Seasonal variations in accumulated particulate matter on leaves of four major tree species in Korea

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ABSTRACT
Tree planting is a countermeasure used to mitigate the effects of the high concentrations of atmospheric particulate matter (PM), because trees have a strong ability to adsorb PM. However, owing to the limited information on PM accumulation on leaves throughout the year, guidelines for the implementation of urban forests cannot be provided. Therefore, in this study, we measured PM accumulation in leaves to compare the PM-reducing ability of four common species (Metasequoia glyptostroboides, Prunus yedoensis, Spirea prunifolia f. simpliciflora, and Zelkova serrata) during one growing season. We collected leaves almost every two weeks from 7 May to 23 October 2019, in Seoul Forest Park. We then measured the PM quantities on the leaves before and after rainfall (28 mm/h) to determine the amount of PM that washed off. We found that the average PM10 (PM <10 µm in diameter) accumulation on the leaves of S. prunifolia f. simpliciflora, Z. serrata, M. glyptostroboides, and P. yedoensis during one growing season was 68.1, 58.3, 43.5, and 28.2 mg/m², respectively. The average PM2.5 (PM <3 µm) accumulation on the leaves of S. prunifolia f. simpliciflora, Z. serrata, M. glyptostroboides, and P. yedoensis was 18.2, 11.6, 7.8, and 6.5 mg/m², respectively. However, the accumulation of both PM10 and PM2.5 on the leaves of the four species fluctuated during the sampling period. The average ratio of PM2.5 to PM10 accumulation in the leaves of S. prunifolia f. simpliciflora and P. yedoensis was significantly higher than that of Z. serrata and M. glyptostroboides; however, this ratio fluctuated throughout the sampling period. Rainfall considerably reduced the levels of PM10 on the leaves of P. yedoensis and PM2.5 on the leaves of M. glyptostroboides. We found no significant removal of PM from the leaves of other species. These results indicate that the PM-reducing ability of trees varies between species and over time. Therefore, urban forests should be managed with a high diversity of tree species.

1. Introduction
Particulate matter (PM) poses a serious threat to human health worldwide (Dockery et al. 1993; Pope et al. 1995; Anenberg et al. 2019). It causes respiratory illness because these particles penetrate deep inside the lungs (Jo et al. 2017; Kyung and Jeong 2020). Moreover, PM with a diameter less than 2.5 µm (PM2.5) can infiltrate the circulatory system, increasing morbidity and mortality (Maté et al. 2010; Achilleos et al. 2017). The main sources of PM are secondary inorganic species, dust, sea salt, traffic, industry, biomass burning, and coal/oil combustion (Hopke et al. 2020). Many countries are working toward reducing PM by replacing old with modern boilers, installing reduction devices in factory chimneys, phasing out diesel-only trains, reducing ammonia emissions from farming, among others (EU 2013; USA The Law Library of Congress 2018; India Ministry of Environment, Forest & Climate Change 2019; UK Department for Environment and Food and Rural Affairs 2019). The U.K. plans to reduce PM emissions by 30% by 2020 and 46% by 2030 (UK Department for Environment and Food and Rural Affairs 2019). The EU National Emission Ceilings Directive requires an ammonia reduction of 27% to achieve new air policy targets for 2030 (EU 2013). The national PM2.5 and PM10 concentration target in India is a 20–30% reduction by 2024 (India Ministry of Environment, Forest & Climate Change 2019).

Among the various efforts implemented to reduce PM emissions in many countries, tree planting is a cost-effective and environmentally friendly approach. Forests and trees act as efficient natural sinks for removing PM from the atmosphere. As trees and forests in the conterminous United States removed an estimated 696,000 tons of PM2.5 in 2010, 557 deaths, 169,701 acute respiratory symptoms, 129 hospital admissions, and 203 emergency room visits were avoided (Nowak et al. 2014). In addition, trees in the central part of Beijing removed an estimated 772 tons of PM10 in 2002 (Yang et al. 2005). Forests remove PM through four regulatory processes: deposition (43.6%), blocking (34.8%), accumulation (21.5%), and intake (0.1%) (Yu 2017). The forest canopy structure creates a specific microclimate that changes both air
current and velocity, thus facilitating the deposition or blocking of PM (Yu 2017). The microstructure of leaf surfaces and large leaf surface areas remove PM through accumulation (Yu 2017). PM intake occurs through the stomata of the leaves, but the amount removed through intake is low compared with the amount removed through the other processes mentioned above (Yu 2017).

According to the Korea Forest Service, more than 728.8 ha of forest will be established by 2025 to block and reduce airborne PM and increase human well-being (Korea Forest Service 2022). However, prior to forestation, the tree species most effective at reducing PM, although covering a limited land area, must be selected. Researchers have estimated the deposition capacity of PM based on foliar characteristics and the amount of PM accumulated on the leaves of various tree species (Dzierzanowski et al. 2011; Saebø et al. 2012; Nowak et al. 2014; Chen et al. 2015; Song et al. 2015; Zhang et al. 2015; Chen et al. 2017). These researchers generally examined the amount of PM on leaves several times per year and were limited to a short period of a single growing season; thus, the amount of PM accumulated on leaves was evaluated based on limited data (Saebø et al. 2012; Popek et al. 2019; Jin et al. 2021; Xu et al. 2021). However, trees grow throughout the year, and both the leaf morphological features and chemical composition of epicuticular wax change (Jetter and Schäffer 2001). In addition, weather conditions, such as wind speed and rainfall, change throughout the year, which affect the ability of the species to capture and retain PM on leaves (Pullman 2009; Wang et al. 2013; 2015; Jin et al. 2021). Thus, the limited period and frequency of PM adsorption research on plant species might have led to misconceptions regarding the true PM adsorption ability of plants.

Therefore, to select more effective tree species for reducing PM in urban forests, we studied 1) the PM accumulation capacities of four tree species under various PM concentrations and 2) the impact of rainfall on PM accumulation on the leaves of the four tree species.

2. Materials and methods

2.1. Study site and plant materials

The study site was Seoul Forest Park, a public open space located in Seoul, South Korea (37°33'N, 126°58'E). We selected three deciduous species (Metasequoia glyptostroboides Hu & W. C. Cheng, Prunus × yedoensis Matsum., and Zeikova serrata (Thunb.) Makino Hu & W. C. Cheng), and one deciduous shrub species (Spirea prunifolia f. simpliciflora Nakai) to evaluate their capacity to trap both PM_{10} and PM_{2.5} on their leaf surfaces. These four tree species are common on roadsides and urban woodlands in South Korea. P. yedoensis, Z. serrata, and M. glyptostroboides were the 1st, 4th, and 8th most planted tree species on roadsides from 2015 to 2017, respectively (Korea Forest Service 2018). S. prunifolia was the 5th most planted shrub species on roadsides in 2017 (Korea Forest Service 2018). The above four tree species were randomly distributed at the study site of Seoul Forest Park, so we selected them as the study tree species.

2.2. Atmospheric PM and weather data

We downloaded the atmospheric PM and weather data (wind speed and rainfall) collected at the monitoring station nearest to the Seoul Forest Park for analysis (http://www.airkorea.or.kr, http://data.go.kr). Table 1 presents the PM data collected during the study period. The mean PM concentration is the mean hourly PM data for each data collection period (Figure 1(a)). We calculated the sum of the PM inflows as

\[
S = \sum_{i} (C_i \times W_i)
\]

where \(S\) is the sum of the PM inflow (\(\mu g/m^2\cdot s\)), \(i\) is the data collected on an hourly basis, \(n\) is the last hour of the data collection period, \(C\) is the hourly PM concentration (\(\mu g/m^3\)), and \(W\) is the hourly wind speed (\(m/s\)) (Figure 1(b)). From July 16 to 17, the sum of the PM inflow was the lowest because this period had the lowest mean wind speed and the data collection period was the shortest compared to other periods. We calculated the mean ratio of atmospheric PM_{2.5} to PM_{10} using the average ratio of hourly PM_{2.5} to PM_{10} within each data collection period (Figure 1(c)). We calculated the mean rainfall and wind speed using the average hourly rainfall and hourly wind speed data for each data collection period (Figure 1(d)).

2.3. Sample collection and leaf-washing protocol

Table 1 lists the sampling dates, PM data, and weather data we obtained during the experimental period. We sampled leaves almost every two weeks from 7 May to 23 October 2019, from four or five individual trees (replicates) of each species. To calculate the amount of PM washed off tree leaves by rain, we immediately sampled leaves before (on July 15) and after (on July 17) a rainfall event on July 15 (the maximum precipitation was 28 mm/h, seasonally the heaviest rainfall) (Figure 2). The rainfall duration of this experiment was three hours on July 15 (3–5 p.m.), and the intensity was 28 mm/h for the first hour, 12.5 mm/h for the next hour, and 1 mm/h for the last hour, and 2 h on July 16 (2–3 a.m.), with a rainfall intensity of 6 mm/h for the first hour and 9.5 mm/h for the next hour. A heavy rainfall (≤15 mm/h) event occurred for 6 days (13% of total rainy days) during the experimental period.

We randomly collected the bottom outermost branches bearing leaves from three or four different sides of individual trees. Immediately after cutting, we placed the branches in labeled paper bags, which we transported to the laboratory. We collected a total of 40 leaves from M. glyptostroboides and S. prunifolia.
Table 1. Sampling dates, particulate matter (PM) data, and weather data during the experimental period in 2019.

| Sampling date (data collection period) | Mean PM concentration ($\mu g/m^3$) | Sum of PM inflow ($\mu g/m^2\cdot s$) | Mean wind speed (m/s) | Mean rainfall (mm/h) |
|----------------------------------------|-------------------------------------|--------------------------------------|-----------------------|----------------------|
|                                        | PM$_{10}$  | PM$_{2.5}$                  | PM$_{10}$  | PM$_{2.5}$ |                           |                         |                         |
| May 7 (Apr 22–May 7)                   | 61.9       | 30.3                        | 45545.6   | 21182.4   | 2.1                       | 0.05                    |
| May 20                                 | 63.1       | 35.0                        | 36686.8   | 18655.1   | 1.9                       | 0.08                    |
| May 30                                 | 64.6       | 32.9                        | 34754.6   | 16537.9   | 2.4                       | 0.02                    |
| Jun 13 (May 31–Jun 13)                 | 39.3       | 23.4                        | 26416.4   | 15297.8   | 2.1                       | 0.18                    |
| Jun 27 (Jun 14–Jun 27)                 | 29.0       | 17.2                        | 18746.1   | 10793.8   | 2.0                       | 0.04                    |
| Jul 15 (Jun 28–Jul 15)                 | 26.5       | 18.5                        | 24787.4   | 16161.6   | 2.4                       | 0.13                    |
| Jul 17                                 | 47.8       | 39.0                        | 2406.7    | 1938.5    | 1.1                       | 0.32                    |
| Jul 29 (Jul 16–Jul 17)                 | 23.8       | 16.7                        | 12680.2   | 8017.8    | 2.0                       | 0.45                    |
| Aug 20 (Jul 30–Aug 20)                 | 20.3       | 10.7                        | 19794.5   | 9969.4    | 1.9                       | 0.40                    |
| Sep 2 (Aug 21–Sep 2)                   | 26.3       | 14.4                        | 13938.8   | 7445.2    | 1.7                       | 0.17                    |
| Sep 17 (Sep 3–Sep 17)                  | 17.3       | 7.5                         | 10916.0   | 3967.7    | 2.0                       | 0.35                    |
| Oct 10 (Sep 18–Oct 10)                 | 26.7       | 12.1                        | 20159.6   | 8482.8    | 1.5                       | 0.09                    |
| Oct 23 (Oct 11–Oct 23)                 | 26.4       | 13.4                        | 10161.3   | 4599.5    | 1.4                       | 0.00                    |

PM$_{10}$ refers to particulate matter with a diameter less than 10 $\mu m$. PM$_{2.5}$ refers to particulate matter with a diameter less than 2.5 $\mu m$. The sum of the PM inflow was calculated as the sum of the mean hourly PM concentration multiplied by the mean hourly wind speed. Mean wind speed and mean rainfall are based on mean hourly data.

Figure 1. Atmospheric particulate matter (PM) and weather data during the data collection period. (a) Mean concentration of atmospheric PM, (b) Sum of PM inflow, (c) Mean ratio of PM$_{2.5}$ to PM$_{10}$, (d) Mean rainfall and wind speed data.
For *P. yedoensis* and *Z. serrata*, we collected 20 leaves because of their large leaf area.

To wash off the PM on the leaves without damaging the leaf surfaces, we modified the methods reported by Dzierzanowski et al. (2011), Sgrigna et al. (2015), and Liu et al. (2018) through preliminary experiments. We soaked the leaves collected from each tree in a glass jar containing 700 mL of distilled water. We shook the jar at 140 rpm for 10 min at 24°C and sonicated for 5 min to desorb PM from the leaf surfaces. We measured leaf area to quantify the PM mass per unit leaf area after we removed the leaves from the glass jar. We used a leaf-area meter (Li-3100, Li-COR, USA) to measure leaf area, which we then doubled to include both leaf sides (Saebø et al. 2012).

### 2.4. Quantitative analysis of PM on leaves

We used the filtration-weighing method to quantitatively analyze the PM on the leaves (Dzierzanowski et al. 2011; Song et al. 2015; Chen et al. 2017; Xu et al. 2019). Prior to filtration, we placed all the filters in a desiccator (KA-33, Kastech, Korea) at 20°C and 30% humidity for 24 h, which we then weighed (cpa2p-f, Sartorius, Germany). We filtered distilled water containing desorbed PM with 10.0 μm nylon filters, followed by 3.0 μm mixed cellulose ester (MCE) membrane filters, and finally 0.22 μm MCE membrane filters (Millipore Merck, Germany). After filtration, we dried the filters in a desiccator for 24 h under the same conditions, which we then reweighed. We calculated the mass of PM$_{10}$ on the leaf surfaces from the sum of the mass of PM on the 3.0 and 0.22 μm filters (10 μm > PM$_{10}$ > 0.22 μm). We used the mass of PM on the 0.22 μm filter as the mass of PM$_{2.5}$ on the leaf surfaces (3 μm > PM$_{2.5}$ > 0.22 μm).

To analyze the effect of PM washed off tree leaves by rain, mean reduction rate was calculated by

\[
\sum\frac{n}{i} \left(1 - \frac{i \text{ after rainfall}}{i \text{ before rainfall}}\right) \times 100
\]

where *i* is surface PM mass of each sample tree and *n* is sample tree number of same species.

### 2.5. Quantitative analysis of leaf morphological traits

To understand the effect of morphological leaf traits on the PM accumulation on the leaf surface, we used leaf trait data from Je et al. (2020) and Son et al. (2021) for correlation analysis (Table 2). The traits we used for the analysis included the leaf area, length and density of stomata on each of the adaxial and abaxial leaf surfaces, length and density of trichomes on each of the adaxial and abaxial leaf surfaces (each leaf surface divided by vein and leaf blade), and roughness of adaxial and abaxial leaf surfaces, which we measured using a 3D optical microscope at 50× magnification (Contour GT-K, Bruker) (Kwak et al. 2020).

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**Table 2. Leaf surface morphological traits (data from Je et al. (2020) and Son et al. (2021), mean ± SD).**

| Traits                  | *Metasequoia glyptostroboides* | *Prunus yedoensis* | *Zelkova serrata* | *Spirea prunifolia f. simpliciflora* |
|-------------------------|-------------------------------|--------------------|-------------------|---------------------------------------|
| Leaf area (cm$^2$)      | 18.9 ± 4.4                    | 45.9 ± 8.0         | 28.9 ± 5.6        | 6.4 ± 1.0                             |
| Roughness (μm) adaxial  | 3.9 ± 1.3                     | 3.8 ± 0.3          | 0.9 ± 0.1         | 1.4 ± 0.2                             |
| abaxial                 | 3.3 ± 0.4                     | 4.1 ± 0.3          | 0.6 ± 0.2         | 1.4 ± 0.3                             |
| Trichome Length (μm)    |                               |                    |                   |                                       |
| adaxial                 | –                             | –                  | 159.3 ± 86.7      | 82.8 ± 38.6                           |
| abaxial                 | –                             | –                  | 367.6 ± 75.6      | 153.5 ± 83.2                         |
| Density (no./mm$^2$)    |                               |                    |                   |                                       |
| adaxial main vein       | –                             | –                  | 92.6 ± 46.0       | 78.6 ± 19.5                           |
| lateral vein blade      | –                             | –                  | 100.5 ± 20.7      | –                                     |
| abaxial main vein       | –                             | –                  | 121.1 ± 4.9       | –                                     |
| lateral vein blade      | –                             | –                  | 112.4 ± 24.9      | 36.7 ± 21.9                           |
| Stomata                 |                               |                    |                   |                                       |
| Length (μm) abaxial     | 22.9 ± 11.7                   | 12.0 ± 4.1         | 346.5 ± 48.3      | 913.6 ± 195.0                        |
| Density (no./mm$^2$) abaxial | 233.2                       | 524.0 ± 51.9       | 7.2 ± 2.3         |                                       |

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For *P. yedoensis* and *Z. serrata*, we collected 20 leaves because of their large leaf area.

To wash off the PM on the leaves without damaging the leaf surfaces, we modified the methods reported by Dzierzanowski et al. (2011), Sgrigna et al. (2015), and Liu et al. (2018) through preliminary experiments. We soaked the leaves collected from each tree in a glass jar containing 700 mL of distilled water. We shook the jar at 140 rpm for 10 min at 24°C and sonicated for 5 min to desorb PM from the leaf surfaces. We measured leaf area to quantify the PM mass per unit leaf area after we removed the leaves from the glass jar. We used a leaf-area meter (Li-3100, Li-COR, USA) to measure leaf area, which we then doubled to include both leaf sides (Saebø et al. 2012).
2.6. Statistical analysis

We statistically analyzed the data using freeware R, and we tested significant differences between the mean values using the Bonferroni correction method after the Kruskal–Wallis test, Dunn’s test after the Kruskal–Wallis rank sum test, paired t-test, or Spearman’s rank correlation.

3. Results

Table 3 lists the mass of PM10 on the leaf surface per unit leaf area (termed “surface PM10”) of the four study species. The mean masses of surface PM10 on Z. serrata (58.3 ± 39.5 mg/m²) and S. prunifolia f. simpliciflora (68.1 ± 34.7 mg/m²) were significantly higher than those on M. glyptostroboides (43.5 ± 35.8 mg/m²) and P. yedoensis (28.2 ± 20.3 mg/m²). We found no significant correlation between the mean mass of PM10 and leaf traits of the four study species (data not shown).

Species with a high accumulation of surface PM10 changed with the sampling day. The mass of surface PM10 on Z. serrata was significantly higher on May 20, May 30, and September 2 (Figure 3(a)). The mass of surface PM10 on S. prunifolia f. simpliciflora was significantly higher on June 13, June 27, August 20, September 17, and October 10, and significantly lower on May 20 (Figure 3(a)).

Table 4 shows the mean mass of PM2.5 on the leaf surface per unit leaf area (termed “surface PM2.5”) of the four study species. The mean masses of surface PM2.5 on Z. serrata (11.6 ± 7.1 mg/m²) and S. prunifolia f. simpliciflora (18.2 ± 12.9 mg/m²) were significantly higher than those on M. glyptostroboides (7.8 ± 6.6 mg/m²) and P. yedoensis (6.5 ± 4.4 mg/m²). We found no significant correlation between the mean mass of PM2.5 and leaf traits of the four study species (data not shown).

The mass of the surface PM2.5 on Z. serrata was significantly higher on May 20 and September 2 (Figure 3(b)). The mass of surface PM2.5 on S. prunifolia f. simpliciflora was significantly higher on May August.

Table 3. The mass of surface particulate matter with a diameter less than 10 μm (PM10) accumulated on leaves in 2019.

| Sampling date | Metasequoia glyptostroboides | Prunus yedoensis | Zelkova serrata | Spirea prunifolia f. simpliciflora |
|---------------|------------------------------|-----------------|----------------|----------------------------------|
| May 7         | 119.5 ± 64.6 a A             | 78.9 ± 39.1 a A | 82.0 ± 37.5 a A | 78.7 ± 29.1 a A                  |
| May 20        | 28.6 ± 15.2 ab AB            | 21.1 ± 4.7 ab AB| 60.2 ± 25.3 a A| 19.2 ± 13.9 b A                  |
| May 30        | 42.7 ± 32.4 ab AB            | 27.9 ± 8.0 b AB | 97.8 ± 35.0 a A| 58.4 ± 17.7 ab A                  |
| Jun 13        | 36.2 ± 20.1 ab AB            | 21.3 ± 4.1 b AB | 56.6 ± 18.6 ab A| 84.9 ± 24.4 a A                  |
| Jun 27        | 54.4 ± 41.6 ab AB            | 35.8 ± 18.0 b AB| 66.6 ± 19.8 ab A| 105.2 ± 54.2 a A                 |
| Jul 15        | 40.9 ± 15.4 a AB            | 31.5 ± 5.7 a AB | 31.8 ± 6.2 a A  | 60.9 ± 20.2 a A                  |
| Jul 17        | 30.4 ± 12.2 b AB            | 22.8 ± 8.6 a AB | 30.7 ± 7.4 a A  | 44.0 ± 6.8 a A                   |
| Jul 29        | 22.6 ± 11.0 a AB            | 20.5 ± 6.5 a AB | 39.2 ± 25.6 a A| 61.7 ± 16.2 a A                  |
| Aug 20        | 30.6 ± 18.1 ab AB            | 25.7 ± 5.7 b AB | 48.5 ± 28.2 a B| 120.9 ± 14.8 a A                 |
| Sep 2         | 34.2 ± 9.9 b AB             | 35.7 ± 8.0 a AB | 101.2 ± 61.4 a A| 46.1 ± 14.7 a B                  |
| Sep 17        | 18.1 ± 7.6 b B              | 10.6 ± 3.0 b B  | 29.2 ± 19.3 a B| 53.4 ± 13.0 a A                  |
| Oct 10        | 26.8 ± 10.2 ab AB           | 15.6 ± 5.9 b B  | 72.1 ± 74.9 a B| 73.3 ± 41.9 a A                  |
| Oct 23        | 80.1 ± 15.5 a A             | 19.3 ± 5.9 b AB | 42.5 ± 24.8 a B| 78.3 ± 33.8 a A                  |
| Mean          | 43.5 ± 35.8 b 6             | 28.2 ± 20.3 c   | 58.3 ± 39.5 a   | 68.1 ± 34.7 a                     |

n = 4 (S. prunifolia f. simpliciflora), n = 5 (all others), mean ± SD. Different small letters in the same row and different capital letters in the same column indicate significant differences at p < 0.05, according to the Bonferroni correction method after the Kruskal-Wallis test.

Figure 3. The mass of surface particulate matter (PM) per unit leaf area of four tree species. (a) PM10, with a diameter less than 10 μm, (b) PM2.5, with a diameter less than 2.5 μm.
Table 4. The mass of surface particulate matter with a diameter less than 2.5 μm (PM$_{2.5}$) accumulated on leaves in 2019.

| Sampling date | Metasequoia glyptostroboides | Prunus yedoensis | Zelkova serrata | Spirea prunifolia f. simpliciflora |
|---------------|-----------------------------|-----------------|----------------|----------------------------------|
| May 7         | 22.6 ± 11.2 a A             | 14.7 ± 7.2 a A  | 13.7 ± 2.3 a A | 19.6 ± 7.9 a A                  |
| May 20        | 2.0 ± 1.4 b B               | 3.9 ± 1.3 ab AB | 12.8 ± 5.8 a A | 3.0 ± 3.2 b A                   |
| May 30        | 9.0 ± 5.3 ab AB             | 7.1 ± 2.0 b AB  | 18.3 ± 7.6 a A | 23.6 ± 6.8 a A                  |
| Jun 13        | 12.7 ± 3.9 ab AB            | 7.7 ± 1.4 b AB  | 18.1 ± 3.4 a A | 34.6 ± 3.9 a A                  |
| Jun 27        | 10.8 ± 6 ab AB              | 8.0 ± 3.2 b AB  | 12.1 ± 3.8 a AB| 30.7 ± 13.0 a A                 |
| Jul 15        | 6.0 ± 1.2 a a               | 3.8 ± 1.7 a AB  | 5.4 ± 0.8 a A  | 13.4 ± 7.9 a A                  |
| Jul 17        | 3.2 ± 1.9 a a               | 4.1 ± 2.4 a AB  | 4.4 ± 1.5 a A  | 6.6 ± 4.1 a A                   |
| Jul 29        | 5.5 ± 3.4 a AB              | 4.3 ± 1.2 a AB  | 5.7 ± 2.7 a A  | 15.2 ± 3.3 a A                  |
| Aug 20        | 7.8 ± 4.6 b AB              | 9.9 ± 1.6 a AB  | 14.7 ± 10.1 a A| 43.4 ± 6.2 a A                  |
| Sep 2         | 5.5 ± 1.6 b AB              | 11.1 ± 3.3 a A  | 18.1 ± 8.2 a A | 4.4 ± 2.6 b A                   |
| Sep 17        | 6.5 ± 2.8 ab AB             | 2.8 ± 0.6 b B   | 9.2 ± 4.4 a A  | 11.8 ± 2.2 a A                  |
| Oct 10        | 7.3 ± 1.9 ab AB             | 5.2 ± 2.2 b AB  | 12.0 ± 9.0 a B | 18.2 ± 2.9 a A                  |
| Oct 23        | 2.9 ± 2.1 a B               | 2.1 ± 2.5 b AB  | 5.9 ± 2.8 a B  | 11.6 ± 4.7 a A                  |
| Mean          | 7.8 ± 6.6 b                 | 6.5 ± 4.4 b     | 11.6 ± 7.1 a  | 18.2 ± 12.9 a                   |

n = 4 (S. prunifolia f. simpliciflora), n = 5 (all others), mean ± SD.

Different small letters in the same row and different capital letters in the same column indicate significant differences at p < 0.05, according to the Bonferroni correction method after the Kruskal-Wallis test.

Table 5. The mean ratio of particulate matter with a diameter less than 2.5 μm to particulate matter with a diameter less than 10 μm for each species during the study period.

|                     | Metasequoia glyptostroboides | Prunus yedoensis | Zelkova serrata | Spirea prunifolia f. simpliciflora |
|---------------------|------------------------------|-----------------|----------------|----------------------------------|
| Mean Ratio          | 0.21 ± 0.12 a                | 0.24 ± 0.10 b   | 0.21 ± 0.08 a  | 0.26 ± 0.12 b                    |

n = 4 (S. prunifolia f. simpliciflora), n = 5 (all others), mean ± SD.

Different letters indicate significant differences at p < 0.05, according to Dunn’s test after the Kruskal-Wallis rank sum test.

The mean ratios of surface PM$_{2.5}$ to PM$_{10}$ in P. yedoensis and S. prunifolia f. simpliciflora were significantly higher than those of M. glyptostroboides and Z. serrata (Table 5). The mean ratio of surface PM$_{2.5}$ to PM$_{10}$ also changed throughout the experimental period (Figure 4). However, the pattern of surface PM ratio fluctuations did not coincide with that of the mean ratio of atmospheric PM$_{2.5}$ to PM$_{10}$ (Figure 1(c)). We observed no correlation between the ratio of surface PM$_{2.5}$ to PM$_{10}$ and the ratio of atmospheric PM$_{2.5}$ to PM$_{10}$ in Z. serrata and S. prunifolia f. simpliciflora (Spearman’s rank correlation, data not shown). For M. glyptostroboides and P. yedoensis, we found a correlation between the ratio of surface PM$_{2.5}$ to PM$_{10}$ and the ratio of atmospheric PM$_{2.5}$ to PM$_{10}$, but the correlation coefficient was very weak (−0.25 for M. glyptostroboides and −0.31 for P. yedoensis).

The mean masses of surface PM$_{10}$ on P. yedoensis before and after rainfall were 31.5 ± 5.7 mg/m$^2$ (on July 15) and 22.8 ± 8.6 mg/m$^2$ (on July 17), respectively, indicating that 29% of the PM$_{10}$ on the leaves was significantly removed by the rainfall event (Table 6).

The mean masses of surface PM$_{2.5}$ on M. glyptostroboides before and after rainfall were 6.0 ± 1.2 mg/m$^2$ (at July 15) and 3.2 ± 1.9 mg/m$^2$ (at July 17), respectively, meaning that 45% of PM$_{2.5}$ on the leaves was significantly washed away by the rainfall event. These were the only cases in which the mass of surface PM was significantly reduced by rainfall. Overall, the surface PM masses of the four study species were not significantly affected by rainfall.

4. Discussion

In this study, the decreasing order of mean mass of surface PM$_{10}$ that accumulated on the leaves of the
four considered tree species was *S. prunifolia* f. *simpliciflora* = *Z. serrata* > *M. glyptostroboides* > *P. yedoensis*. The mean mass of surface PM$_{2.5}$ that accumulated on the leaves of the four tree species was *S. prunifolia* f. *simpliciflora* = *Z. serrata* > *M. glyptostroboides* = *P. yedoensis*. The PM adsorption ability of trees is related to their leaf surface morphological characteristics, such as leaf size, surface roughness, trichome length and density, wax composition, contact angle, and stomatal size and density (Beckett et al. 2000; Räsänen et al. 2013; Wang et al. 2013; Leonard et al. 2016; Zhang et al. 2020; Jin et al. 2021). Increased PM accumulation on leaf surfaces is related to increased hair density, length, and leaf roughness, and decreased leaf size (Beckett et al. 2000; Liu et al. 2012; Saebø et al. 2012; Weerakkody et al. 2017; Zhang et al. 2017; Jin et al. 2021). The leaves of *S. prunifolia* f. *simpliciflora* and *Z. serrata* reportedly have trichomes on their adaxial leaf surface (Je et al. 2020; Son et al. 2021) (Table 2). In contrast, trichomes we observed no on the adaxial leaf surfaces of *M. glyptostroboides* and *P. yedoensis*. The high PM accumulation abilities of *S. prunifolia* f. *simpliciflora* and *Z. serrata* leaves that we observed in this study might be related to the presence of trichomes on the adaxial leaf surfaces of these species. However, the longer length and higher density of trichomes on the adaxial leaf surfaces of *Z. serrata* did not result in higher PM accumulation on the leaf surface compared with *S. prunifolia* f. *simpliciflora* (Table 2). The leaf area and roughness of *S. prunifolia* f. *simpliciflora*, *Z. serrata*, *M. glyptostroboides*, and *P. yedoensis* were 6.4 cm$^2$ and 1.4 mm, 28.9 cm$^2$ and 0.9 mm, 18.9 cm$^2$ and 3.9 mm, and 45.9 cm$^2$ and 3.8 mm, respectively (Je et al. 2020; Son et al. 2021) (Table 2). We found no significant correlation between the PM accumulation ability on the leaf and leaf area or leaf roughness of the four tree species in this study. This result coincides with the prior findings of no significant correlations between PM retention ability on the leaf surface and leaf size, leaf surface roughness, or trichome density (Saebø et al. 2012; Kwak et al. 2020; Su et al. 2021). The results are conflicting regarding the factors that determine the amount of PM adsorbed by the leaves (Corada et al., 2021). Because of the variations in leaf characteristics and the complex relationships between leaf characteristics, determining the factors affecting the PM accumulation ability of the leaves is difficult. Therefore, it is necessary to find the factors using a simple model, such as experiments using the front and back sides of leaves with different micromorphologies of the same tree, using leaves of different sizes of the same tree, and so on.

Among the four study species, *S. prunifolia* f. *simpliciflora* and *P. yedoensis* were the most efficient at capturing PM$_{2.5}$, which is known to induce respiratory diseases in humans (Kampa and Castanas 2008). *S. prunifolia* f. *simpliciflora* showed the strongest PM$_{10}$ and PM$_{2.5}$ accumulation ability, indicating that this species had the best ability to improve the atmospheric PM quality among the four study species. Moreover, *S. prunifolia* f. *simpliciflora* is a shrub with an activity range close to human breathing height. Therefore, *S. prunifolia* f. *simpliciflora* may be the most effective species for reducing atmospheric PM during one growing season among the four species we considered in this study. The mean ratio of surface PM$_{2.5}$ to PM$_{10}$ of the four study species ranged from 0.21 to 0.26 in this study. In contrast, the mean ratio of atmospheric PM$_{2.5}$ to PM$_{10}$ was 0.54, which was higher than the leaf surface PM ratio. Song et al. (2015) also showed that the ratio of average PM$_{2.5}$ to PM$_{10}$ on the leaves was 0.10, so further studies are required to determine why leaves adsorb more PM$_{10}$ than PM$_{2.5}$. The pattern of surface PM ratio changed among the sampling dates, but did not coincide with the pattern of atmospheric PM ratio. The surface PM ratio of *S. prunifolia* f. *simpliciflora* was the highest on June 13 and the lowest on September 2. From this result, we found that the surface PM ratio widely fluctuated during the growing season. Therefore, to reduce atmospheric PM$_{2.5}$ effectively, various tree species must be planted.

Rainfall intensity affects the surface PM wash-off mass and rate of *Pinus tabuliformis* (Xu et al. 2019). This species showed the lowest wash-off rate among the six study species, reaching 45% of coarse and fine PM under a 30 mm/h rainfall intensity (Xu et al. 2017). Weerakkody et al. (2018) concluded that a 16 mm/h intensity rainfall washed surface PM of four evergreen species and that the interspecies variations were probably owing to micromorphologies such as smooth or waxy surfaces. Our results showed significant surface PM wash-off at a 28 mm/h intensity rainfall in two cases: PM$_{10}$ on the leaves of *P. yedoensis* and PM$_{2.5}$ on *M. glyptostroboides*. Despite the

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**Table 6.** The mass of particulate matter (PM) with a diameter less than 10 μm (PM$_{10}$) and a diameter less than 2.5 μm (PM$_{2.5}$) accumulated on leaves before (on 15 July 2019) and after (on 17 July 2019) rainfall event.

| Type of PM | Sampling date | Metasequoia glyptostroboides | Prunus yedoensis | Zelkova serrata | Spirea prunifolia f. simpliciflora |
|-----------|---------------|------------------------------|-----------------|----------------|---------------------------------|
| PM$_{10}$ | Before rainfall | 40.9 ± 15.4                  | 31.5 ± 5.7$^a$  | 31.8 ± 6.2     | 60.9 ± 20.2                     |
|           | After rainfall | 30.4 ± 12.2                  | 22.8 ± 6.0$^b$  | 30.7 ± 7.4     | 44.0 ± 6.8                      |
| PM$_{2.5}$| Before rainfall| 6.0 ± 1.2$^a$                | 3.8 ± 1.7        | 5.4 ± 0.8      | 13.4 ± 12.9                     |
|           | After rainfall| 3.2 ± 1.9$^b$                | 4.1 ± 2.4        | 4.4 ± 1.5      | 6.6 ± 4.1                       |
|           | Mean reduction rate (%) | 20 ± 38                   | 29 ± 17          | 1 ± 27         | 21 ± 26                         |

$n = 4$ (S. prunifolia f. simpliciflora), $n = 5$ (all others), mean ± SD. Mean reduction rate is mean of five PM reduction rates of individual trees before and after rainfall event. Different letters indicate significant differences at $p < 0.05$ according to Paired t-test.
sufficiently strong rainfall intensity (28 mm/h) to wash off surface PM in our study, we found no significant surface PM washed off in the other cases, such as PM$_{2.5}$ on the leaves of P. yedoensis or PM$_{10}$ on Z. serrata. This might have occurred because the high concentration of atmospheric PM and the position of the leaves were under the crown, blocking contact with the raindrops and increasing the wet deposition of the lower tree leaves, as mentioned by Wang et al. (2015). This means that the PM wash-off rates we estimated in this study may be low, indicating that a study of PM wash-off by rainfall throughout an entire tree crown is needed.

In conclusion, the PM-reducing ability of tree leaves varies among species and over time. Reduced PM from tree leaves owing to rainfall also varied among species and PM diameter in this study. The presence or absence of trichomes on the leaf surface was the only factor that explained the difference in PM adsorption between tree species. The change in the amount of PM adsorbed by S. prunifolia f. simpliciflora over time showed no correlation with that of the other three species. However, we think this was owing to the discrepancy between the microclimate around the shrub species of S. prunifolia f. simpliciflora and the other three tall tree species. To understand why the amount of PM accumulated on the leaves of one species changes during the growing season, we need to study the microclimate near the investigated individual trees and seasonal leaf characteristic changes. Because the period of highest PM adsorption and PM$_{2.5}$ adsorption ratio differed for each tree species, the diversity of tree species in urban forests should be high to effectively reduce atmospheric PM concentrations throughout the year.

**Disclosure statement**

No potential conflict of interest was reported by the author(s).

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