



A branch scale analytical model for predicting the vegetation collection efficiency of ultrafine particles

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ABSTRACT

The removal of ultrafine particles (UFP) by vegetation is now receiving significant attention given their role in cloud physics, human health and respiratory related diseases. Vegetation is known to be a sink for UFP, prompting interest in their collection efficiency. A number of models have tackled the UFP collection efficiency of an isolated leaf or a flat surface; however, up-scaling these theories to the ecosystem level has resisted complete theoretical treatment. To progress on a narrower scope of this problem, simultaneous experimental and theoretical investigations are carried out at the “intermediate” branch scale. Such a scale retains the large number of leaves and their interaction with the flow without the heterogeneities and added geometric complexities encountered within ecosystems. The experiments focused on the collection efficiencies of UFP in the size range 12.6–102 nm for pine and juniper branches in a wind tunnel facility. Scanning mobility particle sizers were used to measure the concentration of each diameter class of UFP upstream and downstream of the vegetation branches thereby allowing the determination of the UFP vegetation collection efficiencies. The UFP vegetation collection efficiency was measured at different wind speeds ($0.3\text{--}1.5\text{ m s}^{-1}$), packing density (i.e. volume fraction of leaf or needle fibers; 0.017 and 0.040 for pine and 0.037, 0.055 for juniper), and branch orientations. These measurements were then used to investigate the performance of a proposed analytical model that predicts the branch-scale collection efficiency using conventional canopy properties such as the drag coefficient and leaf area density. Despite the numerous simplifications employed, the proposed analytical model agreed with the wind tunnel measurements mostly to within 20%. This analytical tractability can benefit future air quality and climate models incorporating UFP.

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1. Introduction

Ultrafine particles (UFP) are characterized by their large surface area and small sizes (particle diameter $<100\text{ nm}$) with a lifetime in the atmosphere ranging from few seconds to several days (Ketzel and Berkowicz, 2004; Riipinen et al., 2011; Williams et al., 2002). Interest in UFP is now exponentially proliferating due to their role in climate change and human health. UFP can form cloud condensation nuclei (CCN) through condensation or coagulation (Pierce and Adams, 2007) and can modify cloud albedo (Kazil et al., 2010; McFiggans et al., 2006). Adverse health effects such as

respiratory (Dockery, 2001; McConnell et al., 2006; Samet et al., 2000) and cardiovascular diseases (Delfino et al., 2005) have also been linked to UFP. Hence, risk and hazard predictions necessitates the quantification of UFP concentration, which, in turn, requires knowledge of the primary UFP sinks.

One important sink for UFP is their removal by vegetation, a sink that was recognized as early as 1915 for gases when O’Gara showed that SO_2 emissions from elevated smoke stacks at smelter operations induced crop damage (Hosker and Lindberg, 1982; Thomas, 1951). The main collection mechanism by vegetation is Brownian diffusion primarily due to the small size of UFP with other mechanism such as interception and inertial impaction being less significant (Litschke and Kuttler, 2008; Petroff et al., 2008a). Phoretic effects, including turbo-phoresis, electro-phoresis and thermo-phoresis may modify the collection mechanism of UFP. Phoretic effects suggest that particles are more prone to move in the direction of decreasing turbulent energy, electric field, and

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temperature, respectively. Phoretic effects have been shown to increase the UFP collection efficiency in wet deposition (Andronache, 2004; Andronache et al., 2006). Turbo-phoresis, a term first coined in the mid 1970s (Caporali et al., 1975), has also received significant attention in designs of indoor pipes and ducts (Zhao and Wu, 2006). Petroff et al. (2008b) discussed a form of phoretic effect – mainly related to turbo-phoresis but they call this term turbulent impaction (in analogy to inertial impaction). Likewise, Katul et al. (2010, 2011) discussed turbo-phoresis in detail and showed that this effect appears to be significant when particle sizes exceed 100 nm. Hence, turbo-phoresis in isolation is not likely to play a primary role in the vegetation collection mechanisms of UFP. Electro-phoresis may also play a role in particle deposition on the tip of the top needles of trees for low winds (Tamm et al., 2001), but its effects might be sheltered in a canopy (Petroff and Zhang, 2010). Studies on the effects of thermo-phoresis mostly focused on particle deposition to a tube (Lin et al., 2008; Pratsinis and Kim, 1989) and their importance is connected to the difference between the air and skin temperature. In general, if the canopy is well-coupled to the atmosphere, this effect is likely to be small. However, it should be emphasized that understanding of the effects (especially the combined effects) from phoretic terms on particle collection by vegetation is still in its infancy (Petroff et al., 2008a; Petroff and Zhang, 2010; Slinn, 1982). In addition to the many uncertainties originating from phoretic effects, a large number of inter-related factors, mostly pertinent to the spatial scale of the problem, add ambiguity to modeling the UFP collection efficiency by ecosystems.

Studies of particle deposition onto vegetated surfaces have benefited from developments in filtration theory (Brown, 1993; Baron and Willeke, 2001; Spurny, 1998) and studies of particle deposition onto uniform boundaries. Filtration theory elucidates particle deposition to fibers of various shapes and has been shown to adequately describe particle deposition to vegetation (Davidson et al., 1982; Lin and Khlystov, 2012). Moreover, particle deposition onto uniform cylindrical surface has been studied (Friedlander, 2000). Other uniform shapes such as plates and ducts provide foundation for single leaf (or big leaf) models (Feng, 2008; Wesely and Hicks, 2000). The big leaf representation is now common when parameterizing the deposition of particles onto vegetated surfaces, especially in air quality and climate models (Feng, 2008). In fact, a big-leaf representation remains the basis for representing the effects of the vegetation particle collection mechanism as a “deposition velocity”, shown to vary by more than 3 orders of magnitude for various particle sizes (Sehmel, 1980). The term “big leaf” refers to models that “compress” the canopy vertical dimension and all its concomitant effects on the canopy microclimate into ‘effective’ surface roughness and resistance parameters that can then be used in estimating deposition velocity. Such assumptions may explain why some big-leaf models predict particle deposition onto forested surfaces that differ from actual measurements by a factor of 3–5 (Pryor et al., 2009), especially in the UFP range. However, for ecosystem scale particle deposition modeling, it is unrealistic to resolve the collection mechanism at a scale commensurate to an isolated single leaf given the large number of leaves within ecosystems, the complexity in their spatial arrangement, and the variations in micro-climatic conditions (e.g. mean flow and turbulent stresses) around each single leaf. Hence, understanding the deposition at some intermediate scale between leaf and ecosystem is needed. The branch scale appears to be a logical starting point for any spatial up-scaling beyond the single leaf, the subject here. Such a scale still provides a sufficiently large number of leaves for collecting UFP, but their spatial arrangement remains much simpler than an ensemble of leaves on numerous branches within an ecosystem.

Wind tunnel tests represent a convenient way of studying aerosol deposition onto vegetation because the flow parameters (speed and orientation) can be varied in a controlled manner. Moreover, any thermo-phoretic effect is likely to be minor given the near equivalence between air and skin temperature of the foliage in wind tunnels. However, most wind tunnel studies so far have focused on the accumulation mode (0.1–2 μm) and coarse mode (2–50 μm) particle deposition (Belot et al., 1976; Fujii et al., 2008; Little and Wiffen, 1977; Reinap et al., 2009). Recently, our group reported wind tunnel measurements of UFP removal by *Pinus taeda* (pine) and *Juniperus chinensis* (juniper) branches (Lin and Khlystov, 2012). The choice of these two species is primarily due to structural differences in foliage smoothness properties and their clumping at the branch scale. This setup enables the development and testing of particle size-resolving models for predicting the branch-scale vegetation collection efficiency of UFP without the added complexity of phoretic terms. Here, such a model is proposed to explore the UFP collection efficiency measured in the wind tunnel. The analytical model uses conventional canopy attributes such as the dimensionless drag coefficient and leaf area index and is “forced” by upwind mean wind speed. The model predicts the concentration, turbulent flux, and collection efficiency along the segment vegetation length. While this wind tunnel setup and model framework do not address all the complexities encountered in UFP deposition onto ecosystems, they do zoom onto a key process common to all UFP deposition – Brownian diffusion.

2. Experimental

Details of the experimental setup can be found in Lin and Khlystov (2012) but a brief review is provided for completeness. The measurements of UFP vegetation collection efficiency were made in a wind tunnel having a test section that is 16 cm wide (W_T), 18 cm high (H_T), and 226 cm in length (Fig. 1). A digital thermometer and barometer (Cole-Parmer Workstation) was used to record the air temperature and atmospheric pressure. A mixer was placed in front of the wind tunnel to obtain aerosol uniformity. The tunnel was sampling room air. To improve signal to noise ratio of the measurements, solid ammonium sulfate aerosol was added at the entrance of the tunnel. The ammonium sulfate aerosol was generated using a constant output atomizer (model 3076 TSI) by spraying 0.01% aqueous solution of ammonium sulfate with the targeted size distribution being centered at 20 nm after drying. The area-averaged mean entrance velocity (U_{in}) was recorded by a vaneometer (DWYER) located just upwind from the branches. The test section populated by fairly uniform vegetation elements has a length $L_x = 1$ m and begins 1.2 m downstream from the tunnel entrance (Fig. 1). A Scanning Mobility Particle Sizer (SMPS model 3080 TSI) was used to measure the mean particle concentration through the two sampling ports located before (C_{in}) and after (C_{out}) the vegetation (Fig. 1). The two sampling ports were connected via a three-way valve, enabling us to switch between the two ports. The relative transmission efficiency through both lines was measured prior to each experiment, i.e. without branches. The data obtained through one of the lines was corrected to match that measured through the other. This correction, which accounts for any differences in deposition through the two lines and any tunnel wall losses, did not exceed 10% in any of the experiments.

Five different wind speeds (0.3, 0.5, 0.7, 1.0, 1.5 m s^{-1}) similar to wind speed in natural settings (i.e. 0–3 m s^{-1}) (Harman and Finnigan, 2007; Poggi et al., 2004; Queck and Bernhofer, 2010) were used. We also selected two packing densities (PD, defined as volume of the branches divided by the volume of the tunnel section occupied by them) for each vegetation. “Packing density” is commonly used in filtration studies, while “volume porosity”

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