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Indirect method for determining the dry deposition velocity of submicron particulate matter on leaves

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HIGHLIGHTS

- A novel way of determining PM_1 deposition velocity (V_d) with controllable conditions.
- Comparison of wind tunnel was performed using 3 broadleaved and 3 coniferous species.
- The indirect and the wind tunnel methods have no significant difference statistically.
- The sampling of different tree organs may lead to the different results of V_d.
- Application in real situations, the meteorological and PM conditions should be considered.

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ABSTRACT

Plant leaves, as natural receptors of airborne particles, can retain particles onto their surfaces, and absorb gaseous pollutants, thus mitigating air pollution and improving air quality. Dry deposition is considered the main process for particle removal from the atmosphere, and its velocity (V_d) is a crucial parameter for describing the process. Wind tunnels, a conventional approach to determine V_d, are costly and require substantial space, and regular inspections to maintain their systematic integrity. Hence, this study established a simpler and more straightforward method based on relevant research to obtain the submicron particulate matter (aerodynamic diameter $\leq$1 μm; PM_1) V_d on plant leaves. This method involves determining the attenuation pattern of particle concentration in a smog chamber. The V_d values of six tree species (three broadleaved and three coniferous) were obtained through the indirect method. In addition, we determined the V_d of the same tree species with a wind tunnel and compared the values from both methods. Through the paired-samples *t*-test, it's found that there is no significant difference (sig. = 0.59) between these two methods, which means that the indirect method is feasible to determine V_d. We also compared our results with those from other researches, and we found that the V_d values in our study might be lower because leaves and shoots were used in this research, while branches or seedings were selected in the literature. Overall, when applying such values to predictive models or in concrete studies, researchers must consider factors such as real-time meteorological conditions (humidity, temperature, etc.) and pollutant concentration. The indirect method requires less space and is less costly than the wind tunnel method; therefore, it can easily be used to conduct experiments under controlled conditions, which is helpful for simulating various scenarios.

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1. Introduction

Atmospheric aerosols are the primary cause of air pollution, and its concentration is one of the most critical factors that has drawn the attention of the public and scientific community. Submicron particulate matter (aerodynamic diameter ≤1 μm; PM_1) contributes to about 50–60% the concentration of inhalable particles (aerodynamic diameter ≤10 μm; PM_{10}) (Gomišček et al., 2004). Furthermore, PM_1 account for the majority (about 70%) of $PM_{2.5}$ (aerodynamic diameter ≤2.5 μm) (Gomišček et al., 2004; Tan et al., 2016; Zhou et al., 2018). In contrast to larger particles, PM_1 is small in diameter, so it will remain airborne for a longer time, and can be slowly removed from the air through dry deposition, (Costa et al., 2021; Mohan, 2016). Usually, with the increase in particle size by a factor of 10, the dry deposition velocity increases by a factor of 6 (Sehmel and Hodgson, 1978).

Aerosol particles, especially from the biogenic emissions, can lead to the increase of O_3 concentration, thus causing climate changes (Dawson et al., 2014; Shi et al., 2018). As the main cause of air pollution (Sabbagh-Kupelwieser et al., 2010; Urošević et al., 2019), the particles can affect the morphological characteristics of leaves and the flowing of plants (Rai, 2016), and it can also cause strong acidity in rain and leaf litter (Xue et al., 2011; Wu and Zhang, 2018). As to human health, it is reported that submicron PM can penetrate the pulmonary alveoli and enter the blood (Pope, 2000), leading to inflammation (Habre et al., 2018; Wang et al., 2020) or toxicological responses (Jaramillo et al., 2018). Furthermore, the smaller the particle size is, the more its surface-to-volume ratio increases; thus, substances such as toxic metals can accumulate on such particles (Izhar et al., 2016), causing pulmonary and cardiovascular problems (Huang et al., 2016) or even death (Shi et al., 2018).

Dry deposition is the transport of a particle from the atmosphere to a surface. In the absence of precipitation or other wet deposition (Xie et al., 2019), dry deposition is a crucial mechanism that determines the destination of particles and involves the transport and removal of aerosols (Dzierz;anowski and Gawroński, 2011; Willis and Petrokofsky, 2017). Small particles can impact with leaves to get romoved from the atmosphere (Janhäll, 2015) or collide with each other through coagulation effect to form some bigger partcles (Yin et al., 2020), then through the gravitational sedimentation, those bigger particles can get to the leaf surfaces and get adsorbed by them (Janhäll, 2015; Mohan, 2016). Thus, urban green infrastructure is imperative for retaining PM to alleviate air pollution (Abhijith et al., 2020; Escobedo et al., 2015). However, to quantify the particle retention capacity of leaves, the deposition velocity of submicron PM must first be determined.

Dry deposition velocity (V_d) is a parameter primarily used to quantify particle retention capacity and is often applied to help explain deposition phenomena (Giardina and Buffa, 2018) or estimate PM reduction by using predictive models (Jeanjean et al., 2016; Morakinyo and Lam, 2016; Santiago et al., 2017). Wind tunnels are commonly used to determine V_d (Beckett et al., 2000; Freer-Smith et al., 2004; Pullman, 2009), and most relevant studies have determined V_d values at the level of branches and seedlings. We improved an established indirect method for determining $PM_{2.5}$ V_d values (Yin et al., 2019) to determine PM_1 V_d values by using leaves or shoots from six tree species. We compared these values with those obtained through wind tunnel tests to determine why the two methods may yield different results. We aimed to determine which method yields V_d values that more closely resemble those obtained in real situations to assist in estimating the particle retention capacity of leaves through predictive modeling.

The current study attempted to answer the following questions:

- How is the indirect method advantageous for determining the V_d of submicron PM?
- What factors may cause the indirect method and the wind tunnel method to yield different results?
- Compared with field measurements or model evaluation, what factors can cause the difference in V_d values?

2. Materials and methods

2.1. Leaf sampling and preparation

In this study, leaf samples were collected from the plant nursery at Minhang Campus, Shanghai Jiao Tong University. We selected six tree species as materials (Fig. 1); the trees were required to be mature and healthy. All the leaves used in the experiment were gathered from outer canopies at approximately two-thirds of the tree height to ensure favorable growing condition. Three trees from each species were obtained, and each tree was considered a replication. Approximately 30–40 broad leaves and 3–5 small shoots (Fig. 1) were collected from each broadleaf and coniferous tree, respectively. Leaf sampling was conducted in August 2020.

The collected leaves were placed in an ultrasonic cleaner for 30 s to remove PM, rinsed three times with deionized water, and dried in a fume cupboard.

2.2. Tracer selection

NaCl was used to simulate PM_1 as a tracer. NaCl solution with a concentration of 5 g/100 g of H_2O was placed in the aerosol generator (Single-Jet Atomizer, Model 9302, TSI Inc., MN, USA) to form the aerosol. By using the aerosol spectrometer (Laser Aerosol Spectrometer, Model 11-R, Grimm Aerosol Technik, Germany), which contains 30 channels ranging from 0.25 to 30 μm, particles were detected and concentrations of a specific size were recorded. The NaCl aerosol consisted of particles smaller than 1 μm (average $d_p = 0.41$ μm), and the number of PM_1 particles accounted for 99.23% of the total mass of the aerosol.

2.3. Determination of V_d through the indirect method in the smog chamber

As shown in Fig. 2(a), a smog chamber is a closed cylindrical container (Φ = 800 mm, H = 800 mm). A mixing fan was installed at the center of the chamber base; a purification system consisting of nitrogen pipelines was fixed to the top of the chamber, and a nitrogen outlet was connected to a pump at the bottom. Pure nitrogen was pushed through the pipelines into the chamber to carry the particles out of the chamber through the outlet. The purification system enables the particle concentration in the chamber to remain at a low level (lower than 10 μg/m^3).

The aerosol generator was placed at the chamber base. The generated aerosol was dispersed in the chamber by using the mixing fan set to a wind speed of 1 m/s. The concentration of the aerosol was monitored in intervals of 6 s by using the aerosol spectrometer placed at the top of the chamber. When the particle concentration reached 500 μg/m^3, the aerosol generator turned off, enabling the particles to deposit in the chamber. The gradual process of particle concentration reduction (within 1 h) was then analyzed (Fig. 3, control curve) and described using the exponential attenuation model.

When clean leaves were hung in the chamber, the fan was adjusted to maintain a wind speed of 1 m/s; then, the aforementioned particle concentration analysis step was repeated. The observed gradual decrease in particle concentration was described using the exponential attenuation model (Fig. 3, test curve). However, the particle concentration decreased faster with leaves in the chamber than without leaves in the chamber because of the increase in internal surface area.

On the basis of Yin et al. (2019), we refined the V_d determination method. A blank test was required each day for calibrating the performance of the smog chamber and to improve the accuracy of the test results. The derivation of PM_1 V_d is as follows:

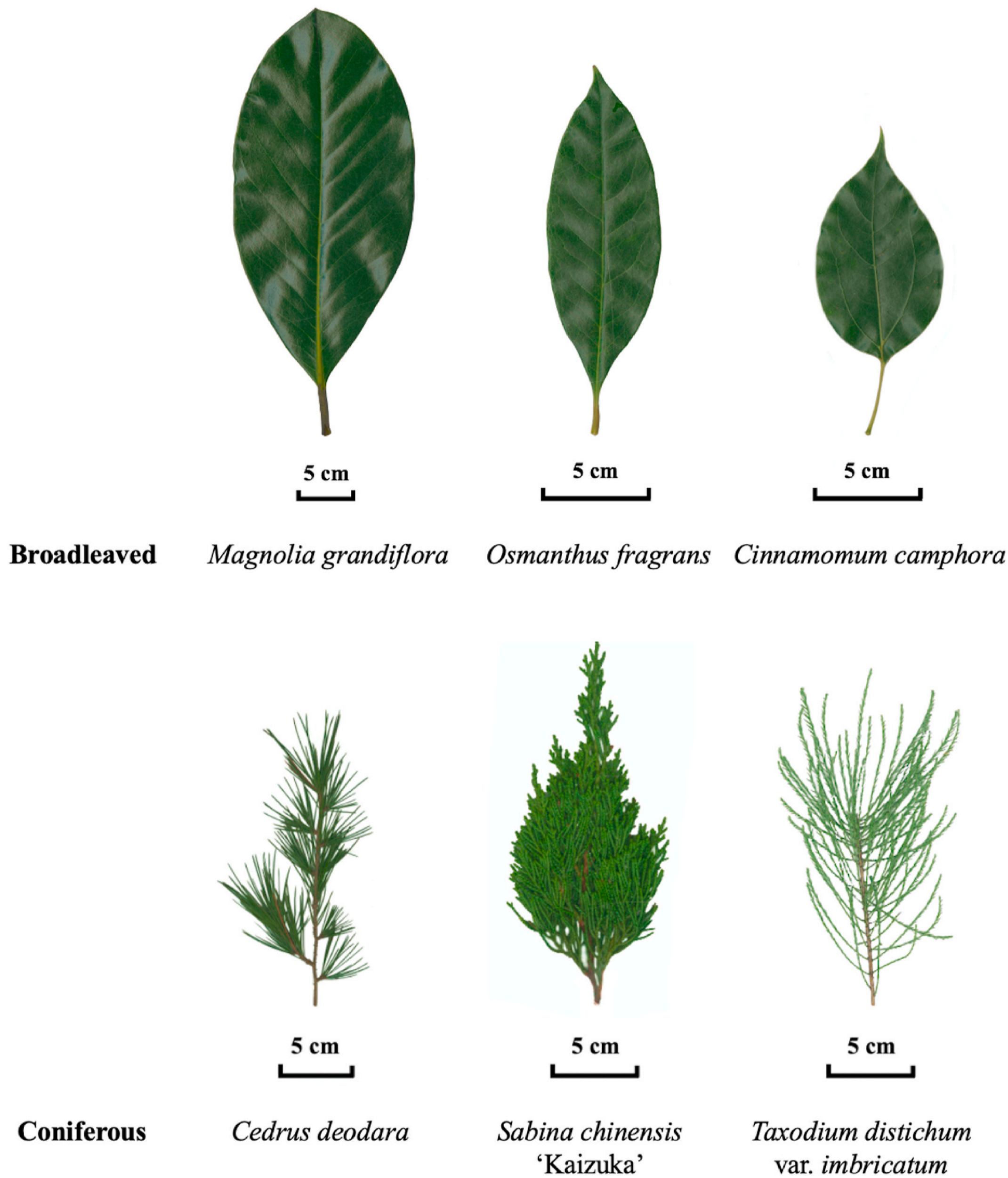


Fig. 1. Six tree species (three broadleaved and three coniferous) selected for the study.

$$y(t)_{leaf} = C_0 \cdot e^{k(t)}$$

$$y(t')_{ctrl} = C_0 \cdot e^{j(t')}$$

When $y(t)_{leaf} = y(t')_{ctrl}$,

then $t' = kt/j$.

After a short period (Δt), the control curve becomes

$$y(t'+\Delta t)_{ctrl} = C_0 \cdot e^{j(t'+\Delta t)} = C_0 \cdot e^{kt+j\cdot\Delta t}$$

On the basis of the definition of V_d, the following equation can be derived:

$$V_d(t) = (e^{j\cdot\Delta t} - e^{k\cdot\Delta t}) \cdot V/(LA \cdot \Delta t)$$

When $\Delta t = 1s$, the PM_1 V_d calculation can be simplified as eq. (1):

$$V_d\,(t) = (e^{j} - e^{k}) \cdot V/LA \tag{1}$$

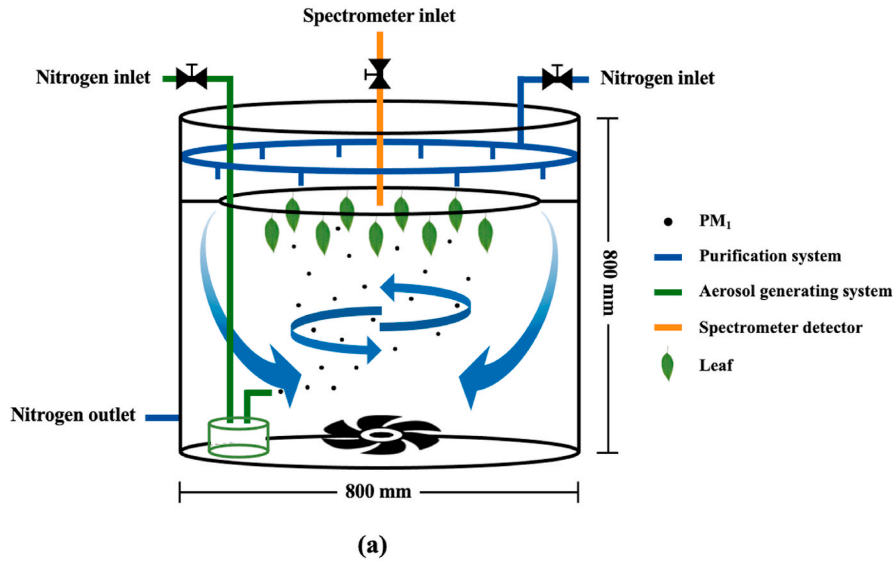
where j is the decay rate constant of particle concentration with no leaves, and k is the decay rate constant of particle concentration with leaves. LA is the leaf area of the samples, and V is the volume of the chamber.

The projected leaf areas of the broad leaves were obtained using WinFOLIA (WinFOLIA Pro 2016, Regent Instruments Inc., Canada), and the areas of the small shoots were obtained using WinSEEDLE (WinSEEDLE Pro 2016, Regent Instruments Inc., Canada).

2.4. Determination of V_d through the wind tunnel method

This experiment adopted methods from relevant studies (Freer-Smith et al., 2004; Pullman, 2009). PM_1 deposition velocity was calculated by plotting the average concentration of PM_1 (μg/m^3) during each NaCl aerosol dosing against the flux of particles onto the leaves (μg/m^2/s).

As shown in Fig. 2(b), The wind tunnel was a semiclosed rectangular columnar tube (L = 3700 mm) fitted with baffles. A mixing fan was



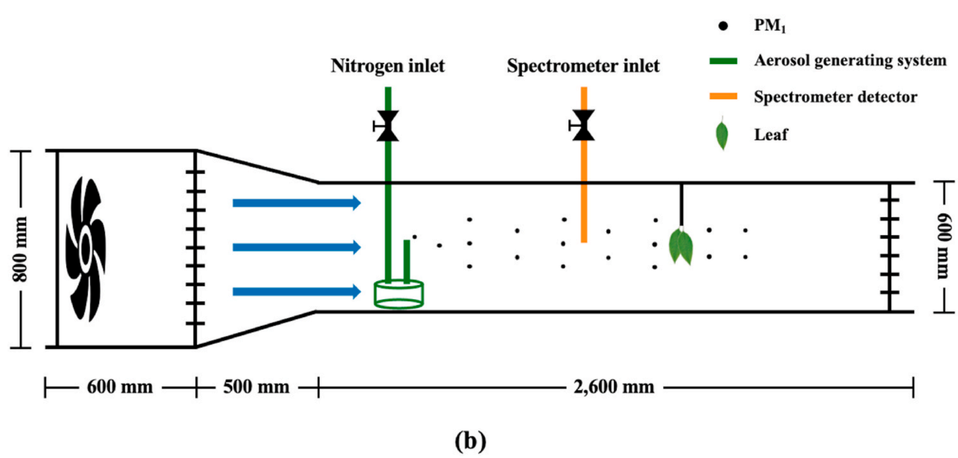


Fig. 2. Schematic of the smog chamber (a) and the wind tunnel (b).

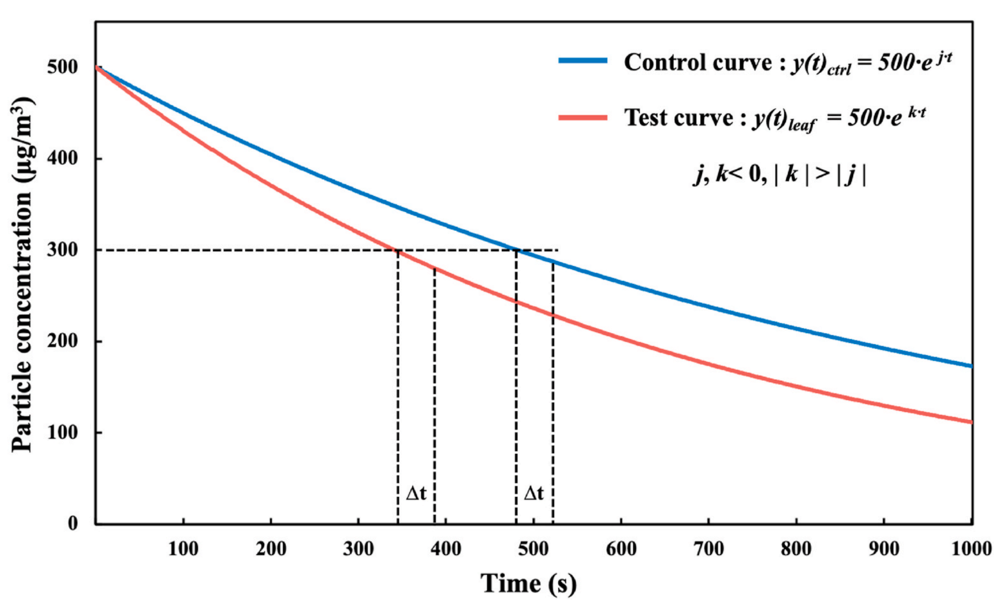


Fig. 3. Exponential decay curves of PM_1 concentration in the chamber. For the control curve (without leaves; k is the decay rate constant in s^{-1}) and test curve (with leaves; j is decay rate constant in s^{-1}), k and j are both negative, and $|k|$ is greater than $|j|$.

installed at one end, and the wind speed was controlled by adjusting the motor. The aerosol generator was placed in the wind tunnel. Leaves were washed in the ultrasonic cleaner for 30 s and then rinsed with deionized water three times. During the last rinse, the solution was diluted with deionized water to the constant volume of 500 mL, and its electrical conductivity (EC) was recorded to provide the background value. The washed and dried leaves were tied on the hanger in the wind tunnel, and the aerosol spectrometer was fixed between the aerosol generator and the tested leaves. During each replication, leaves were exposed for 30 min under a wind speed of 1 m/s, and the concentration of PM_1 was detected using the spectrometer; the average concentration was then obtained through data processing. After the leaves were exposed to NaCl aerosol, the amount of NaCl retained by the leaves was determined by washing the NaCl particles off the leaves with deionized water, diluting the resulting concentration into a fixed volume of 500 mL, and then determining the corresponding EC. The EC after exposure and the background value were calculated and compared subsequently. On the basis of the standard curve of EC and NaCl concentration, the amount of NaCl retained by the leaves was obtained. The total mass of NaCl was corrected by considering the proportion of PM_1 and accounting for the amount of aerosol (PM_1 mass concentration of 69.15%). Leaf areas were measured using WinFOLIA or WinSEEDLE.

2.5. Data processing

One-way analysis of variance and Duncan multiple range test were conducted in RStudio (version 1.3.1056) to identify any significant differences in PM_1 V_d values. The images presented were processed using Microsoft Excel (2019, Version 16.32).

3. Results and discussion

3.1. Comparison between the indirect method and wind tunnel method

As presented in Fig. 4, *C. deodara* had the highest V_d, whereas *C. camphora* had the lowest V_d among the six tested species. Overall, the V_d of the coniferous tree leaves was higher than that of the broad leaves.

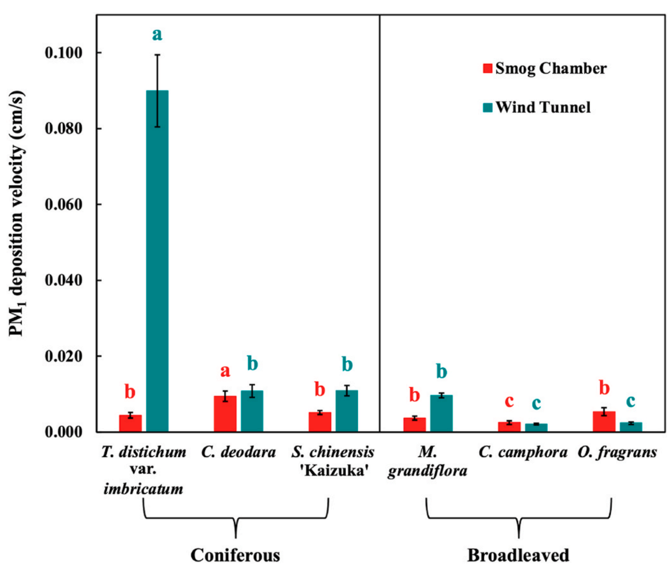


Fig. 4. PM_1 deposition velocity obtained using the indirect method (smog chamber) and the direct method (wind tunnel). Data are presented as mean ± SD (n = 3); comparison is only performed within the same method (shown in the same color); bars with the same letter are not significantly different (Duncan multiple comparisons, significance level $\alpha = 0.05$, $p < 0.001$). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Fig. 4 demonstrates that as indicated through the wind tunnel method, *T. distichum* var. *imbricatum* had the highest V_d and *C. camphora* had the lowest V_d. Overall, the V_d of the coniferous tree leaves was higher than that of the broad leaves, which is the same result obtained using the indirect method.

Through paired-samples *t*-test, we studied the difference between the two methods. The significant value (2-tailed) is 0.59, and it means that there exist no significant difference between these two methods.

3.2. Comparison of the indirect method, wind tunnel method, and field measurements

Deposition velocity is considered to depend strongly on particle size (Yun et al., 2002), wind speed (Mohan, 2016), temperature and humidity (Yin et al., 2020). As presented in Fig. 5, Slinn (1982) predicted V_d according to different particle sizes and at different wind speeds (1, 5, and 10 m/s).

Beckett et al. (2000) used NaCl aerosol generated by a disco smoke machine. The particles were observed to be spherical by using a scanning electron microscope; the mean diameter of the particles was 1.28 μm. Shoots were placed in a wind tunnel and exposed to aerosol for 10 min at a wind speed of 3 m/s. The total mass of NaCl was measured using an atomic absorption spectrometer. Freer-Smith et al. (2004) conducted a similar experiment, but for their study, the mean diameter of NaCl was 0.8 μm and the wind speed was 3 m/s. Pullman (2009) used milled KNO_3 (mean $d_p = 2.5$ μm, unverified) as the tracer particle to determine conifer V_d in a wind tunnel at a wind speed of 6.5 m/s. Hwang et al. (2011) investigated the V_d of submicron soot particles by using a deposition chamber with an aerosol flowrate (Q_a) of 4 L/min. Yin et al. (2019) used diamond powder (d_p 1.8–2.5 μm, verified by spectrometer) to determine $PM_{2.5}$ V_d through an indirect method similar to that used in the current study.

As presented in Fig. 5, the V_d from Hwang et al. (2011) exhibited the same pattern as Slinn's predicted curve of V_d. In addition, the V_d values

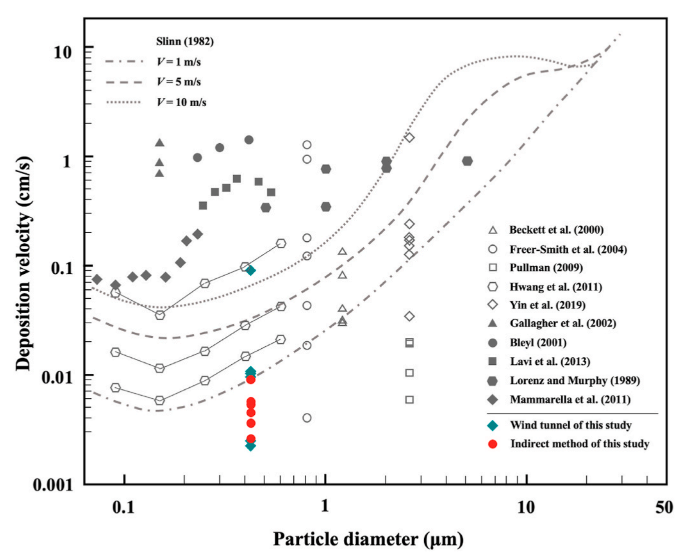


Fig. 5. Predicted V_d according to different particle sizes at wind speeds of 1, 5, and 10 m/s based on data from a study on *Eucalyptus* (Slinn, 1982). V_d data were obtained from wind tunnel experiments, namely those reported in Freer-Smith et al. (2004), Beckett et al. (2000), and Pullman (2009); through a deposition chamber study, namely that of Hwang et al. (2011), through the indirect method, namely that of Yin et al. (2019), and the current study (indirect method and wind tunnel). Those data are presented with hollow legends. Comparison with field measurements or model assessment, namely those reported in Gallagher et al. (2002), Bleyl (2001), Lavi et al. (2013), Lorenz and Murphy (1989), and Mammarella et al. (2011). Those data are presented with filled legends.

reported in Beckett et al. (2000) and Freer-Smith et al. (2004) were similar to the predicted curve, but the V_d values from Pullman (2009) were much lower than the curve. This discrepancy may have resulted because the particle size was not verified in Pullman (2009). The mean of $PM_{2.5}$ V_d from Yin et al. (2019) falls between the predicted wind speed curves at 1 and 5 m/s, a reasonable range. In the current study, the V_d values from the indirect and wind tunnel methods were both similar to the predicted curve at the wind speed of 1 m/s, indicating that the indirect method is practical for determining PM_1 V_d.

In the current study, we determined V_d values at the level of leaves or shoots in the context of both the indirect and wind tunnel methods. At such a small scale, leaves and shoots are prone to environmental conditions, which may have resulted in the low V_d values. However, the V_d values obtained using the wind tunnel method (Fig. 5) were higher in relevant studies than those in the current study. This likely occurred because the relevant studies determined V_d values at the level of branches and seedlings. The leaves of branches and seedlings may not be as susceptible to environmental influences because of the complexity of the spatial distribution of the leaves, thereby leading to less flutter and reduced resuspension. This may be the reason that the V_d values through the wind tunnel method and indirect method in this study were lower than those in the other researches.

Leaf characteristics are also considered some important factors to influence V_d. It's generally found that the coniferous tend to have better capacities of particle adsorption than the broadleaved (Räsänen et al., 2013; Weerakkody et al., 2018). Structurally, the spatial distribution of leaves from the coniferous trees is more complex, which can extend the surfaces to capture more particles (Beckett et al., 2000). Furthermore, a smaller leaf size, bigger trichome density, and shorter petiole length usually have a positive impact on V_d (Zhang et al., 2020, 2021). When V_d values are compared, no matter which method is used, the leaf characteristics should be taken into consideration.

As presented in Fig. 5, Gallagher et al. (2002), Bleyl (2001), Lavi et al. (2013), Lorenz and Murphy (1989), and Mammarella et al. (2011) determined V_d through field tests or model assessment, and resulting V_d values were often higher than those obtained from wind tunnel experiments and indirect methods.

Different from leaves, shoots, branches or seedlings, the whole canopy can strongly affect its surroundings. It's proved that trees can alter the humidity and temperature (Kupper et al., 2011; Manickathan et al., 2018; Shahidan, 2015), and change wind speed (Andújar et al., 2017). Those factors can efficiently influence the deposition of PM. Furthermore, in real situations, deposition progress is more complex, compared with controlled conditions. The higher the wind speed is, the higher the friction velocity is, which accelerates the transport of PM (Mohan, 2016). From the outer canopy of a tree to its inner center, wind speed decreases (Daudet et al., 1999); therefore, V_d also decreases. In comparison with the inner leaves of a tree, the leaves of the outer canopy may play a main role in particle adsorption. The V_d values from field measurements are typically obtained with the aid of various models. Calibrating parameters sometimes requires incorporating existing V_d values from relevant literature (mainly wind tunnel experiments). This can cause the model-calculated V_d values to be higher. The results of the current study can help improve data accuracy when researchers use different models.

When V_d data from indirect or wind tunnel methods are applied to real situations, many limitations must be considered. Compared with controlled conditions, actual conditions (e.g., meteorological conditions, environmental pollution status, and underlying surface properties) can be much more complex. To assess particle retention capacity on a large scale, theoretical values and real-time field measurements must be incorporated to better understand actual deposition progress.

Dry deposition velocity, the most commonly used parameter for quantifying particle deposition progress, can be affected by many factors. Researchers have not only studied how the properties of leaves affect V_d (Weerakkody et al., 2018; Zhang et al., 2021) but have also investigated the effects of environmental conditions (Gomišček et al., 2004; Pullman, 2009). Leaf characteristics and environmental conditions have been proven to affect particle deposition. However, V_d values are typically obtained under controlled conditions through the wind tunnel or indirect methods. Even if researchers use different models to predict air purification efficiency on a large scale and calibrate the models with data under controlled conditions, the results may still contain some inaccuracies. However, the current study may provide a new option for researchers when they search for data to improve the accuracy of their estimations.

3.3. *Advantages of the indirect method*

Compared with a smog chamber, a wind tunnel with uniform velocity profiles is much more costly to purchase and maintain. In addition, it's necessary for a wind tunnel to make calibrations with high precision and accuracy to ensure low turbulences and uniform velocity profile in the test section (Nader et al., 2006). A wind tunnel typically does not contain devices for controlling temperature, humidity, or other conditions; thus, it requires a control room outside of the wind tunnel to manipulate environmental conditions (Welsh, 2013). Unlike wind tunnels, smog chambers are typically smaller and cost less to maintain. Temperature- and humidity-controlling devices can also be installed inside them. Moreover, smog chambers contain purification system, enabling the particle concentration to be maintained at a low level.

Furthermore, the procedures of the two methods also differ. In the wind tunnel, the NaCl concentration must be recorded during each test. The particles deposited on the leaves must then be washed off to determine their EC. The washing duration should not be excessive, otherwise the fluid in the leaves may leak, particularly when the leaves are damaged. According to the standard curve, EC must then be converted to deposition flux. The solubility of NaCl highly depends on temperature, which can further affect EC. Therefore, the standard curve must always be adjusted before converting EC to deposition flux, and the temperature must be strictly controlled during each conversion. To effectively determine V_d through the wind tunnel method, many different steps must be performed; any slight indiscretion during each step can cause an error. An accumulation of errors results in uncertainty, ultimately leading to inaccurate V_d values. By contrast, the indirect method involves determining the attenuation pattern of mass particle concentration in a smog chamber and emphasizes the differences resulting from the presence of leaves. In the corresponding formula, particle concentration in the environment is not included, thereby simplifying calculation.

4. Conclusions

This study established an indirect method to determine PM_1 V_d on plant leaves. Based on the attenuation pattern and the differences resulting from the presence of leaves in a smog chamber, a theoretical V_d was calculated, and the V_d of six tree species (three broadleaved and three coniferous) was obtained. A wind tunnel experiment was also performed, and the V_d values resulting from the two methods were compared. The data from the both methods had no significant difference. The methodology of the indirect method was different from that of the wind tunnel method, but the main purpose of the indirect method is to facilitate simple and straightforward V_d calculation.

Compared with the V_d values by the indirect and wind tunnel methods from the current study and relevant ones, the V_d values obtained in this study were lower. These results likely occurred because this study determined V_d values at the level of leaves or shoots, whereas relevant studies determined V_d values at the scale of branches or seedlings through wind tunnel experiments.

In real situations, the outer canopy of a tree plays a main role in atmospheric particle removal. Thus, if researchers use data from wind tunnel experiments as a parameter for predicting the particle retention ability of trees, the resulting data may contain some inaccuracies. When theoretical data are applied to real situations, factors such as real-time meteorological conditions, environmental pollution status, and underlying surface properties must be accounted for because they can also affect V_d.

CRediT authorship contribution statement

Xuyi Zhang: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. **Shan Yin:** Methodology, Investigation, Writing – original draft, Writing – review & editing. **Junyao Lyu:** Investigation, Formal analysis, Writing – review & editing. **Ningxiao Sun:** Methodology, Investigation. **Guangrong Shen:** Resources, Writing – review & editing. **Chunjiang Liu:** Writing – review & editing, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2021.118692.

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