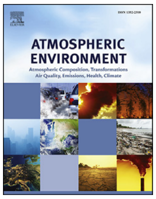
Atmospheric Environment 143 (2016) 133–138

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv



Short communication

Empirical estimates of size-resolved precipitation scavenging coefficients for ultrafine particles

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HIGHLIGHTS

- Below-cloud scavenging coefficients derived from long-term particle measurements.
- Scavenging coefficients decline by three as diameter goes from 15 nm to 100 nm.
- Good agreement with empirical parameterization of Laakso et al. (2003).

ARTICLE INFO

Article history:
Received 10 June 2016
Received in revised form
21 July 2016
Accepted 9 August 2016
Available online 11 August 2016

Keywords:
Below-cloud
Particle removal
Wet scavenging
Observational
Aerosol particles

ABSTRACT

Below-cloud scavenging coefficients for ultrafine particles (UFP) exhibit comparatively large uncertainties in part because of the limited availability of observational data sets from which robust parameterizations can be derived or that can be used to evaluate output from numerical models. Long time series of measured near-surface UFP size distributions and precipitation intensity from the Midwestern USA are used here to explore uncertainties in scavenging coefficients and test both the generalizability of a previous empirical parameterization developed using similar data from a boreal forest in Finland (Laakso et al., 2003) and whether a more parsimonious formulation can be developed. Scavenging coefficients (λ) over an ensemble of 95 rain events (with a median intensity of 1.56 mm h^{-1}) and 104 particle diameter (D_p) classes (from 10 to 400 nm) indicate a mean value of 3.4×10^{-5} s^{-1} (with a standard error of 1.1×10^{-6} s^{-1}) and a median of 1.9×10^{-5} s^{-1} (interquartile range: -2.0×10^{-5} to 7.5×10^{-5} s^{-1}). The median scavenging coefficients for D_p: 10–400 nm computed over all 95 rain events exhibit close agreement with the empirical parameterization proposed by (Laakso et al., 2003). They decline from ~4.1×10^{-5} s^{-1} for D_p of 10–19 nm, to ~1.6×10^{-5} s^{-1} for D_p of 80–113 nm, and show an increasing tendency for $D_p > 200$ nm.

1. Introduction and motivation

Below-cloud scavenging of aerosol particles by hydrometeors plays an important role in defining their atmospheric lifetimes and in situ particle size distributions (Andronache, 2003; Pruppacher and Klett, 1997). Full numerical treatment of below-cloud scavenging in atmospheric chemistry models is computationally demanding and is subject to large uncertainties due to non-linear dependencies of scavenging efficiencies with hydrometeor diameter spectra and phase, precipitation intensity, atmospheric turbulence and particle diameters (Andronache, 2003; Pruppacher and Klett, 1997). Thus, some atmospheric chemistry models continue to use scavenging coefficients expressed as a function of rainfall rate for specific particle diameters or modes (Feng, 2007) to represent this process (e.g. the EMEP MSC-W Eulerian chemical transport model (Simpson et al., 2012), DEHM (Frohn et al., 2001), and MATCH (Robertson et al., 1999)).

A number of previous publications have explained the mechanisms and theorized dependencies of below-cloud scavenging (e.g. (Andronache, 2003; Pruppacher and Klett, 1997; Wang et al., 2010)), so they are described only briefly here. The rate at which particles are scavenged by rain droplets depends on the collision and collection efficiency (where the latter term is often assumed to be 1 for particles with diameters much smaller than the rain droplet diameter), and is thus a function of the rainfall intensity and rain droplet size distribution, as well as the size distribution and composition of the in situ particles. The collision efficiency

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http://dx.doi.org/10.1016/j.atmosenv.2016.08.036
1352-2310/

(E(D,D_p)) between a falling raindrop of a given diameter (D) and an in situ particle (of diameter, D_p) is theorized to decrease with increasing particle diameter in the D_p range ~1–100 nm, to remain fairly constant (i.e. within a factor of approximately 2–5) in the D_p range ~100 nm to 1 μm, and then to exhibit a rapid increase with increasing D_p (Wang et al., 2010). E(D,D_p) for the smallest D_p is dominated by Brownian diffusion with additional contributions from thermophoresis and electrostatic forces (Andronache, 2004), and atmospheric turbulence (Quérel et al., 2014). For larger D_p interception and impaction play a larger role in dictating collision efficiency (Pruppacher and Klett, 1997; Wang et al., 2010). E(D,D_p) increases with D, so given rainfall events with lower intensities (rainfall rates) are typically characterized by smaller rain droplet diameters than those with higher intensities (Marshall and Palmer, 1948), the *a priori* expectation is that higher rainfall rates are typically associated with a larger number of larger rain droplets, and thus higher particle scavenging efficiencies. This expectation has largely been realized in observational analyses e.g. (Castro et al., 2010; Chate, 2005; Laakso et al., 2003; Maria and Russell, 2005; Volken and Schumann, 1993; Zhao et al., 2015). However, there are large event-to-event and site-to-site variations in experimentally derived scavenging efficiencies (Andronache et al., 2006). Of the observational studies of below-cloud scavenging of ultrafine particles (UFP, particles with diameters (D_p) less than 100 nm) that have been conducted to date, only two (Laakso et al., 2003; Zikova and Zdimal, 2016) have considered sufficient rain event sample sizes ($n \approx 100$) to generate statistically robust estimates of size-resolved scavenging coefficients, and only one (Laakso et al., 2003) sought to develop an empirical parameterization of size-resolved scavenging coefficients. Herein, we use a 30-month time series of measured near-surface UFP size distributions and precipitation intensity to explore the generalizability of the empirical parameterization of Laakso et al. (2003), and to evaluate whether alternative, more parsimonious formulations can be advanced.

2. Data and methods

The meteorological and particle size distribution (PSD) data analyzed herein were collected at an AmeriFlux site in southern Indiana (in the Morgan Monroe State Forest (MMSF), 39°19′ N, 86°25′ W, 275 m a.s.l (Schmid et al., 2000)) from December 2006 to April 2009. Precipitation at this site is fairly evenly distributed across the year and has an average occurrence of approximately 1 day in 3 (see précis description of the meteorological conditions in Fig. 1). Size-resolved particle number concentrations in 104 size classes over the D_p range: 10–400 nm were obtained using a TSI scanning mobility particle sizer (SMPS3936) system comprising an electrostatic classifier (TSI-3080), long differential mobility analyzer (TSI-DMA3081) and a condensation particle counter (TSI-3025A). A second SMPS3936 system was also operated on the same manifold and comprised an electrostatic classifier (TSI-3080), nano-DMA (TSI-DMA3085) and a condensation particle counter (TSI-3786) and for which particle concentrations for D_p in the range 6–100 nm are reported. The sampling protocol was such that air was drawn from a height of 46-m (above a forest canopy with a mean height ~26–28 m) through copper tubing for 10-min in each half-hour period, with sampling at two other heights via a common manifold in each 30-min period (Pryor et al., 2010). Copper was chosen for the sampling lines because it was available at very long sections reducing the number of connections to two, is malleable (meaning all bends could be relatively smooth) and is relatively inert and resistant to acquisition of charge, but with such long sampling lines particle losses are inevitable. Thus, transmission efficiencies derived experimentally (which for D_p = 10 nm are

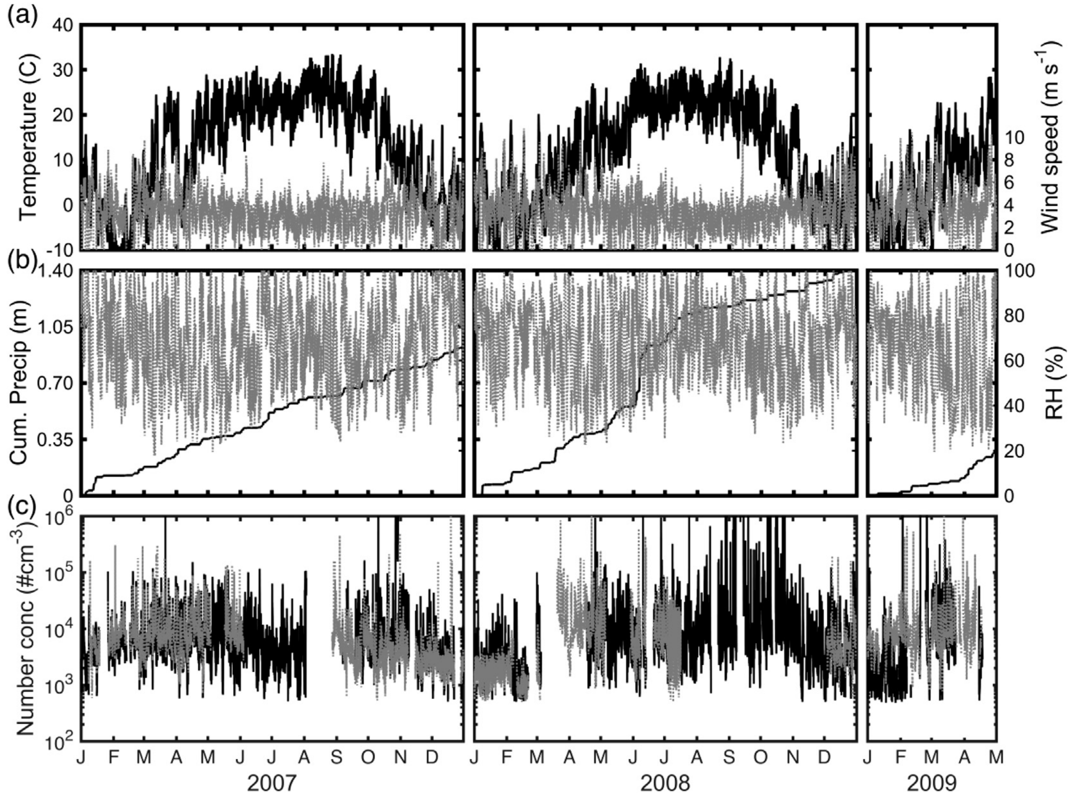

Fig. 1. Overview of meteorological conditions and particle number concentrations at MMSF during January 2007–April 2009. (a) Hourly average air temperature (°C) (black) and wind speed (m s^{-1}) (gray) at 46-m. (b) Cumulative precipitation (Cum. Precip, m) in each year (black) and hourly average relative humidity (RH, %) again at 46-m. (c) Total number particle concentrations (# cm^{-3}) at 46-m (Number conc) from an SMPS with a nano-DMA (D_p: 6–100 nm) (black) and an SMPS with a long-DMA (D_p: 10–400 nm) (gray).