

Original Research

Heavy Metals Concentrations in Deposited Dust of Typical Chinese Tree Species in Different Functional Areas in Nanjing

Jie Tang^{1,2*}, Yan Zha³, Yan Cheng⁴

¹School of Resources and Environment, Anhui Agricultural University, Hefei, Anhui, China

²Hefei Scientific Observing and Experimental Station of Agro-Environment,
Ministry of Agriculture, P.R. China

³Co-Innovation Center for Sustainable Forestry in Southern China, Jiangsu Province Key Laboratory
of Ecological Engineering, Nanjing Forestry University, Nanjing, China

⁴Hefei Environmental Monitoring Central Station, Hefei, Anhui, China

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Abstract

Foliar dust passively adsorbs anthropogenic heavy metals (HM) present in the atmosphere and thus reduces the total suspended particle (TSP) level. Urban plants have been shown to reduce the atmospheric level of ambient particulate matter (PM) via foliar dust adsorption. We studied heavy metal concentrations in the foliar dust of three typical tree species in five functional areas of Nanjing city. The highest levels of Cd (19.89 ± 4.56 mg/kg), Pb (167.33 ± 16.61 mg/kg) and Cr (197.42 ± 13.96 mg/kg) were found in the Traffic Area (TA), whereas the highest levels of Cu (309.27 ± 25.79 mg/kg) and Zn (1036.88 ± 52.77 mg/kg) were found in the Industrial Area (IA). Significant differences were found between tree species. The amount of PM per unit leaf area generally decreased in this order: *Cedrus deodara* > *Pittosporum tobira* > *Cinnamomum camphora*. The highest mass percentages of large, coarse and fine PM were captured by *C. camphora*, *P. tobira* and *C. deodara*, respectively. A scanning electron microscope (SEM) was used to investigate the surfaces of the leaves, as well as the density and size of the stomata of each species. Our results suggest that an oily and coarse leaf surface is the most important factor facilitating PM accumulation, but large high-density stomata also enhance PM adsorption and thus favor HM accumulation in foliar dust. This study shows that the HM concentrations in foliar dust can act as an indicator of air pollution.

Keywords: heavy metals, foliar dust, air pollution, urban plants

*e-mail: ahnydx1989@163.com

Introduction

The unprecedented economic development in China in recent years has been accompanied by unprecedented environmental changes. Particulate matter (PM) in the atmosphere can be utilized to characterize the environmental quality of urban areas [1-3]. The capacity of dust particles to carry pollutants depends largely on their particulate composition in terms of minerals and organic matter [4]. Heavy metals in dust can persist in urban environments, or they may be resuspended in the atmosphere and thus pose a threat to public health and ecosystems [5]. The sources of heavy metal pollutants include domestic waste, the chemical industry and transportation [6]. Most previous studies of heavy metal pollution have determined the concentrations, distributions and sources of heavy metals in roadside dust [7-9]. However, the presence of heavy metals in foliar dust is of greater concern because of the frequent hand-to-mouth activities of children, which increase their exposure to pollutants in dust. In addition, the height of roadside shrubs on which dust is deposited is often similar to that of children (0.5-1.5 m), and this similarity in height could be an important contributor to dust intake by children via direct inhalation [10].

Leaves are highly exposed to air pollution and purify air by absorbing atmospheric pollutants [11-12] such as trace elements, salts and pollens [13]. The leaves of tree species *Robinia pseudoacacia* and *Taxus baccata* have been used as indicators to assess the quality of air in urban environments [14-17]. The efficacy of removing pollutants from the air using plants depends on the particular type of tree canopy, branch density and leaf

morphology [2, 11, 18]. Some species are more effective accumulators of PM because of the microstructure of their leaves [19]. For example, the needles of coniferous species produce a relatively thick epicuticular wax layer that favors PM accumulation. In addition, broadleaf species with rough leaf surfaces capture PM better than those with smooth leaf surfaces [11]. Knowledge of the factors that favor or inhibit accumulation of PM, including heavy metals, from the air on foliage is important for policymakers, farmers and citizens with the responsibility of protecting human health and the environment.

The aim of this study was to determine the concentration of heavy metals (Cd, Cr, Cu, Pb and Zn) in foliar dust and to measure the size of PM on the foliar surface. Scanning electronic microscopy (SEM) analysis was utilized to gain a better understanding of the role of trees in capturing atmospheric pollutants, and heavy metals in particular, in an area of Nanjing that has experienced rapid urbanization and industrialization in the last decade [20-21].

Material and Methods

Sampling and Sample Preparation

Three tree species that are widely planted in Nanjing, *Cedrus deodara*, *Cinnamomum camphora* and *Pittosporum tobira*, were selected for this study. Thirty foliar dust samples were collected from different functional areas in a highly urbanized region of Nanjing during August 2017. Samples were collected from five

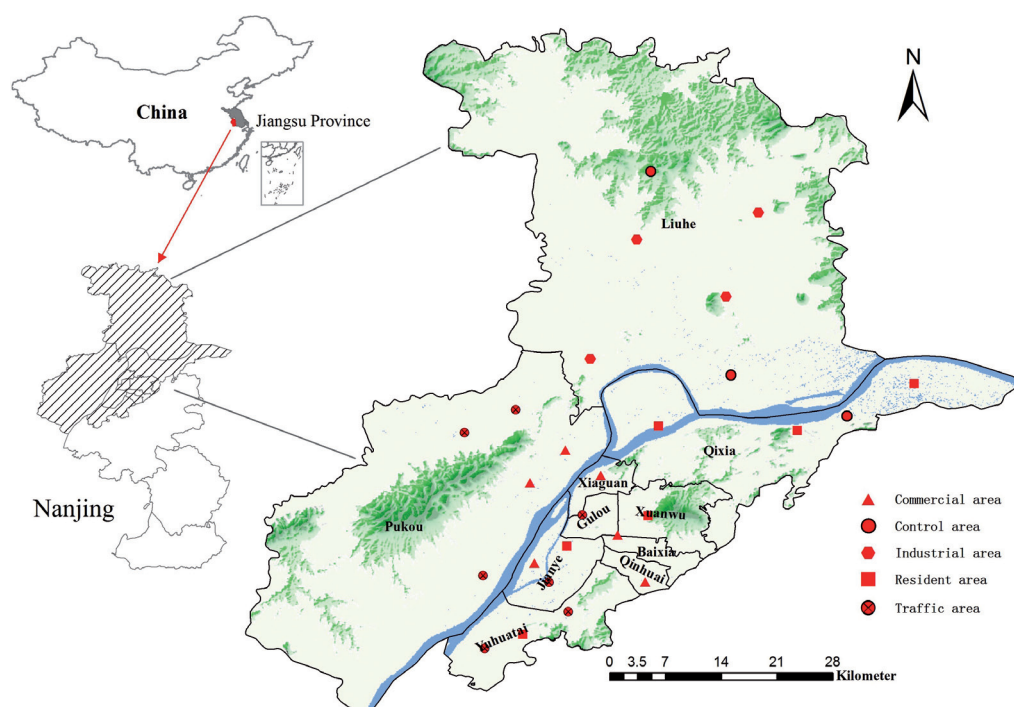


Fig. 1. Study area and sampling sites.

types of urban area that have been identified as being characteristic of urban Nanjing: industrial areas (IA), residential areas (RA), commercial areas (CMA), traffic areas (TA) and control areas (CA). The locations of the selected urban areas are shown in Fig. 1. All samples were collected after at least 28 days had passed with no precipitation, because Liu et al. [22] showed that this practice led to the collection of leaves that had attained the maximum dust-retaining capability. Every sample consisted of approximately 800 g of leaves from mature and healthy trees collected at a height of 0.5-3 m above the ground. Eight individuals of each species were sampled. All leaf samples were collected from a single tree and stored at 4°C in a clean laboratory refrigerator prior to analysis. PM collected from the surface of the leaves was analyzed using the methods of Dzierżanowski et al [23].

Heavy Metal Determination

Concentrations of heavy metals (Cd, Cr, Cu, Pb and Zn) were determined using inductively coupled plasma mass spectrometry (ICP-MS). Before the assay, the dust samples were prepared as follows [24]. The leaves were washed using deionized water and placed into a 500-mL plastic container with 250 mL of Milli-Q water (Millipore, Bedford, MA, USA). The dust-containing suspension was filtered with 50 mL Milli-Q water, and 250 mL of the suspension was dried with a vacuum freeze-drier (Labconco, Kansas City, MO, USA) for three days at -83°C and stored at -20°C until further extraction was performed. Approximately 0.5 g of the prepared dust sample was dried indoors at room temperature, and impurities such as tree leaves and stones were removed. The samples were ground to pass through a 0.15-mm nylon sieve and mineralized on a hot plate in HF-HNO₃-HClO₄ (V:V:V=1:1:1), which was subsequently evaporated to dryness. The sample residue was dissolved in HCl, after which the concentrations of trace metals in the resulting solution were determined by ICP-MS (Perkin Elmer Sciex DRCII, USA). The samples were analyzed in triplicate with standard reference materials and blanks. Before each test, all glassware was submerged in 0.1 M HNO₃ for 48 h, rinsed thoroughly with deionized water, and dried. All chemicals were of analytical grade. Reference samples obtained from the National Research Center for Certified Reference Materials (Beijing, China) were analyzed in parallel with each sample. The recoveries were between 84.72 and 107.25%.

Quantitative Analysis of PM

The filters used for the analysis were first dried for 30 minutes at 60°C in a drying chamber (YB881-4, Suzhou, China). A balance and a humidity controller (WHD48-11, ACREL Co., Ltd., Jiangsu, China) were placed in the balancing box. To avoid electrostatic charges on the filters, they were first placed in a

polytetrafluoroethylene (PTFE) balancing box under constant temperature (25°C) and constant humidity (40%) for 48 hours. Every sample of leaves was placed in a glass container with 250 mL of water and agitated for 60 seconds in order to wash off particles from the leaf surface. The three fractions of particulate matter were collected on filters: (1) large: >10 µm, (2) coarse: 2.5-10 µm and (3) fine: 0.2-2.5 µm. Samples were stored in paper bags in a dryer (DHG-9030A, Shanghai Jinghong Scientific Instrument Co., Ltd., Shanghai, China) at 60°C for 48 hours. Filters were weighed before and after particulate removal and storage using a balance sensitive (Sartorius Beijing Scientific Instrument Co., Ltd., Beijing, China) to 0.00001 g in order to calculate the mass of PM in each fraction of every sample.

Scanning Electron Microscopy of the Leaves

Scanning electron microscopy (FEI Quanta-200, USA) was used to investigate the surfaces of the leaves, as well as all the density and size of the stomata of each species. Small strips (approximately 1 cm²) were cut from the leaves and dehydrated in ethanol solutions (30%, 50%, 70%, 90% and 100%) for 15 min each. The samples were gold-coated to enhance their electrical conductivity before analysis. Photographs were taken of randomly chosen spots at 1000× and 2000× magnification at 5 kV. Particle counting was performed using ImageJ software (National Institutes of Health, USA), according to the method described by Ottelé et al. [25].

Statistical Analysis

One-way analysis of variance (ANOVA) ($p < 0.05$) was applied to detect significantly different amounts of dust on leaf surfaces and significantly different concentrations of heavy metals in foliar dust. The LSD multiple comparison post hoc test was used to explore the significant differences identified by the ANOVA. Calculations were performed using SPSS version 17.0 (IBM Co., Armonk, NY, USA). Graphs were generated using Excel 2012 and Origin 9.0.

Results and Discussion

Heavy Metal Concentrations in Foliar Dust in Urban Areas of Nanjing

The heavy metal concentrations in the foliar dust from five different functional areas in Nanjing differed significantly (Table 1). The concentrations of the five determined elements in the foliar dust decreased in this order: TA>IA>CMA>RA>CA. The highest average levels of Cd, Pb and Cr were found in the TA and greatly exceed those in the foliar dust collected from other areas; this finding was attributed to vehicle emissions. The highest levels of Cu and Zn were found

Table 1. Concentrations of air contaminants (mean \pm SE, mg/kg) in foliar dust of tree species according to the studied areas.

Elements	Species	Site					
		IA	RA	CMA	TA	CA	Average
Cd	<i>Cedrus deodara</i>	17.35 \pm 3.26Aa	11.22 \pm 2.01Db	13.91 \pm 3.14Cb	15.55 \pm 1.43Bc	6.26 \pm 1.03Ea	12.86 \pm 2.71b
	<i>Cinnamomum camphora</i>	14.81 \pm 2.87Db	16.53 \pm 1.83Ca	19.14 \pm 0.35Ba	24.74 \pm 2.01Aa	4.87 \pm 0.46Eb	16.02 \pm 1.50a
	<i>Pittosporum tobira</i>	8.41 \pm 1.78Cc	8.89 \pm 1.24Cc	12.56 \pm 2.25Bb	19.37 \pm 4.23Ab	5.15 \pm 0.94Da	10.88 \pm 2.09c
	Average	13.52 \pm 2.64c	12.21 \pm 1.69c	15.20 \pm 3.91b	19.89 \pm 4.56a	5.43 \pm 0.81d	13.25 \pm 1.92D
Pb	<i>Cedrus deodara</i>	155.62 \pm 17.98Ca	176.25 \pm 17.83Ba	144.57 \pm 12.42Ca	209.02 \pm 21.53Aa	95.67 \pm 11.47Da	156.23 \pm 16.25a
	<i>Cinnamomum camphora</i>	109.75 \pm 12.21Cc	120.32 \pm 12.42Bb	135.81 \pm 10.03Aa	136.72 \pm 15.74Ab	57.95 \pm 7.42Db	112.11 \pm 11.56b
	<i>Pittosporum tobira</i>	123.56 \pm 15.66Bb	133.67 \pm 14.47Bb	103.61 \pm 8.74Cb	156.25 \pm 12.57Ac	60.26 \pm 9.62Db	115.47 \pm 12.21b
	Average	129.64 \pm 15.28c	143.41 \pm 14.91b	127.99 \pm 10.40c	167.33 \pm 16.61a	71.29 \pm 9.50d	127.93 \pm 13.34C
Cu	<i>Cedrus deodara</i>	289.46 \pm 24.47Ab	145.67 \pm 12.31Cb	233.93 \pm 18.46Ba	276.77 \pm 17.62Aa	101.47 \pm 11.03Da	209.46 \pm 18.54a
	<i>Cinnamomum camphora</i>	276.87 \pm 21.46Ab	114.78 \pm 10.93Dc	162.82 \pm 12.93Cc	234.94 \pm 15.58Bb	65.89 \pm 8.62Ec	171.06 \pm 14.12b
	<i>Pittosporum tobira</i>	361.48 \pm 31.45Aa	176.83 \pm 14.14Ca	200.57 \pm 13.78Bb	185.71 \pm 15.75Cc	87.58 \pm 9.72Db	202.43 \pm 14.99a
	Average	309.27 \pm 25.79a	145.76 \pm 12.46d	199.11 \pm 15.06c	232.47 \pm 16.32b	84.98 \pm 9.79e	194.32 \pm 15.88B
Zn	<i>Cedrus deodara</i>	1313.25 \pm 65.71Aa	525.62 \pm 27.76Da	1213.67 \pm 58.62Ba	732.57 \pm 41.67Ca	211.57 \pm 12.64Ea	799.34 \pm 41.28a
	<i>Cinnamomum camphora</i>	814.66 \pm 43.56Ac	498.04 \pm 17.66Db	736.26 \pm 40.09Bc	629.68 \pm 36.71Cb	97.41 \pm 10.08Ec	555.21 \pm 29.37c
	<i>Pittosporum tobira</i>	982.72 \pm 49.04Ab	357.83 \pm 20.41Cc	984.67 \pm 44.61Ab	587.15 \pm 32.72Bc	167.37 \pm 7.13Db	615.95 \pm 31.03b
	Average	1036.88 \pm 52.77a	460.50 \pm 21.94d	978.20 \pm 47.77b	649.80 \pm 37.03c	158.78 \pm 9.95e	656.83 \pm 33.89A
Cr	<i>Cedrus deodara</i>	162.47 \pm 14.56Ba	98.49 \pm 13.56Da	121.57 \pm 14.45Ca	229.04 \pm 16.64Aa	64.31 \pm 11.67Ea	135.18 \pm 14.18a
	<i>Cinnamomum camphora</i>	123.51 \pm 10.42Bc	77.63 \pm 11.68Db	89.82 \pm 6.71Cc	195.79 \pm 13.78Ab	39.26 \pm 8.94Eb	105.20 \pm 9.43b
	<i>Pittosporum tobira</i>	145.68 \pm 12.58Bb	56.84 \pm 9.62Dc	104.39 \pm 10.98Cb	167.42 \pm 11.47Ac	56.72 \pm 10.82Da	106.21 \pm 11.97b
	Average	143.89 \pm 12.52b	77.65 \pm 11.62d	105.26 \pm 10.71c	197.42 \pm 13.96a	53.43 \pm 10.48e	115.53 \pm 11.86C

*IA = Industrial Area, RA = Resident Area, CMA = Commercial Area, TA = Traffic Area, CA = Control Area.

* Values are means \pm SE (n = 30).

*a-d. Medians are significantly different for each metal in rows (among the different functional areas).

*A-B. Medians are significantly different for each metal in columns (among different tree species).

in the IA and greatly exceed those measured in the other functional areas; this finding was attributed to the incomplete combustion of petroleum and coal release in the IA. The Cd concentrations in the IA, RA, CMA, TA and CA were 237.19, 214.21, 266.67, 348.95 and 95.26 times higher, respectively, than the background soil concentration in Jiangsu Province, in which Nanjing is located (CNEMC, 1990). In contrast, the Pb concentrations in the IA, RA, CMA, TA and CA were 5.23, 5.78, 5.16, 6.75 and 2.87 times higher, respectively, than the background soil concentration in Jiangsu. It is worth noting that the heavy metal concentrations in Chinese cities vary greatly. For example, the average concentration of Pb in the TA and RA were higher than the average Pb concentration (136.6 mg/kg) in Shanghai [8], but lower than that in Huizhou (486.0 mg/kg) [7]. The Cu concentrations in the IA, RA, CMA, TA and CA were 9.60, 4.53, 6.18, 7.22 and 2.64 times higher, respectively, than the background soil concentrations in Jiangsu. The Zn concentrations in the IA, RA, CMA, TA and CA were 13.5, 6.00, 12.74, 8.46 and 2.07 times higher, respectively, than the background soil concentrations in Jiangsu. The average concentrations of Cu and Zn measured in our study greatly exceeded those reported in Debrecen, Hungary in 2011 [11]. The Cr concentrations in the IA, RA, CMA, TA and CA were 2.44, 1.32, 1.78, 3.35 and 0.91 times higher, respectively, than the background soil concentration in Jiangsu. The Cr concentrations in the selected areas were lower than those in Huizhou, an urban area of Hangzhou, China [7].

Differences in Foliar Dust Heavy Metal Concentrations Among Species

The concentrations of five elements in the foliar dust of three tree species were determined in the TA, IA, CMA, RA and CA (Table 1). In the case of Cd, the concentration in the foliar dust of *C. deodara* from the IA and CA was significant higher than that in the foliar dust of *C. camphora* or *P. tobira* from the IA and CA. The Cd concentration in the foliar dust of *P. tobira* from the RA and TA was significantly higher than that in the foliar dust of *C. deodara* or *C. camphora* from the RA and TA, which indicated that Cd was more easily captured by the leaves of *P. tobira* and *C. deodara* in comparison with those of *C. camphora*. The concentration of Pb in the foliar dust of *C. deodara* from the CMA was significantly higher than that in the foliar dust of *C. camphora* or *P. tobira* from all of the studied functional areas, with the exception of the CMA, which indicated that *C. deodara* was particularly capable as a remover of lead pollution from the atmosphere. The Cu concentration in the foliar dust of *P. tobira* from the IA and RA was significantly higher than that from the other tree species from the IA and RA, but the Cu concentration in the foliar dust of *C. deodara* was significantly higher than that in the foliar dust from the other tree species from the CMA, TA and CMA.

The Zn and Cr concentrations in the foliar dust of *C. deodara* were significantly higher than that from the other tree species in all areas. The mean concentration of Cd in the foliar dust of *P. tobira* was significantly higher than that of the foliar dust of *C. deodara* and *C. camphora* in all five functional areas. The Pb and Cr concentrations in the foliar dust of *C. deodara* was significantly higher than that in the foliar dust of *P. tobira* or *C. camphora* in all five functional areas, but there was no significant difference between *P. tobira* and *C. camphora*. The Cu concentrations in the foliar dust of *C. deodara* and *P. tobira* were significantly higher than that in the foliar dust of *C. camphora* in all five functional areas. The Zn concentration in the foliar dust of *C. deodara* was significantly higher than that in the foliar dust of *P. tobira* or *C. camphora* in all five functional areas, and the Zn concentration in the foliar dust of *P. tobira* was significantly higher than that in the foliar dust of *C. camphora* in all five functional areas. Our result indicated that the differences in heavy metal concentrations in foliar dust between species was significant, which is consistent with other conclusions [26, 27]. The important traits of leaf micro morphology were the key factor that can influence air contaminant concentrations. Simon demonstrated that leaf micromorphology was important in the dust deposition, and which was further affected by high concentration of air contaminants [26]. TOMAŠEVIĆ and ANIĆIĆ also observed that most of the fine particles were deposited near the stomata, around the over, by which the physiological characteristics of leaves can be affected [27]. In our study we found that *C. camphora* was the most effective at capturing particulate matter and accumulating high heavy metal concentrations, which may be attributed to their coarse leaf surfaces.

Differences in the Amount of PM Accumulation Among Species

A comparison of the dust-retention abilities of different tree species in different functional areas was conducted. One-way ANOVA showed that the dust-retention properties of the three species of trees sampled differed significantly (Fig. 2). The maximum amount of total PM removed by *C. deodara* in the TA, IA, CMA, RA and CA was 112.36 $\mu\text{g}/\text{cm}^2$, 75.12 $\mu\text{g}/\text{cm}^2$, 98.25 $\mu\text{g}/\text{cm}^2$, 117.42 $\mu\text{g}/\text{cm}^2$ and 55.14 $\mu\text{g}/\text{cm}^2$, respectively, which was significantly higher than that removed by *P. tobira* and *C. camphora* in each area. The average amount of PM $>10\ \mu\text{m}$ in diameter retained on *C. deodara* was significantly higher than that retained on *P. tobira* or *C. camphora*, but there was no significant difference between *P. tobira* and *C. camphora*. The average amounts of PM 2.5-10 μm and 0.2-2.5 μm in diameter retained on *C. deodara* were significantly higher than the corresponding amounts retained on *P. tobira* and *C. camphora*. The maximum amounts of PM $>10\ \mu\text{m}$, 2.5-10 μm and 0.2-2.5 μm in diameter were 70.87 $\mu\text{g}/\text{cm}^2$, 26.82 $\mu\text{g}/\text{cm}^2$

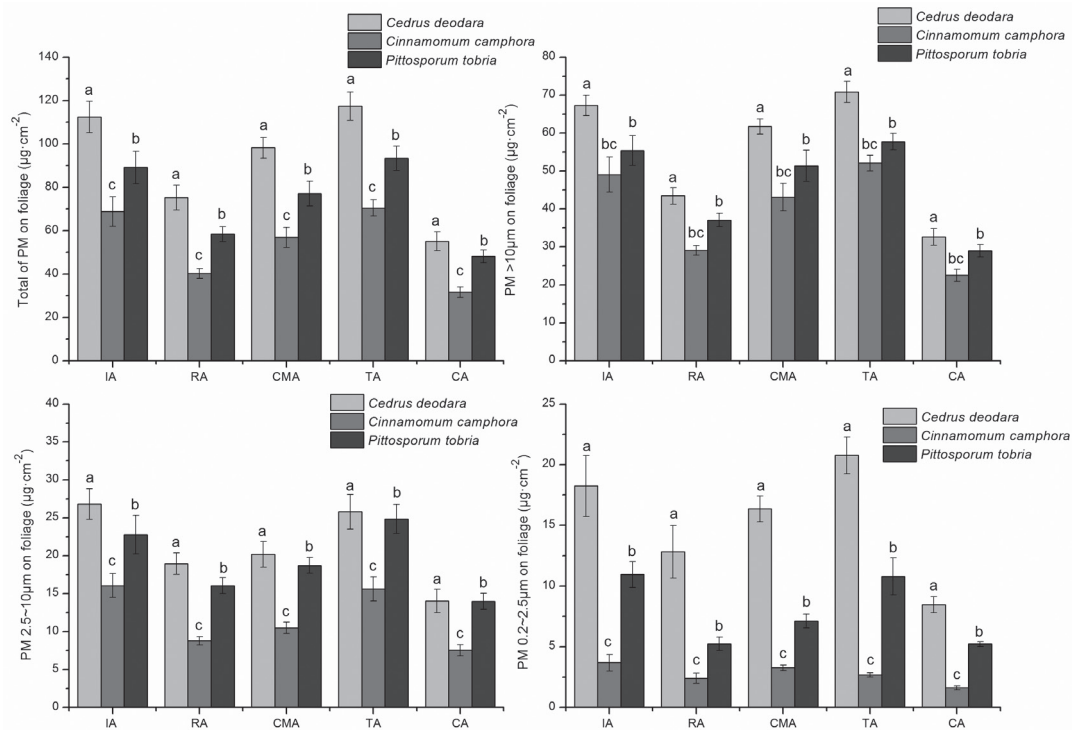


Fig. 2. The mass of PM in different sizes on foliage dust among tree species. Note: Different letters indicated significant difference ($p < 0.05$).

and $20.77 \mu\text{g cm}^{-2}$, respectively. Based on the amounts of total PM, PM $> 10 \mu\text{m}$, PM $2.5-10 \mu\text{m}$, and PM $0.2-2.5 \mu\text{m}$ on the leaf surfaces in each functional area, the areas were ranked as follows: TA $>$ IA $>$ CMA $>$ RA $>$ CA. The presence of large amounts of particles of different sizes in the TA were attributed to fine or ultrafine particles that originated from vehicle exhaust, which was in line with previous studies in Huizhou [7] and Guangzhou [22]. In addition, Song et al. reported fine or ultrafine particles that originated from vehicle exhaust and the burning of coal and biomass in an industrial area [1], which was consistent with our research. In contrast, the RA had less traffic and fewer anthropogenic activities, which led to relatively small amounts of aerial PM in this area.

Mass Distribution Related to Size-Fractionated Particles Among Species

Differences in PM accumulation among the three tested tree species are shown in Figs 3-5. Large PM

($> 10 \mu\text{m}$) made a relatively large contribution to total PM (by wt), with average proportions of 60.0%, 72.9% and 62.8%, respectively, for *C. deodara*, *C. camphora* and *P. tobira*, which indicated that *C. camphora* was most effective at capturing large particles on foliage. Coarse PM ($2.5-10 \mu\text{m}$) made a moderate contribution to total PM (by wt), with average proportions of 26.6%, 23.4% and 21.9%, respectively, for *P. tobira*, *C. deodara* and *C. camphora*. These results showed that the largest proportion of coarse particles was captured by *P. tobira*. Fine PM ($0.2-2.5 \mu\text{m}$) made a small contribution to total PM (by wt), with average proportions of 16.6%, 5.21% and 10.59%, respectively, for *C. deodara*, *C. camphora* and *P. tobira*, which indicated that *C. deodara* was most effective at capturing large particles on foliage. Popek et al. found that large, coarse and fine PM deposited on foliage comprised 65%, 21% and 14% of the total PM, respectively [28]. Although fine particles represented only a small proportion of the captured particles, this accumulation was significant because fine particles are particularly harmful to human health.

Table 2. Comparison of stomatal features and PM_{2.5} amount on the leaf surface of the three investigated tree species in Nanjing.

Tree species	Stomata width (μm)	Stomata length (μm)	Stomata area (μm^2)	Stomata density (number/ mm^2)	PM _{2.5} ($\mu\text{g cm}^{-2}$)
<i>Cedrus deodara</i>	22.14 \pm 0.76a	15.3 \pm 0.25a	265.91 \pm 2.76a	314.37 \pm 3.52a	17.16 \pm 0.78a
<i>Cinnamomum camphora</i>	12.52 \pm 0.14b	5.98 \pm 0.07b	58.77 \pm 1.35c	205.26 \pm 2.86bc	5.36 \pm 0.36b
<i>Pittosporum tobria</i>	14.26 \pm 0.21b	7.16 \pm 0.14b	80.15 \pm 1.68b	238.1 \pm 2.57b	6.27 \pm 0.82b

*Different letters indicate significant difference between means ($p < 0.05$)

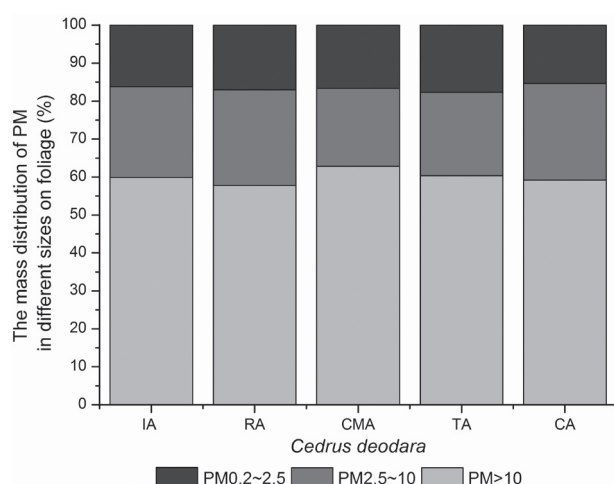


Fig. 3. The mass distribution of PM in different sizes on foliage dust of *Cedrus deodara*.

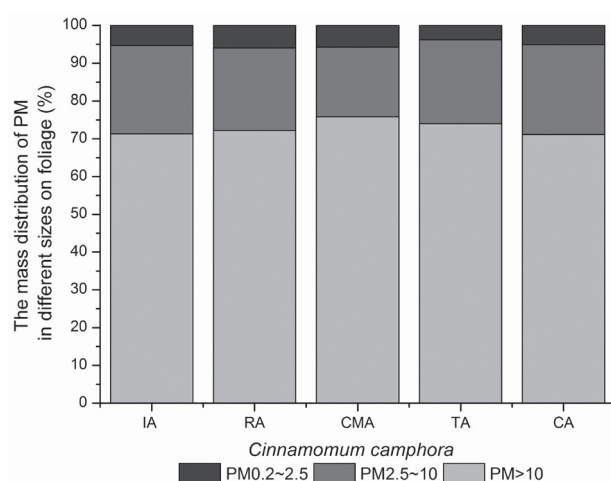


Fig. 4. The mass distribution of PM in different sizes on foliage dust of *Cinnamomum camphora*.

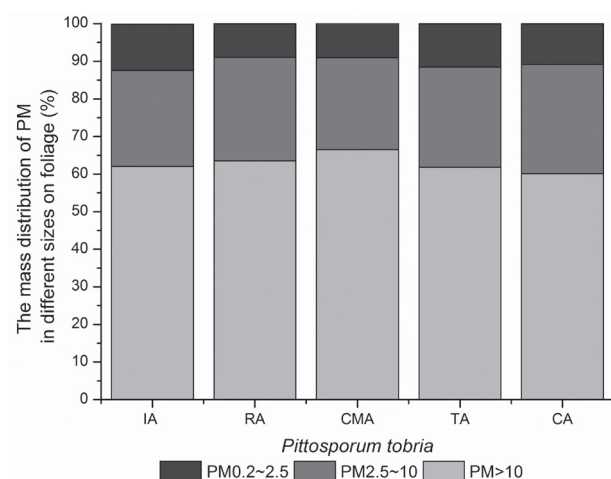


Fig. 5. The mass distribution of PM in different sizes on foliage dust of *Pittosporum tobira*.

Effects of Morphological Structure Characteristics on Dust Adsorption

The characteristics of the stomata of the three tree species selected for this study are shown in Table 2. The stomata width and length of *C. deodara* were significantly greater than those of the other two tree species, and no significant difference was observed between *C. camphora* and *P. tobira* for either measurement. The stomata density of *C. deodara* ($314.37 \pm 3.50/\text{mm}^2$) was significantly higher than that of *P. tobira* ($205.26 \pm 2.86/\text{mm}^2$) and *C. camphora* ($205.26 \pm 2.86/\text{mm}^2$). The stomata area of the tested species was ranked in the following order (largest to smallest): *C. deodara* ($265.91 \pm 2.76 \mu\text{m}^2$) > *P. tobira* ($80.15 \mu\text{m}^2$) > *C. camphora* ($58.77 \mu\text{m}^2$). The high particle capacity of *C. deodara* may be related to their high stomata density and large stomata area. Schönherr et al. indicated that high stomata density and large stomata openings can favor the capture of PM around $2.5 \mu\text{m}$ in diameter [29], which is consistent with our research. In addition, the leaf structure influences the capacity to adsorb atmospheric PM. Foliage surface characteristics influence the ability of plants to retain atmospheric PM. Irregular spheres are clearly shown in Fig. 6. In terms of leaf shape, the needles of coniferous trees like *C. deodara* have a unique microstructure and a relatively thick epicuticular wax layer. Beckett et al. reported that most coniferous trees have slender needles that capture a relatively large amount of particles. The thick epicuticular wax layer on the adaxial needles of *C. deodara* (Fig. 6a) can produce oil that readily captures PM. For example, fly ash particles are easily adsorbed by leaves with oil, leaf hairs or mucilage [18]. Similar to our study, Simon et al. indicated that the roughness of a leaf surface was directly associated with the amount of dust deposited on it [11]. In addition, particle retention by *C. deodara* is enhanced by the rough adaxial surface and thicker epicuticular wax of its needles. The particle capture capacity of *C. camphora* was relatively weak because of its smooth foliar surface and shallow ditch-shaped tissues, which reduce the contact area available for particles (Fig. 6c). Particle retention by *P. tobira* was influenced by the irregularly corrugated shapes on the periphery of its stomata (Fig. 6f). We found that a greater amount of coarse PM was retained on the adaxial leaf surface in comparison with that retained on the abaxial leaf surface.

The deep grooves of *C. deodara* favor particle adsorption in comparison with the shallow grooves of *P. tobira* and *C. camphora*. Xie indicated that blade surface grooves with a depth of $5 \mu\text{m}$ are ideal for retaining PM with a diameter of around $2.5 \mu\text{m}$ [30]. The grooves of *P. tobira* are deeper than those of *C. camphora*, which favored the capture of fine particles by the abaxial leaves of *P. tobira*. As shown in Fig. 6a), the ditch-shaped grooves of the leaves of *C. deodara* increased their roughness, which enhanced their ability to capture particles. As shown in Fig. 6b), there were

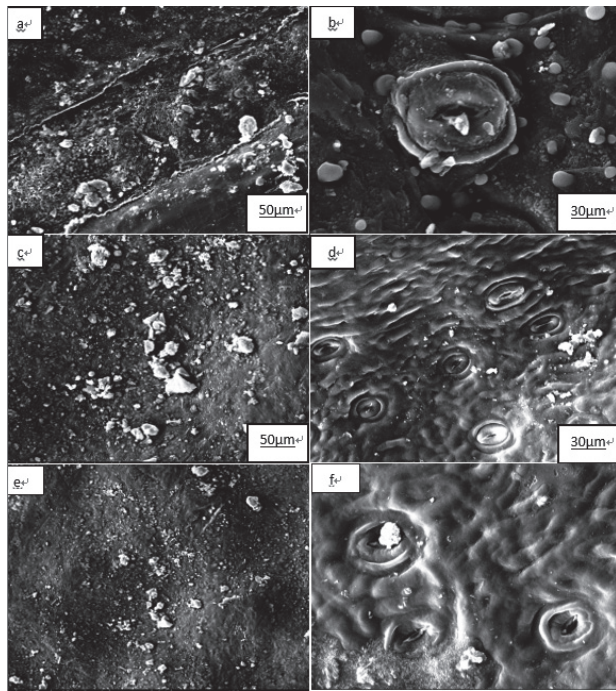


Fig. 6. The SEM images of particulate matter morphology on leaf surface of the three investigated tree species in Nanjing a,c,e) The adaxial leaf of *C. Deodara*, *P. tobira*, *C. camphora*. b) The abaxial leaf of *C. Deodara*, *P. tobira*, *C. camphora*.

more elliptical fine particles in the stomata center and surrounding area of *C. deodara* in comparison with the other tested species. The oily surface of the foliage of *C. deodara* captured more PM in comparison with the other tested leaves and prevented PM from being resuspended in the atmosphere.

Relationship between the Particle Distribution and HM Concentrations

The results of this study show that the particles in the foliar dust on different tree species differed with respect to size and heavy metal concentrations. *C. deodara* had the greatest amount of particles on the leaf surface in each of the investigated areas, as well as the greatest stomata density and area according to the SEM analysis. In addition, *C. deodara* removed more fine particles than did the other two species. Viklander et al. found that particles smaller than 75µm in street dust contained the greatest proportion of heavy metals, which indicated that grain size is an important factor influencing heavy metal accumulation [31]. The SEM images show that the particles on the leaf surface of *C. deodara* were mostly PM with a diameter around 2.5 µm. In contrast, the particles on the surfaces of *C. camphora* leaves consisted of particles larger than 10 µm, which can be observed clearly on the adaxial leaf surface. Similarly, *P. tobira* accumulated more coarse particles on its adaxial leaf surface. In this study, the capacity of tree species to accumulate the heavy metals decreased in the following order: *C. deodara*>*P. tobira*>*C. camphora*.

The different sizes of particles trapped on the surfaces of the leaves of the three tested tree species could explain why we found different concentrations of heavy metals on the surfaces of their leaves.

Conclusions

We studied foliar capture of PM with concomitant accumulation of heavy metals (Cd, Pb, Cu, Zn and Cr) by three tree species in Nanjing, China. The heavy metal concentration in foliar dust can be used as an indicator of atmospheric pollution in urban environments. We found significant differences among the tested tree species in terms of PM and heavy metal accumulation capacity. *C. camphora*, *P. tobira* and *C. deodara* were most effective at capturing large, coarse and fine PM, respectively. The most efficient species was *C. deodara*. Important traits that favored total PM accumulation were oily and coarse leaf surfaces, whereas the capture of fine PM was enhanced by large stomata area and high stomata density. The removal of PM by tree species in this study provides evidence to assist environmental planners in selecting suitable tree species to reduce PM and heavy metal contaminants in the atmosphere.

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Conflict of Interest

The authors declare no conflict of interest.

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