The Nature and Size Fractions of Particulate Matter Deposited on Leaves of Four Tree Species in Beijing, China

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Abstract: Particulate matter (PM) in different size fractions (PM0.1–2.5, PM2.5–10 and PM10) accumulation on four tree species (Populus tomentosa, Platanus acerifolia, Fraxinus chinensis, and Ginkgo biloba) at two sites with different pollution levels was examined in Beijing, China. Among the tested tree species, P. acerifolia was the most efficient species in capturing PM, followed by F. chinensis, G. biloba, and P. tomentosa. The heavily polluted site had higher PM accumulation on foliage and a higher percentage of PM2.5 and PM2.5–10. Encapsulation of PM within cuticles was observed on leaves of F. chinensis and G. biloba, which was further dominated by PM2.5. Leaf surface structure explains the considerable differences in PM accumulation among tree species. The amounts of accumulated PM (PM0.1–2.5, PM2.5–10, and PM10) increased with the increase of stomatal aperture, stomatal width, leaf length, leaf width, and stomatal density, but decreases with contact angle. Considering PM accumulation ability, leaf area index, and tolerance to pollutants in urban areas, we suggest P. acerifolia should be used more frequently in urban areas, especially in “hotspots” in city centers (e.g., roads/streets with heavy traffic loads). However, G. biloba and P. tomentosa should be installed in less polluted areas.

Keywords: air pollution alleviation; accumulation on leaves; PM2.5; encapsulated particles; urban trees

1. Introduction

In Beijing, the capital of China, with rapid economic development, urbanization, and industrialization, ecological problems are becoming increasingly prominent. Air pollution, especially particulate matter (PM), has become one of the most severe problems over the past several decades [1]. Studies have shown that PM has recently been ranked fifth among the major risk factors threatening human health globally, and thus is first among environmental risks [2,3]. PM10 can cause premature mortality, accelerated atherosclerosis, lung cancer, heart disease, asthma, preterm birth, mutagenicity and DNA damage, and inflammatory responses [4–6]. Nevertheless, fine particles (particles with diameter less than 2.5 µm, PM2.5) are more toxic and more strongly associated with human health effects than coarse particles (particles with diameter between 2.5 and 10 µm) [7]. Therefore, the air quality standards for PM10 and PM2.5 in China are set to 75 and 35 µg/m³ (annual mean), and 150 and 70 µg/m³ (daily mean), respectively [8]. However, urban Beijing’s PM10 and PM2.5 concentrations are much higher than the national standards [9]. Thus, reducing PM concentrations, especially PM2.5, is considered one of the most significant tasks related to environmental protection in urban areas. The government has taken measures to control pollutant sources (e.g., adjusting the industrial structure and promoting energy-saving technology). Meanwhile, massive
afforestation is believed to be an additional and helpful measure to alleviate air pollution by filtering and adsorbing PM through forest crown/leaves.

Trees are a significant element in a city’s landscape and are the most effective vegetation types with regards to reducing PM [10]. This is mainly due to the fact that trees have extensive leaf areas [11], and the structure of tree crowns changes the turbulence of air movement above and within tree canopies [12]. McDonald et al. [13] estimated that a 3.7–16.5% increase in tree cover in West Midland, UK, could reduce PM concentrations by 10%. Reductions in total suspended particles (TSP), PM$_{10}$, PM$_{7}$, PM$_{4}$, PM$_{2.5}$, and PM$_{1}$ associated with an increase in canopy cover have been reported in China, the United States, Chile, and Israel [14–17], indicating a direct positive effect on air quality by removing PM$_{2.5}$ by urban trees. If a massive plantation was employed to decrease airborne PM$_{2.5}$, the differences in PM$_{2.5}$ accumulation among tree species should be considered and the most efficient species should be recognized. Some studies compared the PM$_{2.5}$ capturing ability of different plant species. Räsänen et al. [18] investigated the efficiency ($C_p$) of *Pinus sylvestris*, *Betula pubescens*, *Tilia vulgaris*, and *Betula pendula* leaves to capture PM$_{2.5}$ using simulation method (i.e., NaCl particles). They found that $C_p$ is influenced by leaf structures (e.g., leaf size, wettability, stomatal density, and leaf hair density). However, simulation studies are different from field investigations. Sæbø et al. [11] compared the PM accumulation on leaves of 47 species, including 22 trees and 25 shrubs, in Norway and Poland. They found a species-related difference in PM accumulation in both countries. The abilities of leaves to accumulate PM and its size fractions could be attributed to leaf morphology (e.g., leaf hair density, leaf roughness, and wax content) [6,19–21].

The detriments of PM on human health are primarily determined by particle size. Thus, the present study examined plants accumulating PM in two aspects. First, the amount of PM and its size fractions (PM$_{0.1–2.5}$, PM$_{2.5–10}$, and PM$_{>10}$) deposited on leaves in two contrasting urban environments were quantified. Second, the influences of anatomical/physiological leaf characteristics (e.g., stomatal density, stomatal size, single leaf area, wettability) on PM (i.e., PM, PM$_{0.1–2.5}$, PM$_{2.5–10}$, and PM$_{>10}$) accumulation abilities were investigated. Four tree species (*Populus tomentosa*, *Platanus acerifolia*, *Fraxinus chinensis*, and *Ginkgo biloba*) were selected as test species due to their prevalence in urban and suburban environments, widespread in temperate regions, and also since they are recommended for extensive plantation in Beijing. The findings of this study can provide the impetus for using urban trees to improve air quality, and provide guidance for the work of urban planners and those involved in environmental protection.

2. Materials and Methods
2.1. Plant Materials and Experimental Sites

Four tree species, *P. tomentosa*, *P. acerifolia*, *F. chinensis*, and *G. biloba*, were selected for this study at Beijing Botanical Garden (Site 1, located in Haidian District, 39°59′29.66″ N, 116°12′40.25″ E, upwind of Beijing) and Huangcun (Site 2, located in Daxing District, 39°42′45.13″ N, 116°19′08.44″ E, downwind of Beijing) (Figure 1). The sampling plants grow in the center of the garden or near a busy road with a traffic density of ~5680 cars/h at Site 1 and Site 2. The two sites showed different PM$_{2.5}$ and PM$_{10}$ concentrations, as measured at the nearest monitoring station operated by the Beijing Municipal Ecological and Environmental Monitoring Center [1].
2.2. Sampling Procedure

Leaf sampling was conducted on October 1 and 3, 2014 at Site 1 and Site 2, respectively, when there was no previous rainfall for more than a week. For each species, five individual trees under good growth conditions were selected. At Site 2, the distance between the sampled trees and the road center was about 10 m. Thus, the surrounding environment of each sample tree was similar. Small branches with mature and healthy leaves were cut from four dimensions (N, S, E and W) at 2–6 m above ground at each site and for each species. After cutting, small branches bearing leaves were placed in labeled ziplock bags, transported to the laboratory, and analyzed as soon as possible.

2.3. Analysis of PM

For each plant species at Sites 1 and 2, three batches of leaves were initially prepared. For each batch, 20–30 pieces for *P. acerifolia*, or 30–80 pieces for *P. tomentosa*, *F. chinensis*, and *G. biloba* were selected. The leaves were hand washed using a brush, with 200 mL of ultrapure water (ELGA, High Wycombe, Buckinghamshire, UK). The hemi-surface leaf area was measured using Image J software (Version 1.46; Wayne Rasband, National Institutes of Health, Bethesda, ML, USA) after scanning (HP Scanjet 3570c, HP Inc., Palo Alto, CA, USA). For the filtration procedure, membranes with pore size of 10, 2.5, and 0.1 µm were first soaked in ultrapure water for 2 h and then dried at 105 °C in a drying chamber for 6–8 h to remove soluble impurities. Every membrane was pre-weighed before filtration using a balance with 0.1 mg accuracy (SI-114, Denver Instrument Co., Arvada, CO, USA). The washing solution was hand-shaken for several minutes to re-suspend all washed particles before filtration. Washing solution was then pumped through membranes with pore size of 10, 2.5, and
0.1 µm successively. The filtration was carried out using a 47-mm glass filter funnel with stopper support assembly (Millipore Corp., Bedford, MA, USA) connected to a vacuum pump (SHB-III; Greatwall Scientific Industrial and Trade, Co., Ltd., Zhengzhou, China). Three fractions of PM were collected on the filters: (i) PM$_{>10}$ (particles intercepted by membrane with pore size of 10 µm, large), (ii) PM$_{2.5-10}$ (particles intercepted by membrane with pore size of 2.5 µm, coarse), and (iii) PM$_{0.1-2.5}$ (particles intercepted by membrane with pore size of 0.1 µm, fine). Loaded filters were subsequently dried for more than 24 h at 40°C, stabilized in the weighing room for 30 min, and then re-weighed. Consequently, the pre-weight was subtracted from the post-weight to calculate the mass of PM deposited on leaves in every size fraction of each washed sample. The resulting weight was finally divided by leaf area. At this moment, we obtained the total weight of deposited PM per unit leaf area for each sample, and also for PM$_{0.1-2.5}$, PM$_{2.5-10}$, and PM$_{>10}$.

The deposited PM$_{0.1-2.5}$, PM$_{2.5-10}$, PM$_{>10}$, and PM per unit green land was calculated by multiplying PM per unit leaf area and leaf area index (LAI). The LAI values were 2.13, 3.18, 2.67, and 2.52 for P. tomentosa, P. acerifolia, F. chinensis, and G. biloba, respectively, using a LAI-2000 (LI-COR., Inc., Lincoln, NE, USA) at Site 1.

2.4. Analysis of Leaf Surface Characteristics

Field emission scanning electron microscopy (FESEM, Quanta 200 FEG; FEI Company, Hillsboro, OR, USA) was used to determine leaf surface microstructures. Six pieces (three for upper side and three for lower side) of air-dried samples (about 5 mm × 5 mm) for each species at each site were cut from the center of the leaves, and coated with a thin layer of gold-palladium using a precision etching coating system (Model 682, Ga-tan Co., Ltd., Pleasanton, CA, USA). Stomatal density (per mm$^2$) was determined by calculating the number of stomata in ten FESEM images with a magnification of ×500. Particle frequency on stomata of each species was counted from 10 FESEM images with a magnification of ×1000. Fifty stomata were randomly selected to measure stomatal length, width, and aperture. Leaf roughness on upper and lower sides was evaluated on a subjective scale (1= relatively smooth, and 5 = very rough) [11] using FESEM images with a magnification of ×1000.

The wettability of leaf surface was evaluated by contact angle (CA) with distilled water. CAs were determined on upper and lower sides at room temperature using a goniometer (Kino SL200A, KINO Industry CO. Ltd., Somerville, Boston, MA, USA). For every species at every site, thirty pieces (about 5 mm × 5 mm, fifteen for upper side and fifteen for lower side) were cut from the middle of each leaf next to the main vein. Then, these pieces were attached to a glass plate with double-sided tape. A 3-µL water droplet was made with a capillary tube and carefully applied to the leaf surface. A photograph of the profile of each water droplet resting on the leaf surface was taken with a charge-coupled device equipped with a camera within 30 s after placing the water droplet. The digital photographs were downloaded, and CAs were determined using computer software (CAST2.0, KINO Industry CO. Ltd., Somerville, Boston, MA, USA). Then, the mean values were calculated.

Measurements of specific leaf area (SLA) and single leaf area, leaf length, leaf width, and petiole length were made on the same batches as were used for analysis of PM. After scanning, 30 leaves were randomly selected for measurements of single leaf area, leaf length, leaf width and petiole length using Image J software. The batches were dried in a drying chamber at 80 °C for at least 24 h and weighed to produce a value of dry weight. The dry weight and measured leaf area of the batches were used to calculate SLA as cm$^2$/g (dry weight).

2.5. Data Analysis

Statistical tests were performed with Minitab 16 software (Minitab Ltd., Shanghai, China). One-way analysis of variance was undertaken to estimate the differences in PM accumulation and its size fractions (PM$_{0.1-2.5}$, PM$_{2.5-10}$, and PM$_{>10}$) among different species at each site. The main effects of tree species, different sites, and their interaction on
accumulation of PM on leaves and its size fractions (PM$_{0.1–2.5}$, PM$_{2.5–10}$, and PM$_{>10}$) were tested with a two-way analysis of variance. The relative importance of measured leaf characteristics (stomatal density, stomatal length, stomatal width, stomatal aperture, CA upper side, CA lower side, single leaf area, SLA, leaf length, leaf width, petiole length, roughness upper side, roughness lower side) on the amount of PM and its size fractions was evaluated using principal component analysis (PCA) and partial least squares regression. A given effect was assumed to be significant at $p < 0.05$.

3. Results

3.1. Differences in PM and Its Size Fractions among Species

The total PM deposited on leaves was significantly different among four species at each site ($p < 0.001$, Table 1). The amounts of PM$_{0.1–2.5}$, PM$_{2.5–10}$, and PM$_{>10}$ ranged from 3.9–14.2, 5.7–41.2, 80.0–109.1, and 89.6–164.5 µg/cm$^2$; and 9.3–25.0, 15.9–51.7, 98.7–389.5, and 123.9–466.2 µg/cm$^2$, at Site 1 and Site 2, respectively. Among the four species, *P. acerifolia* had the greatest accumulated PM, followed by *F. chinensis*, *G. biloba*, and *P. tomentosa*. The mass of PM$_{0.1–2.5}$, PM$_{2.5–10}$, and PM$_{>10}$ accumulated on leaves also showed significant differences among tree species ($p < 0.001$, Table 1), except for PM$_{>10}$ at Site 1 ($p = 0.363$, Table 1). For all species, PM$_{>10}$ and PM$_{0.1–2.5}$ made up the greatest (66.4–89.2%) and smallest (4.4–10.0%) proportion of accumulated PM, respectively (Table 1).

The deposited PM$_{0.1–2.5}$, PM$_{2.5–10}$, PM$_{>10}$, and PM per unit green land were 8.3, 45.2, 18.7, and 16.1 µg/cm$^2$ (PM$_{0.1–2.5}$); 12.1, 131.0, 29.6, and 21.7 µg/cm$^2$ (PM$_{2.5–10}$); 170.4, 346.9, 249.1, and 209.7 µg/cm$^2$ (PM$_{>10}$), and 190.8, 523.1, 297.4, and 247.5 µg/cm$^2$ (PM), for *P. tomentosa*, *P. acerifolia*, *F. chinensis*, and *G. biloba*, respectively.

3.2. Differences in PM and Its Size Fractions among Sites

The PM deposited on leaves was significantly higher at Site 2 than that at Site 1 ($p < 0.001$, Table 1), with an increase of 40%, 180%, 40%, and 50% for *P. tomentosa*, *P. acerifolia*, *F. chinensis*, and *G. biloba*, respectively (Table 1). The corresponding increase at Site 2 compared with Site 1 was 20%, 260%, 30%, and 70% for PM$_{>10}$, 180%, 30%, 120%, and 70% for PM$_{2.5–10}$, and 140%, 80%, 120%, and 70% for PM$_{0.1–2.5}$. The ratio of PM$_{0.1–2.5}$/PM$_{0.1–10}$ was 0.38 at Site 1 and 0.36 at Site 2. Both are lower than that in ambient air, which was 0.82 at Site 1 and 0.69 at Site 2 during the growing season (May to October, 2014).

3.3. Morphological Structure of Leaf Surfaces

Table 2 and Figure 2 present the leaf surface structural properties of four studied tree species. *P. acerifolia* had the largest single-leaf area, followed by *P. tomentosa*, *F. chinensis*, and *G. biloba*. In terms of leaf wettability, all of the analyzed species, except the lower side of *G. biloba*, had a mean CA less than 90° (i.e., they were wettable) [22].

In the FESEM study, epicuticular wax was observed in tubular form (*G. biloba*, Figure 2j–l), or wax film (*P. tomentosa*, *P. acerifolia*, *F. chinensis*, Figure 2a–i). Wrinkled cuticles were observed on the lower side of *P. tomentosa* (Figure 2b,c), both surfaces of *P. acerifolia* (Figure 2d–f), lower side of *F. chinensis* (Figure 2h–i). The upper side of *F. chinensis* (Figure 2g) and *G. biloba* (Figure 2). Stomata of the investigated species were either level with epidermal cells (*P. biloba*, Figure 2b,c), sunken (*F. chinensis*, Figure 2h,i, G. biloba, Figure 2k–l), or slightly elevated (*P. acerifolia*, Figure 2e,f). *P. tomentosa* had smaller stomata than the other species. However, the greatest stomatal aperture occurred on the foliage of *P. acerifolia*. 
Table 1. Particulate matter and its size fractions accumulated on unit leaf area of trees (µg/cm²), size fraction percentage (%), shown in parenthesis, and the amount ratios (%) as compared between Beijing Botanical Garden (Site 1) and Huangcun (Site 2).

| Tree Species | Beijing Botanical Garden (Site 1) | Huangcun (Site 2) | Amount Ratios |
|--------------|-----------------------------------|-------------------|---------------|
|              | PM₀.₁–₂.₅ | PM₂.₅–₁₀ | PM₁₀ | Total PM | PM₀.₁–₂.₅ | PM₂.₅–₁₀ | PM₁₀ | Total PM | PM₀.₁–₂.₅ | PM₂.₅–₁₀ | PM₁₀ | Total PM |
| P. tomentosa | 3.9 ± 0.6 | 57 ± 1.1 | 80.0 ± 11.6 | 89.6 ± 13.2 | 9.3 ± 0.8 | 15.9 ± 1.7 | 98.7 ± 4.7 | 123.9 ± 5.5 | 2.4 | 2.8 | 1.2 | 1.4 |
| P. acerifolia | 14.2 ± 0.4 | 41.2 ± 7.0 | 109.1 ± 31.7 | 164.5 ± 30.1 | 25.0 ± 4.9 | 51.7 ± 7.6 | 389.5 ± 15.2 | 466.2 ± 26.9 | 1.8 | 1.3 | 3.6 | 2.8 |
| F. chinensis | 7.0 ± 2.0 | 11.1 ± 2.5 | 93.3 ± 23.2 | 111.4 ± 27.4 | 15.6 ± 4.5 | 24.1 ± 4.4 | 117.3 ± 20.6 | 157.0 ± 25.4 | 2.2 | 2.2 | 1.3 | 1.4 |
| G. biloba | 6.4 ± 2.3 | 8.6 ± 1.2 | 83.2 ± 2.7 | 98.2 ± 1.8 | 10.8 ± 3.2 | 18.0 ± 3.8 | 119.5 ± 11.6 | 148.3 ± 9.1 | 1.7 | 2.1 | 1.4 | 1.5 |

Table 2. Means (±SD) of leaf surface structural properties in the Beijing Botanical Garden (Site 1) and Huangcun (Site 2).

| Leaf Surface Structure Properties | Beijing Botanical Garden (Site 1) | Huangcun (Site 2) |
|-----------------------------------|-----------------------------------|-------------------|
|                                  | P. tomentosa | P. acerifolia | F. chinensis | G. biloba | P. tomentosa | P. acerifolia | F. chinensis | G. biloba |
| Stomatal density (/mm²) | 217.1 ± 42.4 | 245.2 ± 56.8 | 201.5 ± 12.6 | 69.0 ± 22.4 | 278.8 ± 84.9 | 288.0 ± 26.3 | 423.5 ± 135.3 | 187.4 ± 37.8 |
| Stomatal length (µm) | 17.8 ± 1.5 | 29.1 ± 4.3 | 23.5 ± 4.6 | 23.1 ± 4.9 | 16.7 ± 3.0 | 20.9 ± 2.7 | 18.6 ± 2.7 | 15.2 ± 2.7 |
| Stomatal width (µm) | 7.4 ± 0.9 | 22.3 ± 3.6 | 11.3 ± 4.4 | 8.7 ± 2.2 | 5.3 ± 1.2 | 15.2 ± 1.5 | 8.0 ± 1.7 | 5.0 ± 1.6 |
| Stomatal aperture (µm) | 2.1 ± 1.0 | 7.3 ± 2.1 | 7.3 ± 2.1 | 2.3 ± 1.2 | n.d. | 1.9 ± 0.7 | 6.3 ± 1.9 | 2.5 ± 0.6 |
| Stomatal with particle (%) | 7.22 ± 1.5 | 66.4 ± 123.9 | 43.0 ± 25.9 | 14.6 ± 18.5 | 92.9 ± 12.2 | 89.9 ± 20.2 | 63.4 ± 18.0 | 11.0 ± 15.2 |
| Contact angle (upper side) (°) | 75.7 ± 4.6 | 67.4 ± 4.9 | 74.6 ± 6.2 | 74.0 ± 7.6 | 53.5 ± 7.0 | 63.8 ± 8.9 | 60.9 ± 3.1 | 68.8 ± 4.6 |
| Contact angle (lower side) (°) | 65.3 ± 3.2 | 67.1 ± 4.6 | 61.1 ± 7.8 | 103.1 ± 10.3 | 63.6 ± 8.1 | 56.3 ± 6.4 | 61.8 ± 5.7 | 93.2 ± 5.1 |
| Single leaf area (cm²) | 84.9 ± 24.7 | 117.3 ± 22.3 | 24.8 ± 6.1 | 17.4 ± 3.6 | 53.6 ± 12.0 | 92.0 ± 18.8 | 19.4 ± 3.3 | 7.6 ± 2.9 |
| Specific leaf area (cm²/g) | 89.6 ± 9.1 | 116.1 ± 9.6 | 191.0 ± 17.6 | 191.8 ± 7.3 | 106.8 ± 6.9 | 140.8 ± 17.1 | 129.7 ± 13.0 | 121.8 ± 10.8 |
| Leaf length (cm) | 10.1 ± 1.6 | 18.2 ± 2.3 | 9.8 ± 1.5 | 4.5 ± 0.6 | 7.8 ± 1.3 | 14.7 ± 1.3 | 8.6 ± 0.9 | 3.4 ± 0.6 |
| Leaf width (cm) | 9.3 ± 0.9 | 19.9 ± 1.0 | 4.6 ± 0.6 | 6.2 ± 1.0 | 7.1 ± 1.0 | 15.5 ± 1.7 | 3.9 ± 0.4 | 4.3 ± 1.2 |
| Petiole length (cm) | 8.6 ± 1.0 | 7.7 ± 1.1 | 1.6 ± 1.1 | 4.5 ± 1.7 | 5.2 ± 0.9 | 4.8 ± 1.2 | 1.2 ± 0.8 | 3.4 ± 1.1 |
| Roughness (upper side) | 1 | 3 | 3 | 4 | 2 | 3 | 3 | 4 |
| Roughness (lower side) | 3 | 4 | 4 | 5 | 3 | 4 | 4 | 5 |

n.d. indicates variables that were not found.
Figure 2. Scanning electron microscopy images of *Populus tomentosa* (a–c), (d–f), (g–i), and (j–l). a, d, g, j, m, and n, the upper side; b, e, h, and k, the lower side; c, f, i, and l, stomata. Particulate matter embedded in leaf epidermis (m: *Fraxinus chinensis*; n: *Ginkgo biloba*). Symbols indicate examples of stomata (triangle) and particulate matter (arrow).
3.4. Encapsulation of PM

PMs within cuticles were observed for *F. chinensis* (Figure 2m) and *G. biloba* (Figure 2n), but not for *P. tomentosa* and *P. acerifolia*. Encapsulation of PM within cuticles was not so common using FESEM observation. No visible damage was observed to either cuticle or epidermal cell. Wax encapsulated particles had diameters less than 6 μm, which was dominated by PM$_{2.5}$ (>90%).

3.5. The Effects of Leaf Structure on PM Accumulation

Stomatal aperture, stomatal width, leaf length, leaf width, stomatal density, and CA were crucial predictors for PM accumulation and its size fractions (Table 3, Figure 3). Among the 13 dependent variables, the sum of PC#1 and PC#2 was 71.8% (Figure 3). An increase in CA predicted a decrease in PM accumulation, while increased stomatal aperture, leaf length, leaf width, stomatal width, and stomatal density predicted an increase in PM accumulation (Table 3).

**Table 3.** Regression coefficient (B) and standardized regression coefficient (Beta) of the partial least squares regression model for the factors affecting leaf particulate matter capturing of four tree species.

|                      | PM        | PM$_{2.5}$ | PM$_{0.1-2.5}$ |
|----------------------|-----------|------------|----------------|
|                      | B         | Beta       | B              | Beta       | B          | Beta       |
| Stomatal density     | 0.038     | 0.031      | 0.030          | 0.029      | 0.006      | 0.038      | 0.003      | 0.052      |
| Stomatal length      | 0.057     | 0.002      | 0.015          | 0.001      | 0.122      | 0.034      | 0.018      | 0.012      |
| Stomatal width       | 0.833     | 0.039      | 0.648          | 0.036      | 0.182      | 0.065      | 0.053      | 0.046      |
| Stomatal aperture    | 2.757     | 0.060      | 2.254          | 0.058      | 0.470      | 0.077      | 0.162      | 0.064      |
| Contact angle (upper) | −0.473    | −0.030     | −0.362         | −0.027     | −0.074     | −0.035     | −0.041     | −0.047     |
| Contact angle (lower) | −0.255    | −0.035     | −0.235         | −0.039     | −0.036     | −0.037     | −0.015     | −0.039     |
| Single leaf area     | −0.036    | −0.011     | −0.039         | −0.014     | 0.011      | 0.027      | 0.000      | 0.001      |
| Specific leaf area   | 0.062     | 0.015      | 0.059          | 0.017      | 0.003      | 0.005      | 0.003      | 0.014      |
| Leaf length          | 1.12      | 0.045      | 0.929          | 0.044      | 0.217      | 0.065      | 0.067      | 0.049      |
| Leaf width           | 1.049     | 0.050      | 0.847          | 0.048      | 0.188      | 0.067      | 0.054      | 0.047      |
| Petiole length       | −0.194    | −0.004     | −0.138         | −0.003     | 0.053      | 0.008      | −0.040     | −0.015     |
| Roughness (upper side)| 2.224     | 0.018      | 1.238          | 0.012      | 0.279      | 0.017      | 0.163      | 0.024      |
| Roughness (lower side)| 1.641     | 0.010      | 0.636          | 0.005      | 0.112      | 0.005      | 0.096      | 0.011      |

**Figure 3.** Loading plot of principal component analysis of the tree species with 13 dependent variables.
4. Discussion

4.1. Differences in PM Accumulation among Species

The plant species showed significant differences in PM accumulation among tree species and at different sites. In a study conducted by Sæbø et al. [11], they found that *Pinus mugo*, *P. sylvestris*, *Taxus media*, *Taxus baccata*, *Stephanandra incise*, and *B. pendula* showed higher PM accumulation. *Acer platanoides*, *Prunus avium*, and *Tilia cordata* showed lower PM accumulation. Zhang et al. [23] compared foliar PM retention and its size fractions of five plant species, *P. acerifolia* showed higher amount of PM accumulation ability than other species.

The differences in PM accumulation among species should be used for species selection during afforestation. However, tree size and LAI are important in determining the amount of PM accumulated per unit of land area, which is an index for PM retention ability and efficiency [24,25]. The relatively higher LAI of *P. acerifolia* further increased its PM removal potential, suggesting that this species could be used suitably as PM filters. The trees in Beijing could be damaged by air pollution; thus, the actual LAI was lower than healthy trees, which would influence the amount of accumulated PM. Among the investigated tree species, *P. acerifolia* is tolerant of air pollution, *G. biloba* and *P. tomentosa* are sensitive [26]. Therefore, we suggest that *P. acerifolia* should be used more in urban areas, especially in “hotspots” of PM pollution (e.g., the middle of the city, edges of streets/roads with heavy traffic loads). However, *G. biloba* and *P. tomentosa* should be installed in less polluted areas.

The contribution of PM in different size fractions to total PM on leaves decreased with the decreasing of PM diameter. Terzaghi et al. [27] found that large, coarse, and fine particles accounted for 87–95%, 5–12%, and 0.1–0.5% of the total PM for *Cornus mas*, *Acer pseudoplatanus*, and *Pinus pinea*. A study taken along a busy road in UK found the amount of PM captured on leaves increased with decreasing particle diameter [28]. These varied deposition level of PM with different aerodynamic diameter probably reflects the different aerodynamic properties of PM and their interactions with different leaf characteristics. Meanwhile, this difference can be ascribed to two groups of factors. The first group include PM concentration and compositions, plant species, and filtering materials (filter paper or membrane). The second group are mainly the used methods (measured mass or mass derived from particle number). Particles were assumed to be spherical and particle density equal for each size class [27].

4.2. Differences in PM Accumulation between Sites

More PM was found on the foliage of plants grown at heavily polluted site (Site 2) than less polluted site (Site 1). Deposition of pollutants depends on deposition velocity and pollutant concentration [29,30]. Micrometeorological conditions have been demonstrated to strongly impact on deposition dynamics, which may be partly explain the differences in PM accumulation between the two sites in our study. Furthermore, the deposition of PM on leaves depends on PM diameter. Deposition of PM with sizes of 0.1–1 µm is influenced by Brownian diffusivity and Stokes’ law, and which is independent of size. For PM with diameter large than 1 µm, Stokes’ law dominates the process of deposition, and which depends mostly on particle size. Larger particles will, therefore, deposit faster than smaller ones, either by sedimentation under the influence of gravity or by turbulent transfer resulting in impaction and interception [31], leading to a lower ratio of PM$_{0.1–2.5}$/PM$_{0.1–10}$ in Site 2 than the less polluted site.

In the present study, we observed a relatively higher percentage of fine (0.1–2.5 µm) and coarse (2.5–10 µm) size fractions in Site 2 than those in Site 1, while the large fractions were in lower percentages at Site 2. This finding seems to contrast with the results of Przybysz et al. [32], who obtained a relatively high rate of fine particles (6.2–9.8%) on plants at a rural site. Here, the lower ratio of PM$_{0.1–2.5}$/PM$_{0.1–10}$ accumulated on leaves, compared with that in ambient air, may be caused by three factors. First, some particles, elements, or ions are dissolved in water during the washing process [33]. Second, during the process of filtration, the membrane with pore sizes of 10 µm and (or) 2.5 µm may intercept some
particles with diameters less than 10 µm and (or) 2.5 µm after saturation [34]. Third, the leaf cuticle encapsulated some particles, especially tiny ones [27].

4.3. Importance of Leaf Structure for PM Accumulation

Urban trees accumulating PM is a complex and poorly characterized process which is influenced by many factors such as leaf surface micro-, and macro-structure (e.g., leaf wettability, stomatal density, leaf area, leaf roughness, and the shape and amount of trichomes) [11,23,24,35]. Research has shown that leaves with small size, low CAs, low stomatal density, high stomatal conductance, and higher amount of leaf hairiness can capture more PM [11,18,19,21,31]. In this study, we also found that high CAs decreased the amount of captured PM. The contact area between a particle and the underlying leaf surface is considerably reduced on surfaces with high CAs. Consequently, the physical adhesion forces between particles and leaf surfaces are reduced, leading to a lower PM accumulation [19]. However, we also found that increased stomatal aperture, density, and width increased the amounts of captured PM. Large stomatal density, aperture, and width could result in increased transpiration which can make particles more deliquescent. And as a consequence, deposition rates increase [36]. Transpiration of water through stomata can cool leaf surfaces, but increase PM deposition by thermophoresis [18], which may also partly explain the higher ability of leaves to capture PM under more polluted conditions than comparatively less polluted areas.

We found that P. acerifolia, the species with the largest leaves, had the highest PM accumulation among the investigated species. This is in opposition to the findings of previous studies, in which the authors found small leaves increased the number of captured particles [18,28]. According to Nobel [37], particles in the air could more easily collide with small leaves than large and flat leaves, which have thicker boundary layers. This contrasting finding may be caused by the microstructure of P. acerifolia leaves, which had a rough surface that can influence the boundary layer [38].

Some PM with diameters less than 6 µm (mainly PM$_{2.5}$) were encapsulated in cuticles of leaves of F. chinensis and G. biloba, but not for P. acerifolia and P. tomentosa, by FESEM observation. These results suggest that the potential of PM embedded in wax layer depends on the quantity of wax as well as the composition and structure of the epicuticular wax layer; these are species-specific characteristics [39]. Terzaghi et al. [27] found that particles with diameters less than 10.6 µm were encapsulated into cuticles. The amount of encapsulation and the capacity of leaves to capture PM changed over time. They attributed this to the degradation of cuticular waxes, from a perfect wax crystal to an amorphous one. Dzierzanowski et al. [39] demonstrated that large particles appeared mainly on the leaf surface rather than in the wax layer of some plant species. The encapsulated particles always had small diameters and could not be easily washed off during rain events or dislodged by wind. If most of the accumulated PM is immobilized which can be considered to be beneficial in the planning of PM phytoremediation. However, if the washed- or blown-off PM is considered to be filter cleaning, leaving the leaves ready for additional deposition. This process may result in underestimating the PM removal effect [23]. The dynamics of deposition, including the amounts of PM washed-off by rain and blown-off by wind, need further investigation.

5. Conclusions

(i) The amounts of accumulated PM differed significantly among species, in the order of P. acerifolia > F. chinensis > G. biloba > P. tomentosa. Most of the accumulated PM belonged to the largest fraction (>10 µm). Some PM was encapsulated in cuticles of F. chinensis and G. biloba, and which was dominated by PM$_{2.5}$ (>90%).

(ii) Trees at polluted site had higher rates of PM accumulation and higher percentage of fine and coarse fractions than less polluted site. With the increase of pollution level, the PM retention ability of tree species increased with the decrease of particle size, indicating that plant leaves could accumulate fine particles and purify local air.
(iii) Leaf structures affect PM accumulation and its size fractions. Large leaves, along with low stomatal aperture, width, and density, as well as low CA, all resulted in increased PM capture.

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