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

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9 **Highlights**

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

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 hr^{-1}) and 104 particle diameter (D_p) classes (from 10 to 400 nm) indicate a mean value of $3.4 \times 10^{-5} \text{ s}^{-1}$ (with a standard error of $1.1 \times 10^{-6} \text{ s}^{-1}$) and a median of $1.9 \times 10^{-5} \text{ s}^{-1}$ (interquartile range: -2.0×10^{-5} to $7.5 \times 10^{-5} \text{ s}^{-1}$). 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 $\sim 4.1 \times 10^{-5} \text{ s}^{-1}$ for D_p of 10-19 nm, to $\sim 1.6 \times 10^{-5} \text{ s}^{-1}$ for D_p of 80-113 nm, and show an increasing tendency for $D_p > 200 \text{ nm}$.

Key words: below-cloud, particle removal, wet scavenging, observational, aerosol particles

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 ($E(D, D_p)$) 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 μ m, 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 (Qurel 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 Figure 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-minutes in each half-hour period, with sampling at two other heights via a common manifold in each 30-minute 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 below 0.3, but increase to over 0.5 for $D_p > 20$ nm) were used to determine empirical correction factors (Pryor et al. 2010) that have been applied to the observed PSD. We assume the particle losses in the copper sampling lines are independent of variations in humidity and other environmental conditions.

Rainfall rates derive from measurements using a Texas Electronics TE525 tipping bucket rain gauge also deployed at 46 m (Figure 1b). Since this instrument is deployed without a wind-

shield it is subject to under-capture during high wind events. The modal correction factor computed using the approach of (Hildebrandt et al. 2007) and wind speed data from a sonic anemometer deployed at 46-m is ~ 1.1 (Figure 2b). Frozen hydrometeors (snow, ice and hail) differ greatly from liquid droplets both in terms of the particle capture efficiencies (Sparmacher et al. 1993) and the accuracy of their measurement by tipping bucket rain gauges (Savina et al. 2012). To avoid inclusion of snow events in the analysis, METAR reports from the National Weather Service station KIND-Indianapolis, IN (located approximately 45 km to the north of MMSF) are used to screen out periods when snow or hail was observed. The procedure used is as follows; any event during which precipitation is reported by the TE525 at MMSF and for which snow/ice/hail were reported at KIND during that hour or the two hours prior to or subsequent to that time stamp are excluded from consideration. The resolution of the tipping bucket rain gauge is 0.254 mm (equal to one tip). To be consistent with the analysis of (Laakso et al. 2003) and avoid inclusion of very light and comparatively poorly quantified rain events, events were only included in the analysis if the precipitation intensity $\geq 0.4 \text{ mm hr}^{-1}$.

The in situ measured PSDs and rainfall rates are used to derive size-resolved scavenging coefficients ($\lambda(D_p)$) using:

$$\lambda(D_p) = -\frac{1}{t_1 - t_0} \ln \left(\frac{c_1(D_p)}{c_0(D_p)} \right) \quad (1)$$

Where D_p is the particle diameter, $t_1 - t_0$ is the length of the rain event in seconds, and $c_0(D_p)$ and $c_1(D_p)$ are the average number concentration of particles in each size bin in the 30-minute periods before and after the rain event, respectively.

A premise of the empirical approach to determining size-resolved scavenging efficiencies is that with the exception of removal by below-cloud scavenging the particle population is stationary over the course of the rain event. To avoid strongly non-stationary conditions such as

frontal passages or new particle formation occurrence, events are only included in the analysis if two criteria are met: (i) the wind direction (as measured using a 03001-5 R.M. Young Wind Sentry Set deployed at 46-m) at the start and end of the rain event did not differ by more than 90° and (ii) new particle formation (NPF) was not observed in the hour following cessation of rain (i.e. particle concentrations $< 1 \times 10^3 \text{ cm}^{-3}$ for $D_p = 10\text{-}15 \text{ nm}$, inclusive and visual inspection of plots of the time evolution of PSD did not show evidence of NPF (see a detailed description of robust subjective and objective ways to detect NPF given in (Kulmala et al. 2012))). To avoid unstable estimates of the concentration ratio, an individual diameter class was only included in the analysis of a specific rain event if both $c_0(D_p)$ and $c_1(D_p)$ exceeded 10 cm^{-3} .

(Laakso et al. 2003) proposed the following functional form for the relationship between the median $\lambda(D_p)$ (for each of the 29 logarithmically distributed size channels (D_p) in the diameter space 10-500 nm) and precipitation rate:

$$\log_{10} \left(\frac{\lambda}{\lambda_0} \right) = a + b \times d_p^{-4} + c \times d_p^{-3} + d \times d_p^{-2} + e \times d_p^{-1} + f \times \left(\frac{p}{p_0} \right)^{0.5} \quad (2)$$

Where λ_0 is 1 s^{-1} , $d_p = \log_{10}(D_p/D_{p0})$, $D_{p0} = 1 \text{ m}$, p is the rain rate in mm hr^{-1} , p_0 is 1 mm hr^{-1} , and a - f are the fitted coefficients.

In this analysis, we evaluate the degree of agreement between the empirical coefficients (a - f) as derived by (Laakso et al. 2003) and observational estimates of $\lambda(D_p)$ as derived from the MMSF data set. We also use the framework described by (2) to develop least squares best-fit estimates from our observations. Our fitting procedure is as follows: the coefficient f (which describes the scaling term for precipitation intensity) is first derived by conditionally sampling the scavenging coefficients in three precipitation intensity classes (light ($0.4\text{-}2 \text{ mm hr}^{-1}$), moderate ($2\text{-}5 \text{ mm hr}^{-1}$), and heavy ($>5 \text{ mm hr}^{-1}$)) in addition to treating the whole sample

ensemble (i.e. all rainfall intensities). Then f and the median value of p is used with the estimates of $\log_{10}\left(\frac{\lambda}{\lambda_0}\right)$ as a composite dependent variable to generate the other fitting coefficients in equation (2). Application of regression to fit complex (multi-predictor) equations (such as (2)) to small data sets can lead to model over-fitting. Hence we use stepwise regression to examine if a more parsimonious fit can be derived. In this analysis we use stepwise regression with a threshold minimum p-value of 0.1 for a term to be removed.

There are four primary potential sources of discrepancies between empirically derived $\lambda(D_p)$ from different studies:

- a) Variations in the methodology used to select the cases (e.g. the stationarity criteria) and those used to compute the scavenging coefficients (e.g. the discretization of the precipitation data). The precipitation data at the MMSF site are reported with hourly discretization, while (Laakso et al. 2003) had precipitation rates discretized at 15-minute intervals and they included all events with a duration > 0.5 hours. The hourly discretization of precipitation data from MMSF will inevitably result in lower precipitation intensities than if the precipitation data were discretized at higher frequency. Also, herein we compare UFP concentrations before and after rain events to capture the net effect of the precipitation event. That is we compare distributions at $t=0$, prior to the commencement of precipitation, and at time $t=n$, after precipitation and thus each individual event results in a single estimate of λ for each particle diameter so each event is equally weighted in the estimation of median and mean $\lambda(D_p)$ regardless of the event duration. However, previous authors calculated λ from the UFP concentrations during rain events and thus compare the PSD at time $t=0$, just prior to precipitation and at time $t=n$, and at $t+n$ and $t+2n$, (where n is the temporal

discretization of the PSD measurements) during the rain event (Laakso et al. 2003; Zikova and Zdimal 2016).

- b) Variations in the sampling system. Here we present data that were sampled well above a forest canopy to avoid local contamination of the PSD, and thus used long sampling lines that result in large corrections for tubing losses. Other sampling locations may not require use of long sampling lines.
- c) Differences in particle instrumentation both in terms of operating principle (e.g. SMPS versus Fast Mobility Particle Sizers (Hornsby and Pryor 2014)) and the discretization of the PSD. Also differences in instrument maintenance and deployment protocols (e.g. the presence of a wind shielding on, or heating of, rain gauges).
- d) Differences in the type and intensity of precipitation and thus the rain droplet size distribution.

To address some of these sources of uncertainty we compute $\lambda(D_p)$ from two different SMPS3936 systems connected to the same sampling lines and manifold system. One SMPS was operated with a long-DMA in order to capture a wide range of particles diameters (10-400 nm), and one with a short-DMA to provide estimates of scavenging coefficients for $D_p < 100$ nm for a larger event sample (i.e. more rain events), and also undertake an assessment of $\lambda(D_p)$ for the diameter range 10-100 nm for rainfall events sampled by both SMPS systems.

3 Results

The rain events used to compute the scavenging coefficients from the SMPS3936 with a long-DMA exhibit a mean duration of almost 3 hours (median of 2 hours) and a mean intensity of 2.47 mm hr⁻¹ (median of 1.57 mm hr⁻¹) (Figure 2 and Table 1). Thus, the average rain

intensities are higher than those reported in (Laakso et al. 2003) (median value of 0.8 mm hr^{-1}), but cover the same range of values.

Although a substantial fraction of the scavenging coefficients (λ) from (1) are close to (or below) zero, the mean- λ is significantly different from zero and positive. We postulate that the negative λ values (that have also been observed in previous studies (Laakso et al. 2003; Zikova and Zdimal 2016)) reflect causes such as the uncertainty in PSD measurements and inclusion cases where the PSD was non-stationary events (although the event passed our selection criteria). The empirical cumulative density function of λ over the ensemble of rain events and 104 diameter classes indicate a mean value of $3.4 \times 10^{-5} \text{ s}^{-1}$ (with a standard error of $1.1 \times 10^{-6} \text{ s}^{-1}$), a median of $1.9 \times 10^{-5} \text{ s}^{-1}$, and an interquartile range of -2.0×10^{-5} to $7.5 \times 10^{-5} \text{ s}^{-1}$ (Figure 3a, Table 1). To provide further context for these estimates of λ an analysis was conducted in which number concentrations from the 104 size classes were sampled across 95 randomly sampled “no rain” events (that have the same characteristics in terms of duration and stationarity conditions applied above) and used to compute scavenging coefficients. The results indicate a mean value (computed over all diameters and all events) of $4.8 \times 10^{-6} \text{ s}^{-1}$ (with a standard error of $2.8 \times 10^{-6} \text{ s}^{-1}$), and a median of $-1.0 \times 10^{-7} \text{ s}^{-1}$ (interquartile range: -7.7×10^{-6} to $1.0 \times 10^{-5} \text{ s}^{-1}$). Thus, the mean- λ for non-rain events is almost an order of magnitude below that obtained for our rain event ensemble, and the median is over an order of magnitude smaller than that computed for the rain events. This analysis thus further indicates that, while there is considerable event-to-event variability in derived λ estimates, the estimates derived herein are likely reflective of physical removal of particles by below-cloud scavenging. Further, although as in the study of (Zikova and Zdimal 2016) the interquartile range of $\lambda(D_p)$ spans an order of magnitude for virtually all diameters considered (Figure 3b), only two measured D_p exhibit mean- λ values that are negative

and they are confined to diameters (10.9 nm and 399.5 nm) near the edges of the measured interval for this SMPS where the instrument has lower precision, and for the smaller diameter where the correction for tubing losses is largest.

The magnitude of median- $\lambda(D_p)$ agree with previously reported theoretical predictions (Wang et al. 2010) for the range of rainfall intensities represented in the data set. Also in accord with theoretical considerations and prior experimental research (Andronache et al. 2006; Wang et al. 2010), median scavenging coefficients by particle diameter computed over all 95 rain events decline from $\sim 4.1 \times 10^{-5} \text{ s}^{-1}$ for D_p of 10-19 nm to $\sim 1.6 \times 10^{-5} \text{ s}^{-1}$ for D_p of 80-113 nm, and then show an increasing tendency for $D_p > 200 \text{ nm}$ (Figure 3b).

Scavenging coefficients for all D_p were conditionally sampled by rainfall intensity (Figure 3a) to derive an estimate of the coefficient f in (2) as the slope of the fit between median- λ and rainfall intensity. The results indicate a value of 0.185 (versus 0.244984 in (Laakso et al. 2003)). The other coefficients in (2) were then determined by first averaging the median λ estimates across three size channels (so median $\lambda(D_p)$ is the average value of $\lambda(D_{p(n-1)})$, $\lambda(D_{p(n)})$, and $\lambda(D_{p(n+1)})$) and then fitting (2) using least squares regression (Figure 4). The resulting fit has an adjusted variance explanation (adjusted- r^2) of 0.46, and all values of the coefficients have p-values < 0.10 . No more parsimonious model could be found using stepwise regression. Thus it appears that all the terms in equation (2) contribute to the overall quality of the fit.

Although the fitting coefficients from MMSF exhibit marked discrepancies from those developed by (Laakso et al. 2003), the median $\lambda(D_p)$ values computed from the MMSF data set also exhibit good agreement with the parameterization of (Laakso et al. 2003) (Figure 4). The root mean squared error (RMSE) between λ from the fit derived herein and the observations for the 104 diameter classes is $1.23 \times 10^{-5} \text{ s}^{-1}$ (for the median precipitation intensity of 1.56 mm hr^{-1}),

while for the (Laakso et al. 2003) parameterization is only slightly higher ($1.63 \times 10^{-5} \text{ s}^{-1}$). Thus, the parameterization of median $\lambda(D_p)$ derived by (Laakso et al. 2003) appears to be broadly applicable. Interestingly, in contrast to the convergence with estimates from the boreal forest site in Finland (reported by (Laakso et al. 2003)), for all diameters data from MMSF indicate substantially smaller $\lambda(D_p)$ than were derived using data from a site in the Czech Republic (Zikova and Zdimal 2016) (Figure 4). This discrepancy may reflect the occurrence of higher rainfall intensities at the site in the Czech Republic, differences due to the criteria used for event selection with regards to stationarity, and/or difference in the way in which scavenging coefficients are calculated (described above).

Median- $\lambda(D_p)$ computed from the SMPS3936 with the nano-DMA for a larger set of rainfall events at MMSF ($n = 121$, see Table 1) generally exceed those from the SMPS with a long-DMA (Figure 4). The λ estimates from the nano-DMA SMPS data set at MMSF are also smaller than those from the site in the Czech Republic, but by a lower margin than those from the SMPS with a long-DMA (Figure 4). This may be in part due to the inclusion of more summertime rainfall events in the sample used to compute $\lambda(D_p)$ from the SMPS with the nano-DMA (see Figure 1c). To provide a context for these results computed independently from the two SMPS systems at MMSF (over different rain events), λ estimates for D_p 10-100 nm from the two systems for 87 common rainfall events were also computed. The results indicate relatively good agreement between $\lambda(D_p)$ estimates from the two systems (Table 1, Figure 5) adding support to the inference that the discrepancies shown in Figure 4 reflect differences in the nature of the rainfall events.

4 Concluding remarks

Below cloud scavenging is an important removal process for atmospheric aerosol particles and plays a key role in determining particle concentrations and size distributions. The magnitude of the effect remains uncertain in part because there remain very few robust observational data sets with which to evaluate numerical models of scavenging efficiencies. In situ measurements of particle size distributions and rainfall intensities collected over a 30-month period are used to develop a data set of rain events from which scavenging efficiencies for particle diameters between 10 and 400 nm are derived. The ensemble mean scavenging efficiency is $3.4 \times 10^{-5} \text{ s}^{-1}$ (and has a standard error of $1.1 \times 10^{-6} \text{ s}^{-1}$), while the median is $1.9 \times 10^{-5} \text{ s}^{-1}$ (interquartile range: -2.0×10^{-5} to $7.5 \times 10^{-5} \text{ s}^{-1}$). The median scavenging coefficients by particle diameter exhibit good agreement with the empirical expression formulated by (Laakso et al. 2003).

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346 *Table 1. Summary of the data sets from which scavenging coefficients were computed for MMSF*
347 *and the resulting statistics.*

Statistic ↓ /Instrument ⇒	SMPS3936: Long-DMA	SMPS3936: Nano-DMA	Common events Long- DMA	Common events Nano- DMA
D_p range (nm)	10-400	6-100	10-100	
# rainfall events	95	121	87	
Mean rainfall intensity (mm hr ⁻¹) (median)	2.47 (1.56)	2.46 (1.59)	2.55 (1.48)	
Mean- λ (10 ⁻⁵ s ⁻¹) (standard error)	3.4 (0.11)	3.5 (0.32)	4.6 (0.25)	3.2 (0.28)
Median- λ (10 ⁻⁵ s ⁻¹) (IQR)	1.9 (-2.0 to 7.5)	5.7 (-1.7 to 11)	2.6 (-1.5 to 8.8)	2.2 (-2.3 to 8.1)
RMSE of median- $\lambda(D_p)$ versus fit (10 ⁻⁵ s ⁻¹)	1.23	1.26		
RMSE of median- $\lambda(D_p)$ versus fit of Laakso et al. (10 ⁻⁵ s ⁻¹)	1.63	3.95

Figure 1. Overview of meteorological conditions and particle number concentrations at MMSF during January 2007-April 2009. (a) Hourly average air temperature ($^{\circ}\text{C}$) (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 ($\# \text{ cm}^{-3}$) 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).

Figure 2 (a) Summary of the intensity (in mm hr^{-1}) and duration of the 95 rain events from which scavenging coefficients are computed for the SMPS3936 with the long-DMA (shown by the bars). Also shown are the intensities of the rainfall events (dots) included in the analysis of data from the SMPS with a nano-DMA ($n=121$). (b) Histogram of the correction factor (C_w) for wind-related under-catch by the rain gauge computed from $C_w = \exp[-0.001 \times \ln(R) - 0.0122 \times U \times \ln(R) + 0.034 \times U + 0.0077]$, where U is the wind speed (m s^{-1}), and R is the measured rainfall rate (mm hr^{-1}) (Hildebrandt et al. 2007).

Figure 3 (a) Empirical cumulative distribution plot of scavenging coefficients (λ) for each of the 104 diameters (D_p) reported by the SMPS3936 with the long-DMA for the ensemble of 95 rain events at MMSF (denoted by All in the legend). Also shown are cumulative distributions of λ values conditionally sampled by three rain intensity classes (in mm hr^{-1}). The numbers shown in parentheses are the fraction of the total number of valid λ values in that rainfall intensity class. Thus, results from the highest rainfall intensity class are the least robust since they derive from a small sample size. (b) Mean, median and inter-quartile range of scavenging coefficients (λ) for each of the 104 diameters (D_p) reported by the SMPS fitted with the long-DMA.

Figure 4 Median scavenging coefficients (λ) as a function of particle diameter (D_p) at MMSF as used in the fitting procedure (i.e. averaged over three adjacent size classes) (Obs). Also shown is a fit from equation (2) (the adjusted variance explanation (r^2) for this fit is 0.46), and the median scavenging coefficients as computed from the SMPS with the nano-DMA (Obs-nano). The lines denote results of the parameterization of (Laakso et al. 2003) for two different precipitation rates ($p = 0.78 \text{ mm hr}^{-1}$ and 2.84 mm hr^{-1}). The asterisks show median scavenging coefficients as presented for 311 rain events by (Zikova and Zdimal 2016) for selected particle diameters. The fitting coefficients for equation (2) from MMSF and those from (Laakso et al. 2003) (shown in parentheses in the format provided in the original reference) are as follows: $a = 3.4549076 \times 10^3$ (274.35758), $b = 1.0040252 \times 10^7$ (332839.59273), $c = 5.484421066 \times 10^6$ (226656.57259), $d = 1.12147496 \times 10^6$ (58005.91340), $e = 1.01768617 \times 10^5$ (96588.38582), $f = 0.185$ (0.244984).

Figure 5 Median scavenging coefficients (λ) as a function of particle diameter (D_p) for diameters between 10 and 100 nm, for all 87 events for which data from the SMPS with the Long-DMA (SMPS-Long DMA) and SMPS with nano-DMA (SMPS-Nano DMA) are both available. See Table 1 for the statistics of these events.