.9 µg cm^{-2}). The coarse particles in-wax deposition on *P. armandi* (6.5 µg cm^{-2}) and *S. matsudana* (6.6 µg cm^{-2}) were marginally higher than for the remaining trees. *F. pennsylvanica* (1.1 µg cm^{-2}) and *A. altissima* (0.9 µg cm^{-2}) had low deposition of fine particles in waxes. The average coarse and fine particles size fraction accumulations in wax were 3.8 µg cm^{-2} and 2.5 µg cm^{-2}, which corresponded to 13% and 9% of the total in-wax particles deposition. Particle size fraction is very similar for surface and in wax PM accumulated by tress, but differs for shrubs showing amount of large PM on surface about 5 more than in wax, which leaded to less fine and coarse PM fractions on surface. Dzierżanowski et al (2011) detected a small shrub accumulated the largest amounts of PM as compared to trees, and more large PM also deposited on surface than wax. As we explained above, shrubs growing low to the ground were presumably more exposed to soil splash and traffic dust on the leaves than trees with an upright growth habit, which were mostly large particles. However, these large PM deposited on surface cannot be fixed by epicuticular wax in shrubs, which was significantly different with trees.

Overall, PM depositions by mass for the three size fraction was in the order: large > coarse > fine. The average proportions of large, coarse and fine PM were 73%, 16% and 11%. Previous studies reported slightly different proportions of these three size fractions (Popek et al. 2013; Song et al. 2015; Zhang et al. 2014), which may be due to the local ambient PM composition. The proportions of the three size fractions were slightly different between species. This might be explained by the leaf and structural characteristics of species (Song et al. 2015). However, as we introduced that the average background PM_{10} and $PM_{2.5}$ concentrations in this area were 139 and 84 μg m^{-3}, respectively. The concentration of fine PM was higher than that of coarse particles, which was different with fine and coarse PM deposited in leaf surface and waxes. Freer-smith et al. (2005) found that sedimentation under gravity principally leads to large PM deposited on leaf surfaces. Impaction and interception affects the deposition of coarse and fine PM on leaf surfaces.

Therefore, particles sedimentation is the main process by which PM deposits to foliage. The mass of fine PM deposited by impaction and interception was 5.5 μg cm^{-2} on average, which contributed only 11% of the total. However, the number of fine particles on leaves was large. Previous studies proposed that the number of particles in the fine fraction contributed over 90% of total insoluble PM (Li et al. 2015; Zhao et al. 2014). Therefore, plant also showed high efficiently accumulation of fine PM in urban areas. The amount of coarse and fine PM contributed 18% and 12% of total in-wax PM respectively. This ratio was higher than for deposition to leaf surfaces. Shrub leaf surfaces accumulated greater mass of the three size fractions PM than most of trees. Shrub and tree leaves grow at different heights, which may have an effect on PM accumulations on leaf surface because there is more dust near the ground (Dzierżanowski et al. 2011; A. Sæbø et al. 2012; Mo et al. 2015). The significant correlation between the mass of PM size fraction was detected (R^2 for large, coarse and fine PM are 0.99 ($p < 0.0001$), 0.78 ($p < 0.0001$) and 0.52 ($p = 0.001$), respectively). So, species that accumulated more total particles also have the high capability to capture greater amounts of fine particles, which is the most dangerous for human health. The most efficient species of fine PM accumulation should be used for urban greening, such as *R. typhina*, *P. orientalis*, *P. armandi*, and *M. denudate.* Tree-dimensional configuration of the most efficient shrub and tree species can reduce PM pollution on different spatial scales. We also suggest that conifer species should be priority in urban greening, which show high capability in capturing PM during winter.

Water-soluble ions on leaf surfaces

The foliage analyzed for water-soluble ions was that sampled on July 27, 2014. Fig. 4 shows the results for ions and total insoluble suspended particulate (TSP). The ratio in Fig. 4 represents the percentage of water-soluble ions and the total of soluble and insoluble PM. Across all species, the mass of ions on leaf surfaces ranged from 3.7 µg cm^{-2} (in *M. denudata*) to 31.6 µg cm^{-2} (in *P. armandi*). There were significant differences between plant species, *U. pumila*, *S. matsudana*, *P. orientalis* and *P. armandi* accumulating more ions on leaf surfaces ($p < 0.05$). For other species, accumulations for all ions were low to 10 µg cm^{-2}. The mean value for ions deposition was 12.9 µg cm^{-2}. Significant positive correlations were observed between the amounts of ions and TSP across species (R^2=0.48, p=0.001). The highest and lowest percentages of water-soluble ions to total PM on leaves were 50% and 7% respectively, with an average of 28%. The proportions of water-soluble ions in total PM varied greatly among different plant species, especially for trees. Variations of proportion between species were also detected in the study of Freer-Smith et al. (2005). The data show that the contributions of ions to total PM was in the order $NO_3^- > Ca^{2+} > SO_4^{2-} > Mg^{2+} > Cl^- > Na^+ > K^+ > NH_4^+$. The proportion of NH_4^+ in the ions was lowest across all species, which is surprising considering the ambient levels of NH_4^+ in Beijing. However, the findings are consistent with Freer-Smith et al. (2005) who also showed that NO_3^- was the main ion component and that the proportion of NH_4^+ was low. Cheng et al. (2016) reported average concentrations of SO_4^{2-}, Cl^- and NO_3^- on plant leaves of 0.9098 μg cm^{-2}, 0.7298 μg cm^{-2} and 0.0878 μg cm^{-2}, respectively, which are slightly different from this study. The difference may be accounted for by the composition of inorganic compounds in the air. Shen et al. (2011) showed ions comprised 33% PM_{10} at the Dongbeiwang experiment site, near to this study site, which was relatively high compared with this study. The contribution of NH_4^+ to ions was 16%. Liu et al. (2015) used ^{15}N tracer techniques to show that water-soluble ions (NH_4^+ and NO_3^-) in $PM_{2.5}$ can be absorbed effectively by *P. euramericana* seedlings. Given that plant leaves absorb ultrafine PM through their stomata (Bell and Treshow 2002), we postulate that NH_4^+ is translocated into stomata which leads to the low contribution of NH_4^+ to ions on the leaf surface. The concentration of NO_3^- was in the range of previous studies. In summary this study has shown that water-soluble ions on leaf surfaces are also an important component of total PM deposited onto plant leaves. As many past studies did not analyze the soluble PM components this means that the interception capacity of plant species for PM has previously been undervalued. According to studies on water soluble ions in atmosphere particles, without considering the mass of soluble ions was slightly undervalued the effects of plants on PM deposition and may be a deviation in the estimation of the PM accumulated by the whole trees (Song et al. 2015). In this study, the percentages of water-soluble ions to total PM on coniferous leaves ranged from 24% to 29%.

Based on analysis of their data (Song et al. 2015), undervalued mass of water ions on conifer species was about 38.59 μg cm^{-2} on average. This may have more serious impatient on the whole plant assessment. The elemental composition of the ions helps the identification the source of the particles, which provides further important new information. In previous studies, the water-soluble ions in PM in Beijing were classified into 4 sources (Sun et al. 2004; Wang et al. 2005): NO_3^-, SO_4^{2-} and NH_4^+ were assigned to type 1, which comes from secondary inorganic aerosol; Ca^{2+} and Mg^{2+} were to type 2, which comes from road and construction dusts; Na^+ and Cl^- is assigned to type 3, which mainly come from waste incineration; and K^+ is assigned to type 4, which mainly comes from biomass combustion. Figure 5 shows the average concentrations of ions in each type. The greatest contribution to was from type 1 secondary aerosol; NO_2 and SO_2 from coal combustion and vehicle exhausts are converted to NO_3^- and SO_4^{2-} by gas-to-particles reactions, which are mainly of anthropogenic origin. The contribution of Ca^{2+} and Mg^{2+} to total PM were the next greatest and was significantly higher than contribution from type 3 and 4. Type 3 and 4 were low in our study site, which indicated the burning of waste and biomass. The important feature of the data reported is that traffic was the main source of pollution for the background air at this study site.

Conclusion

Seventeen urban plant species showed significantly different capacity in accumulating PM. The average accumulation of PM on leaves was 58.9 μg cm^{-2}, of which 65% was deposited on the leaf surface and 35% in the leaf wax. The greatest PM accumulations on leaves occurred in *P. orientalis* and *P. armandi*, both of which are evergreen conifer species. *F. pennsylvanica*, *P. tomentosa*, *A. altissima* and *S. japonica* was less effective for PM deposition. *P. tomentosa*, *P. orientalis* and *P. armandi* were the only three of seventeen species, which accumulated greater than 50% of total insoluble PM in waxes. PM larger than 10 μm comprised 73% of PM deposited on leaves, with the coarse and fine particles size fractions comprising 16% and 11% of the deposited PM, respectively. Water-soluble ions comprised 28% of total PM on leaves on average. Lack of knowledge about ions captured by leaves obviously leads to underestimation of the ability of plant species to intercept PM. This study has shown that trees and shrubs should be considered as an effective approach to remove aerial PM in urban areas.

Acknowledgments

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Table 1. Species and allometric data for the trees sampled. (mean ±SD, n = 3)

Classification	Species	Height (m)	Diameter at breast height (cm)	Crown diameter (m)
Shrubs	*Amygdalus triloba* (Lindl.) Ricker	2.1±0.4	7.7±1.4	2.8±0.2
	Euonymus japonicus Thunb.	1.9±0.2	5.0±1.1	2.4±0.2
	Lonicera maackii (Rupr.) Maxim.	3.2±0.3	10.8±3.4	5.6±0.4
	Prunus Cerasifera Ehrh	2.4±0.3	23.0±2.4	5.4±1.2
Trees	*Magnolia denudata* Desr.	10.5±0.4	23.6±3	6.8±1.7
	Rhus typhina Nutt.	3.6±0.2	8.8±1.3	2.3±1.1
	Platanus occidentalis L.	14.4±0.6	53.7±6.2	6.6±2.1
	Fraxinus pennsylvanica Marsh.	9.4±0.6	25.0±2.1	7.9±2.6
	Populus tomentosa Carr.	13.4±0.7	51.9±3.6	5.3±0.9
	Ginkgo biloba L.	14.5±0.5	53.5±5.6	5.8±1.4
	Ulmus pumila L.	8.4±0.3	77.1±7.4	6.8±2.3
	Salix matsudana Koidz.	12.6±0.4	34.4±5.8	6.9±2.3
	Pinus tabulaeformis Carr.	10.0±0.8	29.4±5.3	5.6±0.8
	Platycladus orientalis (L.) Franco	6.1±0.1	16.1±2.3	1.7±0.6
	Pinus armandi Franch.	5.4±0.6	16.5±1.6	3.8±0.8
	Ailanthus altissima (Mill.) Swingle	9.8±0.5	27.8±3.5	4.6±0.7
	Sophora japonica L.	10.7±0.3	24.5±0.3	3.4±0.5

Figure Captions

Fig. 1. Total amount of PM accumulated on leaf surfaces and in-wax. Data are mean ±SE, n = 18. Bars marked with different letters are significantly different ($p < 0.05$).

Fig. 2. The sum of PM accumulation of three size fractions on leaf surfaces. Data are mean ±SE, n=18. Bars marked with different letters are significantly different ($p < 0.05$).

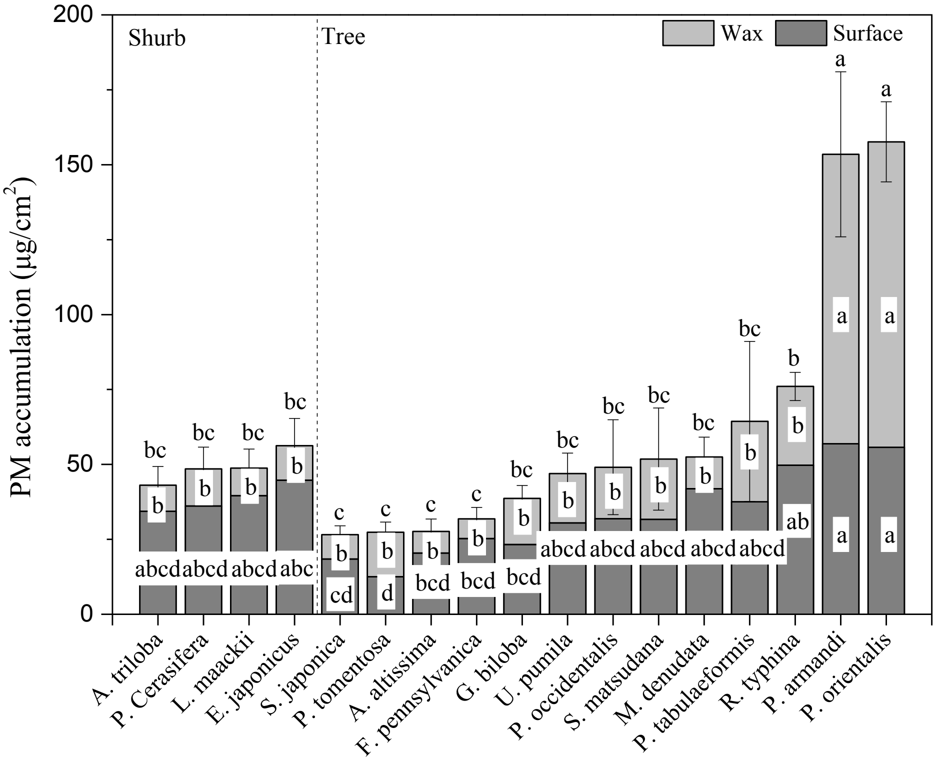
Fig. 3. The sum of PM accumulation of three size fractions in waxes. Data are mean ±SE, n=18. Bars marked with different letters are significantly different ($p < 0.05$).

Fig. 4. Total insoluble suspended particulate and water-soluble ions accumulated on leaf surfaces. The ratio of ions to TSP. Data are mean ±SE, n =3.

Fig. 5. The concentrations of dissolvable inorganic ions presented as the sum mass of four types ions. Data are mean ±SE, n=3.

Fig. 6. Monthly change of PM accumulation on leaf surface between shrubs and trees. Data are mean ±SE, n=3. Bars marked with different letters are significantly different ($p < 0.05$).

Fig. 7. Monthly change of PM accumulation in waxes between shrubs and trees. Data are mean ±SE, n=3. Bars marked with different letters are significantly different ($p < 0.05$).



PM accumulation (μg/cm²)
0
50
100
150
200
Shurb
Tree
Wax
Surface
bc
b
abcd
c
cd
d
bcd
abc
ab
a
A. triloba
P. Cerasifera
L. maackii
E. japonicus
S. japonica
P. tomentosa
A. altissima
F. pennsylvanica
G. biloba
U. pumila
P. occidentalis
S. matsudana
M. denudata
P. tabulaeformis
R. typhina
P. armandi
P. orientalis

Fig. 1.

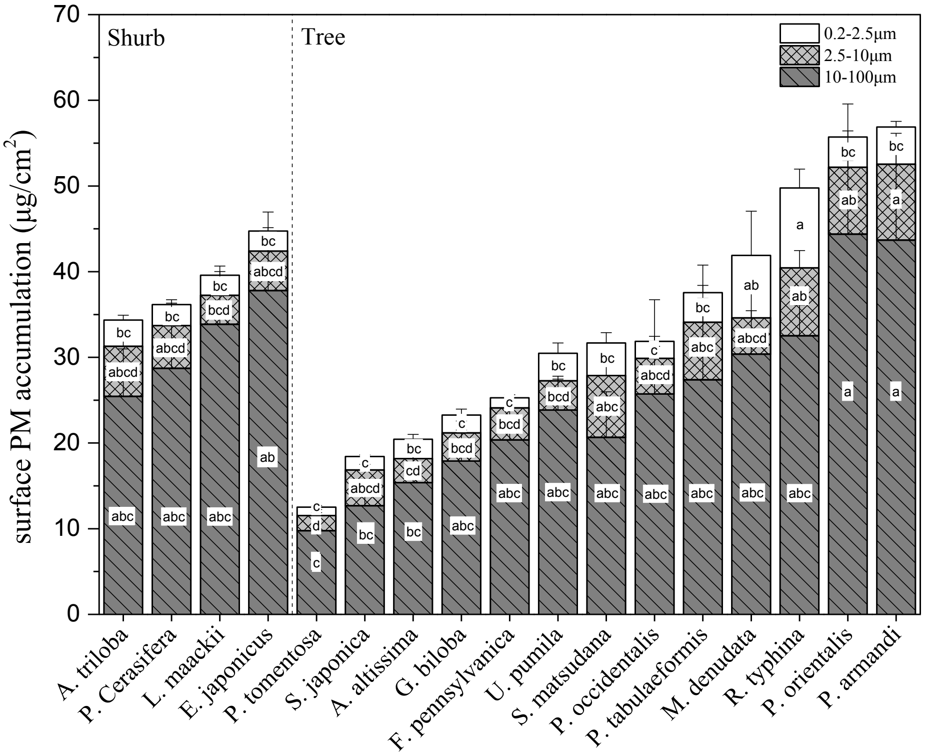
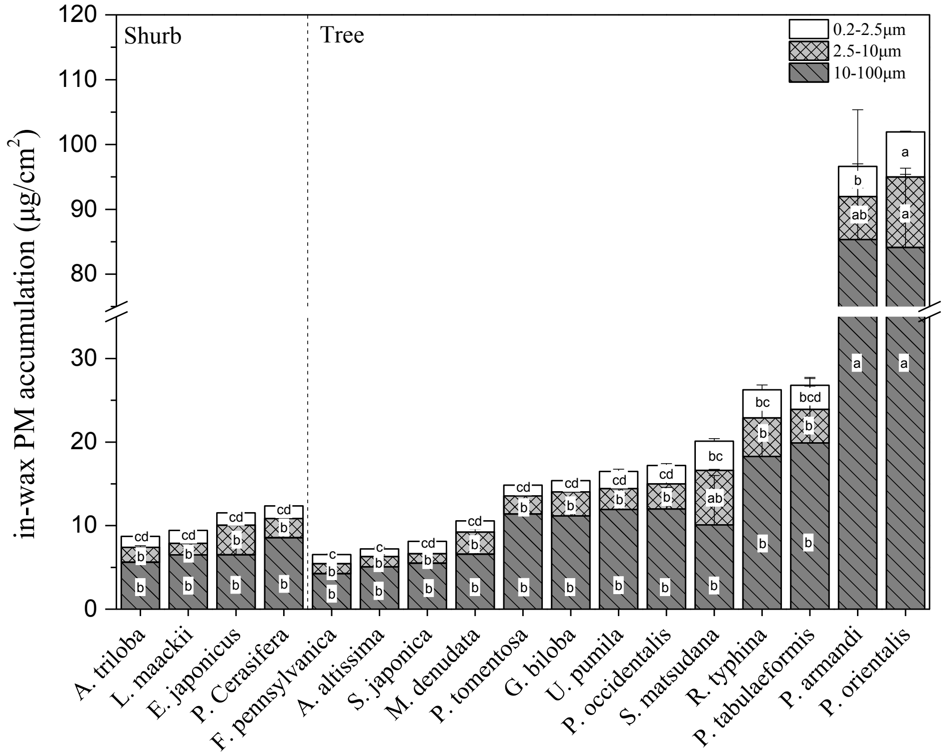

surface PM accumulation (μg/cm^2)
0
10
20
30
40
50
60
70
Shurb
Tree
0.2-2.5μm
2.5-10μm
10-100μm
A. triloba
P. Cerasifera
L. maackii
E. japonicus
P. tomentosa
S. japonica
A. altissima
G. biloba
F. pennsylvanica
U. pumila
S. matsudana
P. occidentalis
P. tabulaeformis
M. denudata
R. typhina
P. orientalis
P. armandi

Fig. 2.



0.2-2.5μm
2.5-10μm
10-100μm
Shurb
Tree
in-wax PM accumulation (μg/cm^2)
0
10
20
30
80
90
100
110
120
A. triloba
L. maackii
E. japonicus
P. Cerasifera
F. pennsylvanica
A. altissima
S. japonica
M. denudata
P. tomentosa
G. biloba
U. pumila
P. occidentalis
S. matsudana
R. typhina
P. tabulaeformis
P. armandi
P. orientalis
cd
b
c
bc
bcd
ab
a

Fig. 3.

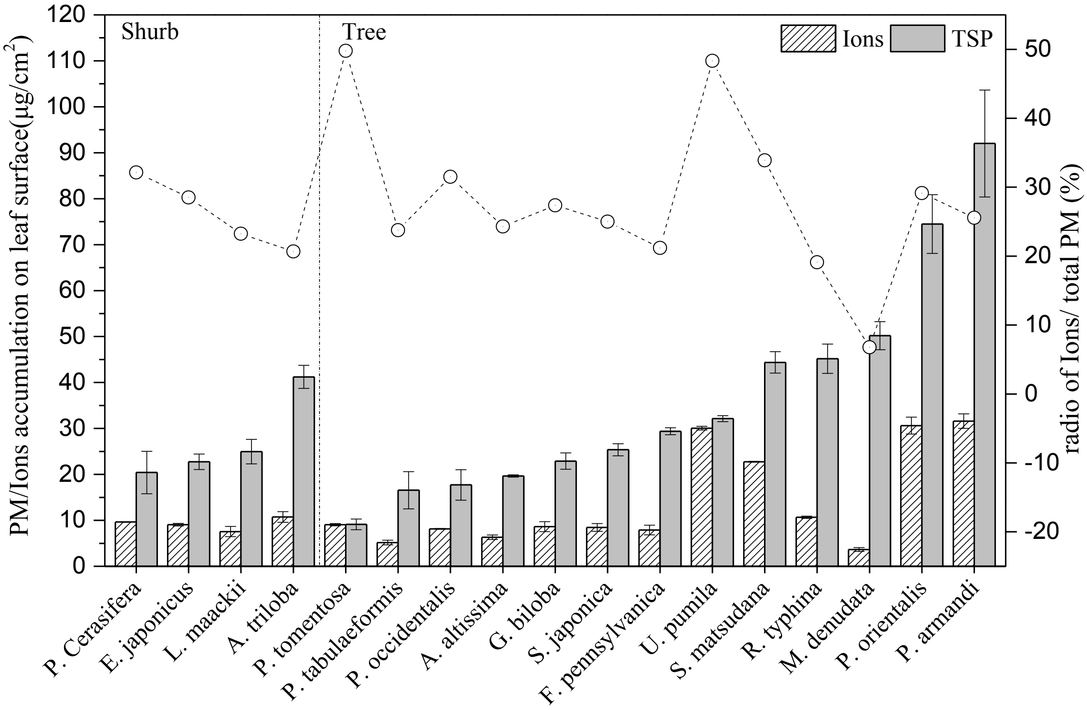

Shurb
Tree
Ions
TSP
PM/Ions accumulation on leaf surface(μg/cm^2)
radio of Ions/ total PM (%)
0
10
20
30
40
50
60
70
80
90
100
110
120
-20
-10
0
10
20
30
40
50
P. Cerasifera
E. japonicus
L. maackii
A. triloba
P. tomentosa
P. tabulaeformis
P. occidentalis
A. altissima
G. biloba
S. japonica
F. pennsylvanica
U. pumila
S. matsudana
R. typhina
M. denudata
P. orientalis
P. armandi

Fig. 4.

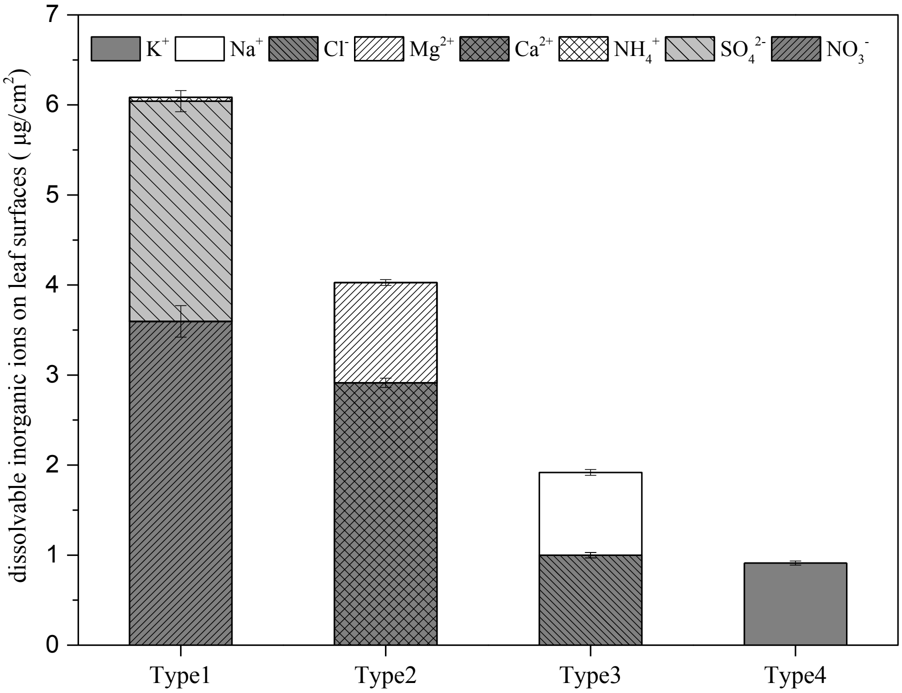

K+
Na+
Cl-
Mg2+
Ca2+
NH4+
SO42-
NO3-
dissolvable inorganic ions on leaf surfaces (μg/cm2)
0
1
2
3
4
5
6
7
Type1
Type2
Type3
Type4

Fig. 5.

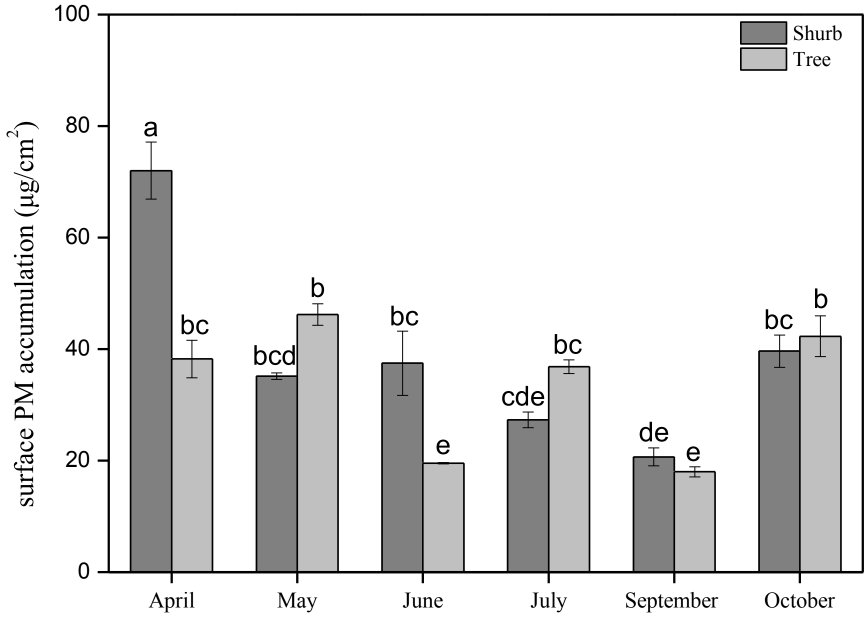

surface PM accumulation (μg/cm^2)
100
80
60
40
20
0
Shurb
Tree
a
bc
bcd
b
bc
e
cde
bc
de
e
bc
b
April
May
June
July
September
October

Fig. 6.

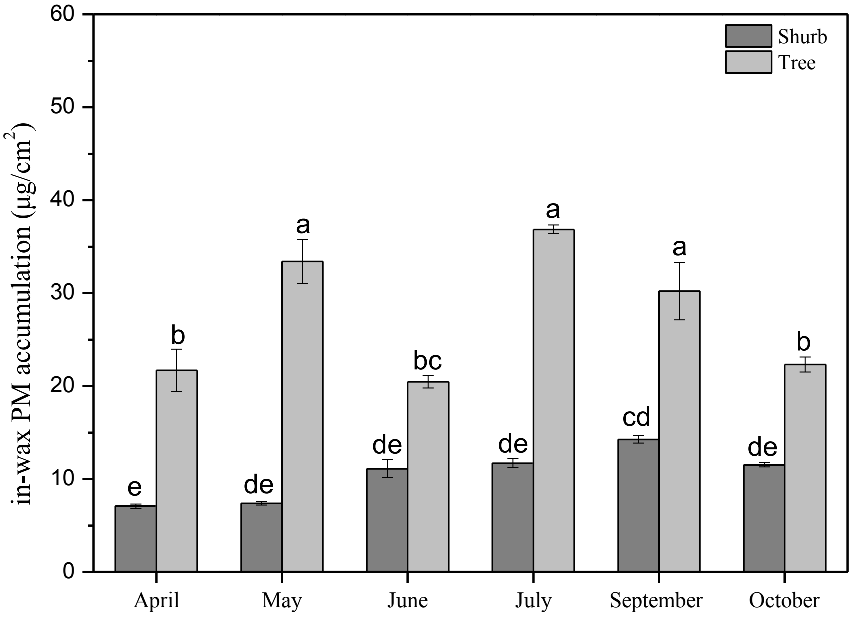

in-wax PM accumulation (μg/cm^2)
0
10
20
30
40
50
60
Shurb
Tree
e
b
de
a
de
bc
de
a
cd
a
de
b
April
May
June
July
September
October

Fig. 7.