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Uncertainty assessment of current size-resolved parameterizations for below-cloud particle scavenging by rain

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Abstract

A detailed review has been conducted of current size-resolved parameterizations of below-cloud scavenging by rain, including their formulation in terms of scavenging coefficient (Λ), their associated input parameters and comparisons with size-resolved Λ values obtained from field measurements. The three dominant factors in the theoretical formulations of Λ – raindrop-particle collection efficiency, raindrop number size distribution and raindrop terminal fall velocity – are investigated through numerical sensitivity tests. It is found that the use of different formulations for raindrop-particle collection efficiency can cause uncertainties in the Λ values of nearly one order of magnitude for particles smaller than 3 µm. The use of different formulations of raindrop number size distribution can cause the Λ values to vary by a factor of 3 to 5 for all particle sizes. The uncertainty in Λ, caused by the use of different droplet terminal velocity formulations, is generally smaller than a factor of 2. All of the current theoretical Λ parameterizations, however, underpredict the Λ values by one to two orders of magnitude for particles smaller than 3 µm, compared with most available field measurements or with empirical formulas generated from field observations. The combined uncertainties from known sources are, thus, not enough to explain the large discrepancies between the theoretical and experimental studies, suggesting a need for further investigations of the collection mechanisms through field, laboratory and numerical studies. The differences in the predicted particle concentrations, due to the use of different Λ parameterizations, can be larger than a factor of 10 for ultrafine and coarse particles even after a small amount of rain (e.g., 2–5 mm). The differences for submicron-sized particles can also be larger than a factor of 2 if sufficient rainfall occurs. Lastly, predicted bulk concentrations (integrated over the particle size distribution) from using different theoretical and empirical Λ parameterizations can differ by up to 50% for particle number and by up to 25% for particle mass after just 2–5 mm of rain.

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1 Introduction

Precipitation scavenging of atmospheric aerosol particles is an important removal process that should be included in atmospheric chemical transport models (CTMs) that simulate aerosol particle number and/or mass concentrations. A parameter, known as the scavenging coefficient (Λ), has been used in the aerosol mass continuity equation in those models to represent below-cloud particle scavenging (Seinfeld and Pandis, 2006). Earlier CTMs only dealt with bulk aerosol mass without the complexity of size-resolved number and mass concentrations (Baklanov, 1999; Rasch et al., 2000; Jacobson, 2003, and references therein). In these models, Λ for bulk mass was commonly parameterized as a function of rainfall intensity (e.g., $\Lambda = A\mathrm{R}^B$, where R is rainfall intensity and A and B are empirical constants) (Balkanski et al., 1993; Mircea et al., 2000; Baklanov and Sorensen, 2001; Andronache, 2003). Recently developed atmospheric aerosol CTMs, on the other hand, explicitly consider size-resolved aerosol number and mass concentrations, where Λ is expressed as a function of the particle size (e.g., Gong et al., 2003; Loosmore and Cederwall, 2004; Tost et al., 2006; Henzing et al., 2006). Model inter-comparisons have shown, however, that both bulk and size-resolved precipitation scavenging parameterizations have large uncertainties (Rasch et al., 2000; Textor et al., 2006).

Calculating size-resolved Λ requires knowledge of the size distributions of aerosol particles and raindrops, droplet terminal velocity and raindrop-particle collection efficiency. In the past few decades, a significant number of theoretical and experimental studies have been carried out to investigate Λ and these related parameters (see reviews in Zhang and Vet, 2006; Sportisse, 2007). However, due to the natural variability of aerosol particles and raindrop populations and the complexity of microphysical collection processes between particles and raindrops, there have been no specific analytical expressions recommended for use in defining these parameters. This leads to large uncertainties in the parameterized size-resolved Λ values. For instance, the present theoretical parameterizations for Λ, which take into account detailed microphysical

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removal processes, including Brownian diffusion, interception, inertial impaction, thermophoresis, diffusiophoresis and electrostatic attraction, are still insufficient to explain observed Λ values. In the submicron range, the theoretical Λ values are generally more than one order of magnitude smaller than the measured ones (Chate, 2005). In addition, the various size-resolved Λ parameterizations that have been developed have differing levels of complexity and assumptions, so that the calculated Λ values vary significantly from one scheme to another (e.g., Chate, 2005). To reduce the large uncertainties in atmospheric aerosol CTMs, there is a need to improve the representation of precipitation scavenging of particles.

The purpose of the present study is to assess the uncertainties in available size-resolved Λ parameterizations in a common framework by means of numerical sensitivity tests and comparisons with available measurements. In the following sections, the theory of below-cloud precipitation scavenging of particle and Λ formulations are first briefly introduced. The sensitivity of Λ to a variety of formulations of raindrop-particle collection efficiency, raindrop number size distribution and raindrop terminal velocity is then examined. Next, the performance of current size-resolved parameterizations of Λ are assessed against available measurements. Finally, predicted aerosol number and mass concentrations, both bulk and size-resolved, after a short period of precipitation are discussed as an example to illustrate the impacts of different existing parameterizations, followed by conclusions.

2 Theory of below-cloud precipitation scavenging of particles

The time-dependent removal of aerosol particles by precipitation is commonly described in CTMs as (Seinfeld and Pandis, 2006)

$$\frac{\partial n(t)}{\partial t} = -\Lambda \cdot n(t), \tag{1}$$

where $n(t)$ is the particle number concentration at time t and the scavenging coefficient Λ has units of inverse time. For CTMs that treat size-resolved aerosol particles, Λ should also be a function of particle size. The size-resolved Λ is parameterized as

$$\Lambda\left(d_{\mathrm{p}}\right)=\int_{0}^{\infty}\frac{\pi}{4}\left(D_{\mathrm{p}}+d_{\mathrm{p}}\right)^{2}\left(V(D_{\mathrm{p}})-v(d_{\mathrm{p}})\right)E\left(d_{\mathrm{p}},D_{\mathrm{p}}\right)N\left(D_{\mathrm{p}}\right)dD_{\mathrm{p}},\tag{2}$$

where d_{p} and D_{p} denote particle and raindrop diameters, respectively, $N(D_{\mathrm{p}})$ is the raindrop number size distribution or size spectrum, and $V(D_{\mathrm{p}})$ and $v(d_{\mathrm{p}})$ are the terminal velocities of raindrop and aerosol particles, respectively. $E(d_{\mathrm{p}},D_{\mathrm{p}})$ is the raindrop-particle collection efficiency: a dimensionless parameter that is defined as the ratio of the total number of collisions occurring between a raindrop and particles to the total number of particles in an area equal to the raindrop's effective cross-sectional area (Slinn, 1983; Pruppacher and Klett, 1997; Seinfeld and Pandis, 2006). Usually the collection efficiency is assumed to be equal with the collision efficiency (Slinn, 1983); that is, the collision between a particle and a raindrop is assumed to result in perfect sticking (the sticking efficiency is unity). This assumption seems reasonable for $d_{\mathrm{p}}/D_{\mathrm{p}}\ll 1$ (Pruppacher and Klett, 1997). Equation (2) shows that the main factors affecting size-resolved Λ include raindrop-particle collection efficiency, raindrop number size distribution and raindrop terminal velocity. A discussion about how these factors can be determined follows.

2.1 Raindrop-particle collection efficiency $E(d_{\mathrm{p}},D_{\mathrm{p}})$

The raindrop-particle collection efficiency $E(d_{\mathrm{p}},D_{\mathrm{p}})$ has been investigated extensively in a series of previous studies (e.g., Wang and Pruppacher, 1977; Grover and Pruppacher, 1985; Slinn, 1983; Pruppacher and Klett, 1997; Pinsky and Khain, 2001). Both experimental and theoretical results have shown that $E(d_{\mathrm{p}},D_{\mathrm{p}})$ is the result of the net action of various forces influencing the relative motion of aerosol particles and hydrometeors. For example, particles following the flow streamlines past a raindrop may be captured by Brownian diffusion, interception, or inertial impaction. Interception takes place when a particle follows a flow streamline that comes within a distance of one particle radius ($d_{\mathrm{p}}/2$) of a droplet. Larger particles tend to experience inertial impaction because of their larger inertia, which prevents them from following the rapidly curving streamlines around falling droplets. Interception and inertial impaction are closely related, but interception occurs as a result of particle size neglecting its mass, while inertial impaction is due to particle mass neglecting its size (Seinfeld and Pandis, 2006).

Brownian diffusion, interception and inertial impaction are believed to be the three most important collection mechanisms for below-cloud particle scavenging. However, accurate prediction of the contribution of each collection mechanism to the overall $E(d_p, D_p)$ is still very difficult due to the complicated flow patterns around the falling droplet. In practical application, various simplified or empirical formulas for $E(d_p, D_p)$ have been induced. Slinn (1983) proposed a semi-empirical approximation for $E(d_p, D_p)$ by using dimensional analysis coupled with experimental data. Based on Slinn (1983), we use the expression for $E(d_p, D_p)$ that is summarized by Seinfeld and Pandis (2006, Eq. 20.56):

$$E\left(d_p, D_p\right) = \frac{4}{ReSc}\left[1+0.4Re^{1/2}Sc^{1/3}+0.16Re^{1/2}Sc^{1/2}\right] \\ +4\frac{d_p}{D_p}\left[\frac{\mu_a}{\mu_w}+\left(1+2Re^{1/2}\right)\frac{d_p}{D_p}\right]+\left(\frac{St-St^*}{St-St^*+2/3}\right)^{3/2}, \quad (3)$$

where

$$Re = \frac{D_p V(D_p)\rho_a}{2\mu_a}, \quad Sc = \frac{\mu_a}{\rho_a D_{diff}}, \quad D_{diff} = \frac{k_b T_a C_c}{3\pi\mu_a d_p},$$

$$St = \frac{2\tau\left(V(D_p)-v(d_p)\right)}{D_p}, \quad \tau = \frac{\left(\rho_p-\rho_a\right)d_p^2 C_c}{18\mu_a}, \quad St^* = \frac{1.2+\frac{1}{12}\ln(1+Re)}{1+\ln(1+Re)},$$

$$C_c = 1+\frac{2\lambda}{d_p}\left(1.257+0.4\exp\left(-0.55\frac{d_p}{\lambda}\right)\right),$$

and all symbols are defined in Table 4.

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The first term in Eq. (3) represents Brownian diffusion, the second term represents interception, and the third term represents inertial impaction. Note that the third term is included only when the Stokes number *St* is greater than the critical Stokes number St^*. The third term is also valid as written only for aerosol particles with a density of $1\,\mathrm{g\,cm^{-3}}$; otherwise, this term should be scaled by $\left(\rho_\mathrm{p}/\rho_\mathrm{w}\right)^{1/2}$ (Slinn, 1983; Seinfeld and Pandis, 2006). The above analytical expression for $E(d_\mathrm{p}, D_\mathrm{p})$ has been widely used in current parameterizations for below-cloud particle scavenging by rain (e.g., Mircea et al., 2000; Chate et al., 2003; Chate, 2005; Andronache, 2003; Andronache et al., 2006; Gong et al., 2003; Loosmore and Cederwall, 2004; Tost et al., 2006; Henzing et al., 2006; Feng, 2007).

Theoretically, Slinn's formula is likely to underestimate $E(d_\mathrm{p}, D_\mathrm{p})$, since it includes only a subset of the mechanisms that influence particle collection by rain. A number of studies have suggested that thermophoresis, diffusiophoresis and electric charges may increase $E(d_\mathrm{p}, D_\mathrm{p})$ for submicron aerosol particles (e.g., Slinn and Hales, 1971; Grover et al., 1977; Wang et al., 1978; McGann and Jennings, 1991; Byrne and Jennings, 1993; Pranesha and Kamra, 1997; Tripathi and Harrison, 2001; Tinsley et al., 2000; Jaworek et al., 2002; Andronache, 2004; Chate, 2005; Andronache et al., 2006). Thermophoresis, which is caused by uneven heating of particles in ambient temperature gradients, drives particles towards evaporating and sublimating hydrometeors. Diffusiophoresis moves particles towards diffusionally-growing hydrometeors due to water vapour concentration gradients (Chate, 2005). According to Andronache et al. (2006), the thermophoretic and diffusiophoretic contributions to $E(d_\mathrm{p}, D_\mathrm{p})$ can be expressed, respectively, as follows:

$$E_\mathrm{th}\left(d_\mathrm{p}, D_\mathrm{p}\right) = \frac{4\alpha_\mathrm{th}\left(2+0.6Re^{1/2}\mathrm{Pr}^{1/3}\right)\left(T_\mathrm{a}-T_\mathrm{s}\right)}{V\left(D_\mathrm{p}\right)D_\mathrm{p}}, \tag{4}$$

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$$E_{\mathrm{dph}}\left(d_{\mathrm{p}},D_{\mathrm{p}}\right)=\frac{4\beta_{\mathrm{dph}}\left(2+0.6Re^{1/2}Sc_{\mathrm{w}}^{1/3}\right)\left(\frac{P_{\mathrm{s}}^{0}}{T_{\mathrm{s}}}-\frac{P_{\mathrm{a}}^{0}\mathrm{RH}}{T_{\mathrm{a}}}\right)}{V\left(D_{\mathrm{p}}\right)D_{\mathrm{p}}}, \tag{5}$$

where

$$\alpha_{\mathrm{th}}=\frac{2C_{\mathrm{c}}\left(k_{\mathrm{a}}+5\lambda/D_{\mathrm{p}}k_{\mathrm{p}}\right)k_{\mathrm{a}}}{5P\left(1+6\lambda/D_{\mathrm{p}}\right)\left(2k_{\mathrm{a}}+k_{\mathrm{p}}+10\lambda/D_{\mathrm{p}}k_{\mathrm{p}}\right)}, \quad \mathrm{Pr}=\frac{c_{\mathrm{p}}\mu_{\mathrm{a}}}{k_{\mathrm{a}}},$$

$$\beta_{\mathrm{dph}}=\frac{T_{\mathrm{a}}\,D_{\mathrm{diffwater}}}{P}\sqrt{\frac{M_{\mathrm{w}}}{M_{\mathrm{a}}}}, \quad \text{and} \quad Sc_{\mathrm{w}}=\frac{\mu_{\mathrm{a}}}{\rho_{\mathrm{a}}\,D_{\mathrm{diffwater}}}.$$

The contribution of an electric charge to the collection efficiency is based on the concept that a raindrop with a charge Q_{r} attracts an aerosol particle with an opposite charge q_{p} and this process enhances the capture efficiency by the raindrop of aerosol particles close to the raindrop's surface (Andronache, 2004). The electrostatic collection efficiency is expressed as

$$E_{\mathrm{es}}\left(d_{\mathrm{p}},D_{\mathrm{p}}\right)=\frac{16KC_{\mathrm{c}}Q_{\mathrm{r}}q_{\mathrm{p}}}{3\pi\mu_{\mathrm{a}}\,V\left(D_{\mathrm{p}}\right)D_{\mathrm{p}}^{2}d_{\mathrm{p}}}, \tag{6}$$

where $K=9\times10^{9}$ (in $\mathrm{Nm^{2}\,C^{-2}}$) and Q_{r} and q_{p} are the mean charges on the raindrop and on the aerosol particle (in Coulomb, C) and are assumed to be of opposite sign. A parameterization, with respect to size, has been proposed for the mean raindrop and particle charges:

$$Q_{\mathrm{r}}=a\alpha D_{\mathrm{p}}^{2}, \text{ and } q_{\mathrm{p}}=a\alpha d_{\mathrm{p}}^{2}, \tag{7}$$

where $a=0.83\times10^{-6}$ and α ($\mathrm{C\,m^{-2}}$) is an empirical parameter that can vary between 0, which corresponds to neutral particles and 7, which corresponds to highly electrified clouds associated with thunderstorms (Andronache, 2004; Andronache et al., 2006).

2.2 Raindrop number size distribution $N(D_p)$

Detailed information about the raindrop number size distribution is essential for understanding the mechanism of below-cloud particle scavenging, estimating the scavenging coefficient Λ and improving microphysical parameterizations in numerical weather models and CTMs. Since the pioneering studies of Marshall and Palmer (1948), extensive research has been devoted to modelling the raindrop size distribution (e.g., Ulbrich 1983; Feingold and Levin, 1986), and various mathematical functions have been proposed to fit the observed number distributions for raindrops. However, almost no guidance is available to recommend a specific function and its parameters for use in characterising natural raindrop size spectra because various factors such as rainfall intensity, precipitation type (e.g., stratiform rain, convective rain, thunderstorm), and the stage of rain development all contribute to the formation and evolution of the raindrop size distribution (e.g., Waldvogel, 1974; Sauvageot and Lacaux, 1995; Brandes et al., 2006; Zhang et al., 2008).

At present, the main mathematical functions used to represent the raindrop number size distribution can be divided into three types based on their formulas: exponential distribution; gamma distribution; and lognormal distribution. The exponential distribution is generally written as (e.g., Marshall and Palmer, 1948)

$$N(D_p) = N_{0e} \exp\left(-\beta_e D_p\right), \tag{8}$$

where N_{0e} is the intercept parameter and β_e is a slope parameter. The general form of the gamma distribution can be written as (e.g., Ulbrich, 1983)

$$N(D_p) = N_{0g} D_p^{\gamma} \exp\left(-\beta_g D_p\right) \tag{9}$$

Here N_{0g} is a number concentration parameter, γ is a distribution shape parameter and β_g is a slope term sensitive to the larger particles. The general form of the lognormal distribution can be written as (e.g., Feingold and Levin, 1986; Cerro et al., 1997)

$$N\left(D_\mathrm{p}\right) = \frac{N_\mathrm{total}}{\sqrt{2\pi}D_\mathrm{p}\log\sigma_\mathrm{D}}\exp\left[-\frac{\left(\log D_\mathrm{p} - \log\bar{D}_\mathrm{p}\right)^2}{2\log^2\sigma_\mathrm{D}}\right], \tag{10}$$

where N_total is the total droplet number density, $\bar{D}_\mathrm{p}$ is the mean droplet diameter and σ_D is the droplet-diameter standard deviation. These three parameters of the lognormal function are expressed as functions of rainfall intensity. In general, gamma and log-normal distributions seem to be better in representing the characteristics of observed raindrop size distributions, especially at the small-particle end than exponential distributions (Mircea and Stefan, 1998; de Wolf, 2001; Mircea et al., 2000; Bae et al., 2006).

These three distribution functions have been widely used in the parameterization of size-resolved below-cloud scavenging. Since this type of parameterization considers the full set of interactions between the size spectra of raindrops and aerosol particles, the numerical calculation of Λ is very complex and computationally intensive. To reduce the computational burden, some large-scale atmospheric models represent the raindrop size spectrum with a representative raindrop diameter D_r (i.e., a monodisperse distribution), generally the median volume diameter (e.g., Gong et al., 2003, 2006; Loosmore and Cederwall, 2004; Tost et al., 2006). Since all raindrops are assumed to have the same diameter D_r, the integral form of Eq. (2) can then be simplified to

$$\Lambda\left(d_\mathrm{p}\right) = \frac{\pi}{4}D_\mathrm{r}^2 V\left(D_\mathrm{r}\right)E\left(d_\mathrm{p}, D_\mathrm{r}\right)N_\mathrm{total}. \tag{11}$$

As the rainfall intensity R (in mm s^{-1}) can be defined by the expression

$$\mathrm{R} = \int_0^\infty \frac{\pi}{6}D_\mathrm{p}^3 V\left(D_\mathrm{p}\right)N\left(D_\mathrm{p}\right)dD_\mathrm{p}, \tag{12}$$

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then for a monodisperse raindrop number size spectrum, the rainfall intensity can be written as

$$R = \frac{\pi}{6} D_r^3 V(D_r) N_{\text{total}}. \tag{13}$$

Combining Eqs. (11) and (13), Λ can then be rewritten for a monodisperse raindrop size spectrum as

$$\Lambda\left(d_p\right) = \frac{3}{2} \frac{E\left(d_p, D_r\right) R}{D_r}. \tag{14}$$

2.3 Raindrop terminal velocity $V(D_p)$

The terminal fall velocity of a raindrop is another parameter that is included in the formula of the below-cloud scavenging coefficient (see Eq. 2). Two general approaches have been employed to describe raindrop terminal velocity in below-cloud scavenging parameterizations: (1) empirical formulas derived directly from experimental data and (2) physically-based parameterizations. Table 1 lists some commonly used empirical formulas for the terminal velocity of falling raindrops.

Physically-based analytical expressions usually divide the population of raindrops into several size ranges that correspond to different, physically-distinct flow regimes (e.g., Beard, 1976; Seinfeld and Pandis, 2006; Jacobson, 2005). Different analytical expressions are employed in different ranges. In the present study, we follow the theoretical formula of Beard (1976) for the most calculations. Beard's scheme assigns each raindrop to one of three physically-distinct flow regimes: Stokes's regime ($D_p \leq 20\,\mu m$ or $Re \leq 0.01$); the transitional regime ($20\,\mu m \leq D_p \leq 1\,mm$ or $0.01 \leq Re \leq 300$); and Newton's regime ($1\,mm \leq D_p \leq 7\,mm$ or $300 \leq Re \leq 4000$). For raindrops in Stokes's regime, the Beard scheme explicitly calculates the terminal velocities using Stokes's formula

$$V(D_p) = \frac{D_p^2(\rho_w - \rho_a) g C_c}{18\mu_a}, \tag{15}$$

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where ρ_a is air density, ρ_w is water density, μ_a is air viscosity, g is the gravitational constant, D_p is the raindrop diameter and C_c is the Cunningham correction factor. However, for larger raindrops ($D_p \geq 20$ µm or $Re \geq 0.01$), the Stokes's formula is no longer valid and there are no explicit expressions for the terminal velocities. In this case, the Beard scheme calculates a Best number, which is based upon the droplet mass and density as well as the gravitational constant and the air viscosity. Then observations are used to derive the Reynolds number from the Best number. Finally, the terminal velocities can be derived using the definition of the Reynolds number. Expressions for the Best number and empirical relations for the Reynolds number in terms of the Best number are given by Beard (1976) and Jacobson (2005).

3 Sensitivity of Λ to different formulations

As described in Sect. 2, there exists a number of different formulas for raindrop-particle collection efficiency, raindrop number size distribution and raindrop terminal velocity, the three factors needed to calculate size-resolved Λ (see Eq. 2). This Section focuses on investigating the sensitivity of Λ to these different formulas.

3.1 Sensitivity to collection efficiency

As discussed in Sect. 2.1, the raindrop-particle collection efficiency $E(d_p, D_p)$ is controlled by many different microphysical processes. Figure 1 shows the contributions of six microphysical processes to $E(d_p, D_p)$ for a wide size range of particles collected by a raindrop 1 mm in diameter. Calculations are performed based on Eqs. (3)–(6), where (a) raindrop terminal velocities $V(D_p)$ are computed from the theoretical formula of Beard (1976), (b) a 3 °C temperature difference has been assumed between the raindrop and ambient air to calculate the thermophoretic and diffusiophoretic collection efficiencies (Slinn and Shen, 1970; Slinn and Hales, 1971; Chate, 2005), and (c) $\alpha=2$ has been assumed for average conditions of electrified clouds to calculate electrostatic collection efficiency (Andronache, 2004).

Clearly, the contribution of Brownian diffusion decreases rapidly as particle size increases. It is the most important collection mechanism for smaller particles, particularly ultrafine particles (d_p<0.01 µm), but it contributes little for supermicron particles. Inertial impaction, by contrast, can only occur for particles with Stokes number St above the critical Stokes number St^* that is close to 1.2 (Phillips and Kaye, 1999; Loosmore and Cederwall, 2004); the corresponding threshold diameter is close to 3 µm for unit-density particles and a 1 mm raindrop. Figure 1 shows that the contribution of inertial impaction dominates $E(d_p,D_p)$ for particles larger than 3.5 µm. The contribution of interception increases with increasing particle size and appears to be important for particles in the 1 to 3.5 µm size range. Thermophoresis makes a comparable contribution to Brownian diffusion for particles with d_p between 0.1 and 1 µm. The contribution of diffusiophoresis is smaller than that of thermophoresis for all particle sizes. Finally, the contribution from electric charges increases with particle size and is dominant for particles with d_p between 0.3 and 3.5 µm.

Because of the combined action of the microphysical processes discussed above, total raindrop-particle collection efficiency varies significantly for different particle sizes. Figure 1 indicates that the collection efficiency is largest for ultrafine particles (d_p<0.01 µm) due to Brownian diffusion and for large particles (d_p>3 µm) due to inertial impaction. However, for submicron particles, although more of these mechanisms, i.e., Brownian diffusion, interception, diffusiophoresis, thermophoresis and electric charges, play a role in the collection process, the overall $E(d_p,D_p)$ is very low (<10^{-2}).

Figure 2 shows a contour plot of raindrop-particle collection efficiency as a function of both raindrop and particle size calculated using the same conditions as in Fig. 1. It is clear from this figure that the collection efficiency decreases with increasing raindrop size for aerosol particles smaller than 3 µm in diameter. The reason is that the dominant collection mechanisms for these particles (i.e., Brownian diffusion for ultrafine particles and Brownian diffusion, interception, thermophoresis, diffusiophoresis and electric charges for aerosol particles between 0.01–3 µm) become less efficient as raindrop size increases (see Eqs. 3–6) because (a) small raindrops have lower terminal velocities and, hence, more time for interactions with nearby particles and (b) small raindrops have more neighbouring aerosol particles relative to their effective cross sections than do large raindrops. In contrast, the collection efficiency for large particles (d_p>3 µm) is not very sensitive to raindrop size. This is due to the fact that inertial impaction dominates collection in this size range and this process has little dependence on raindrop size (see Eq. 3).

Figure 3 shows the sensitivity of the size-resolved scavenging coefficient to three different $E(d_p,D_p)$ formulations for two different rainfall intensities, 1 and 10 mm h^{-1}, based on calculations using Eq. (2). The raindrop terminal velocity and raindrop size spectrum were parameterized using the Beard scheme and the Marshall-Palmer (MP) distribution, respectively. Figure 3 indicates that the addition of the collection processes of thermophoresis, diffusiophoresis and electrostatic forces can significantly enhance the below-cloud scavenging of aerosol particles smaller than 3 µm, particularly for particles in the 0.1–2.0 µm size range. For larger particles, these processes have much less effect since inertial impaction dominates over the other mechanisms.

Note that many early studies used a constant collection efficiency in parameterizing Λ (Scott, 1982; Mircea and Stefan, 1998), for comparison purposes, such a case is also shown in Fig. 3. Since raindrops are generally much bigger than aerosol particles and raindrop terminal velocities are generally much bigger than particle settling velocities, then $(D_p+d_p)^2 \cong D_p^2$ and $V(D_p) \gg v(d_p)$ so that $E(d_p,D_p)$ becomes the only parameter in Eq. (2) that is a function of the collected particle diameter d_p. Therefore, if $E(d_p,D_p)$ is assumed to be a constant for a given raindrop size distribution, then Λ will be close to a constant for all particle sizes and will only change with rainfall intensity R. Clearly, this method does not reflect the reality evident in Fig. 3 since it neglects the dependence of below-cloud scavenging processes on particle size.

3.2 Sensitivity to raindrop size distribution

As discussed in Sect. 2.2, a number of different empirical formulas for the raindrop size spectrum have been used in Λ parameterizations in previous studies. To investigate

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the sensitivity of Λ to the choice of raindrop size spectrum, eight empirical formulas were selected to calculate Λ using Eq. (2). As shown in Fig. 4, these included four exponential distributions (MP, JD, JT and ZH), two gamma distributions (DE and W84), and two lognormal distributions (FL and CE). An immediate observation from Fig. 4 is that the four exponential distributions yield greater numbers of small droplets compared to the other distributions. For example, according to Table 2 the percentage of droplets smaller than 0.1 mm in size for the MP distribution is >65% in drizzle (0.01 mm h^{-1}), 33% in moderate-intensity rain (1 mm h^{-1}), and still 14% even in extremely heavy rain (100 mm h^{-1}). By contrast, the gamma and lognormal distributions have many fewer small droplets and the differences between the gamma and lognormal distributions are not as large, although the gamma distribution has a wider droplet size range than the lognormal distribution. As precipitation intensity increases, the modes of all the distributions shift to larger drops (Fig. 4). The total droplet number concentration (N_{total}) also increases with increasing rainfall intensity (Table 2). For a given rainfall intensity, the N_{total} value predicted by the MP exponential distribution exceeds those predicted by the DE gamma distribution and FL lognormal distribution by an order of magnitude.

The significant differences amongst different raindrop size spectra representations should affect the calculated size-resolved Λ values. A study by Mircea et al. (2000) found only a weak sensitivity of Λ to the raindrop size distribution. However, the two droplet spectra used by Mircea et al. (2000) were quite similar in nature; they were both derived from measurements in the same area (Mediterranean) and were both lognormal distributions. Figure 5 shows a comparison of size-resolved Λ curves derived from the eight different raindrop size spectra considered in Fig. 4 for two different rainfall intensities (0.1 and 10 mm h^{-1}). Note that the terminal fall velocities and collection efficiencies used in the calculations for this figure followed the theoretical formulas of Beard (1976) and Slinn (1983), respectively.

Clearly, Λ depends strongly on the choice of raindrop size distribution. Λ values, calculated using different raindrop size spectra, can differ by a factor of 3 to 5 depending on particle size and precipitation intensity. The differences are more evident for particles smaller than 3 µm under weak rainfall intensities. The MP and the JD raindrop size distributions give higher Λ values than other distributions for all particle sizes for two reasons: (a) their higher droplet total number concentrations and (b) their greater fraction of small droplets, which have higher collection efficiencies (see Table 2 and Fig. 2). As rainfall intensity increases, however, the differences in droplet total number concentration and fraction of small droplets between the different raindrop size spectra decrease (see Table 2) because the modes of all distributions shift to larger droplet sizes (see Fig. 4). Thus, differences in Λ from using different raindrop size spectra decrease with increasing rainfall intensity.

Also shown in Fig. 5 are Λ profiles calculated using two monodisperse raindrop size distributions. One is used in AURAMS (A Unified Regional Air-quality Modelling System), where the typical raindrop diameter D_p is assumed to be D_p=0.7R$^{0.25}$ (with D_p in mm and R in mm h^{-1}) (Gong et al., 2003, 2006; hereafter referred to as AURAMS-Monodisperse), and the other is used in NARAC/LLNL (National Atmospheric Release Advisory Center system of the Lawrence Livermore National Laboratory), where D_p=0.97R$^{0.158}$ (Loosmore and Cederwall, 2004; hereafter referred to as LC-Monodisperse). The Λ values calculated using the monodisperse distributions (i.e., based on a representative droplet diameter, see Eq. 14) are similar to those obtained from using polydisperse droplet spectra. On the other hand, the uncertainty in Λ from using different "representative" drop diameters is as large as using different polydisperse droplet size spectra. It is noted that the LC-Monodisperse "representative droplet diameter" was generated from the W84 droplet size spectrum (Willis, 1984). Interestingly, Λ values from using these two distributions are very close, suggesting that the use of a monodisperse raindrop size distribution can be a reasonable assumption as long as the proper representative diameter is chosen.

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3.3 Sensitivity to raindrop terminal velocity

Figure 6 shows the terminal fall velocities of rain droplets derived from the six empirical formulas listed in Table 1 and the Beard theoretical scheme as well as the measurements reported by Gunn and Kinzer (1949). Note that the formulas from Atlas et al. (1973) and Brandes et al. (2002) give negative values when a droplet has a diameter smaller than 0.1 mm and 0.02 mm, respectively. It can be seen that most formulas, except the power law formula of Kessler (1969), agree well with the experimental data for droplets in the 0.5 mm to 5 mm size range. For droplets larger than 5 mm, however, different formulas produce quite different terminal velocities (e.g., ~20–50% difference depending on size), but droplets at these sizes are very rare. For droplets smaller than 0.5 mm (i.e., in the Stokes and lower transitional regimes), most formulas overestimate the fall speed. Noted that the theoretical formula of Beard (1976) agrees best with the experimental data.

The influence of different $V(D_p)$ formulas on the Λ values is illustrated in Fig. 7 for a rainfall intensity R of 1 mm h^{-1}. The droplet size spectrum used in this calculation was the MP distribution and the collection efficiency formulation was Slinn's (1983) scheme. It can be seen that the uncertainty in Λ, from using different $V(D_p)$ formulas, is generally within a factor of 2 for any particle size, much smaller than uncertainties caused by the choice of different droplet-particle collection efficiencies and different droplet size spectra. The formulas that give higher $V(D_p)$ values also tend to give higher Λ values. For example, the MP droplet distribution has a large number of small drops. Since the Kessler (1969) formula predicts the largest $V(D_p)$ values for small droplets, it also produces the largest Λ values (compare Fig. 7 with Fig. 6). Similar results to the above have also been obtained for gamma and lognormal droplet size spectra.

4 Evaluation of existing size-resolved Λ parameterizations

Available size-dependent Λ parameterizations existing in the literature can be classified into three types based on how they were developed. The first type (hereafter referred to as Type I) calculates Λ based on analytical formulas for raindrop-particle collection efficiency, raindrop size distribution, and raindrop terminal velocity (e.g., Slinn,1983; Mircea et al., 2000; Chate et al., 2003; Chate, 2005; Andronache, 2003; Andronache et al., 2006; Jung et al., 2003; Gong et al. 2003; Loosmore and Cederwall, 2004; Tost et al., 2006; Feng, 2007; Henzing et al., 2006). The second type (hereafter referred to as Type II) employs an empirical fit of pre-calculated Λ values that were generated using a Type I method with an assumed droplet size spectrum and other needed inputs. For example, Henzing et al. (2006) developed a simple size-dependent Λ parameterization based on a three-parameter fit to a set of pre-calculated Λ values (see details in Appendix A1). Thus, Λ values from Type II parameterizations should be similar to those from Type I parameterizations. A major advantage of Type II methods should be a significant reduction in computational burden, since they need only evaluate a simple fitting function rather than performing an explicit integration over the raindrop size spectrum. However, one possible drawback of this method is that it might only be valid for certain rain droplet spectra. The third type of Λ parameterization (hereafter referred to as Type III) uses an empirical fit to Λ values derived from field measurements (Laakso et al., 2003; Baklanov and Sorensen, 2001). Table 3 lists some available size-resolved Λ parameterizations classified by these three types.

Figure 8 shows a comparison of predictions from these 10 parameterizations with each other and with available field measurements for three different rainfall intensities. The majority of the field data display a strong dependence on the Λ values on particle size. Below-cloud scavenging is very fast for particles larger than a few microns in size, quite fast for particles smaller than 0.01 µm in size, and slowest for submicron particles. The measurement data also have a large spread, which is probably due to the very different experimental conditions between these field studies. In general,

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Λ values were determined by measuring concentration changes of the size-resolved aerosol particle at ground stations before, during and after rain events (e.g., Davenport and Peters, 1978; Slinn, 1983; Volken and Schumann, 1993; Laakso et al., 2003; Chate and Pranesha, 2004). Besides the measurement errors caused by the instruments and analysis processes, many other physical (horizontal and vertical advection and turbulent diffusion), microphysical (condensation, nucleation, coagulation and hygroscopic growth), and chemical (both gas- and aqueous-phase chemistry) processes can modify particle concentrations concurrently and, thus, contribute to the large uncertainties in the measured Λ values. Since the submicron particles have the smallest Λ values, it is not surprising that the largest spread of Λ values was also observed for this size range due to the various processes mentioned above.

To eliminate some of the uncertainties existing in the natural environment, a so-called outdoor experiment was designed by Sparmacher et al. (1993) to determine the below-cloud snow and rain scavenging coefficients using monodisperse artificial particles. In this experiment, monodisperse artificial aerosol particles from an aerosol generator were fed into a wind-shielded measuring chamber and suspended. Natural precipitation that fell through the chamber then scavenged the aerosol particles and Λ values were then determined by measuring the changes of particle concentrations inside the chamber. The objective of the design of the experiment was to eliminate as many confounding factors during the scavenging process as possible so that the Λ values observed would represent the actual droplet-particle collection processes (Sparmacher et al., 1993). The Λ values for four selected particle sizes (0.23, 0.46, 0.98, and 2.16 µm in diameter) generated from this study are also shown in Fig. 8. Interestingly, Λ values from the "outdoor" experiment are much lower than those determined from other field observations, suggesting that the above-mentioned physical and chemical processes do contribute substantially and on many occasions play a dominant role, resulting in much higher measured Λ values.

Figure 8 shows that for large particles (d_p>3 µm) the Λ values from the theoretical parameterizations (Types I and II) agree reasonably well with field measurements under various rainfall intensities. However, for submicron particles all theoretical parameterizations underestimate observed Λ values by up to two orders of magnitude and by one order of magnitude for particles smaller than 0.1 µm compared to available measurements, except for the measurements from the "outdoor" experiment by Sparmacher et al. (1993). Although, as discussed in Sect. 3 and also shown in Fig. 8, the uncertainty in the theoretical Λ parameterizations can be larger than one order of magnitude due to various input selections, it is insufficient to explain the nearly two-orders-of-magnitude underestimation for submicron particles. Figure 8 also suggests that the current theoretical parameterizations (Types I and II) seem to be able to correctly predict the droplet-particle collection process in an ideal vertical flow created by raindrops as can be seem from the good agreement between theoretical values and the controlled experiment of Sparmacher et al. (1993). However, in the real world, other processes mentioned above appear to play dominant roles in the overall scavenging process as can be seen from the much higher Λ values found in most field measurements compared to the controlled experiment of Sparmacher et al. (1993).

One example of an important neglected process may be the enhanced scavenging due to turbulent flow fluctuations (e.g., Grover and Pruppacher, 1985; Khain and Pinsky, 1997). More recently, Andronache et al. (2006) developed a simplified scavenging model for ultrafine particles that includes below-cloud scavenging processes, mixing of ultrafine particles from the boundary layer into cloud, cloud condensation nuclei activation, and in-cloud removal by rainfall. They applied the model to the observed data of Laakso et al. (2003) and found good agreement in the overall particle scavenging, suggesting the possible important role of turbulent mixing, cloud droplet activation, and in-cloud scavenging on the observed below-cloud scavenging. More theoretical and field studies of particle removal mechanisms are still clearly needed so the large discrepancies between theoretical and observed results can be reduced.

The empirical formulas derived from measurements (Type III) fit well to the data from which the formulas were generated but not necessarily to other data sets. The parameterization of Baklanov and Sorensen (2001) (see details in Appendix A2) overestimated the Λ values for particles smaller than 3 µm in diameter when compared with the measurements of Laakso et al. (2003), Volken and Schumann (1993) and Sparmacher et al. (1993). This parameterization treats the Λ as a function of rainfall intensity R only and neglects its dependence on particle size for particles smaller than 2.8 µm (Appendix A2), whereas most measurements of Λ show a strong dependence on particle size (e.g., Fig. 8). The empirical formula of Laakso et al. (2003) (see details in Appendix A3) agrees well with most of the observational data; however, this parameterization is only valid for particles with sizes of 0.01–0.5 µm and for rainfall intensities of 20 mm h^{-1} or less. Inspection of Fig. 8 suggests that this parameterization overestimates Λ values for ultrafine particles (d_p<0.01 µm) and for particles larger than 10 µm. Thus, this comparison also suggests that current empirical parameterizations for Λ need to be developed further and need to be verified against new sets of measurement data across a range of different conditions.

5 Impacts of the various Λ parameterizations on below-cloud particle removal

The ultimate goal of parameterizing Λ is to use the parameterization to predict particle mass and number concentration changes through the precipitation scavenging process in aerosol transport models. We can expect uncertainties in representing Λ to introduce corresponding uncertainties on the predicted particle concentrations. In this section, two different aerosol particle size distributions, representing marine and urban aerosols, are taken as examples to investigate the impact of different Λ parameterizations on predicted particle concentrations. The initial size distribution for each aerosol type is described as a sum of three lognormal functions according to Jaenicke (1993).

Figure 9 shows the time evolution over five hours of the bulk particle number and mass concentrations (integrated over the entire particle size spectrum) as calculated using seven different Λ parameterizations from Table 3 and assuming a constant rainfall intensity R of 1 mm h^{-1} (i.e., light to moderate rain). The five solid lines correspond to the five theoretical Λ parameterizations and the two dotted lines correspond to the

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two empirical Λ parameterizations. Differences in the bulk number concentrations predicted by the five theoretical Λ parameterizations are generally within 5% after 2 mm of rain and within 15% after 5 mm of rain for both marine and urban aerosols. However, the bulk mass concentrations predicted by these five schemes can differ by 20% after 2 mm of rain. In contrast, the two empirical Λ parameterizations predict much faster removal compared to the theoretical ones. Given that the empirical parameterization of Baklanov and Sorensen (2001) overestimates Λ values for particle diameters smaller than 3 µm as discussed in Sect. 4, the results generated from the empirical formula of Laakso et al. (2003), which seems to be better at representing the observed Λ values, are used in the following quantitative comparisons. It is found that the number concentrations predicted by the five theoretical Λ parameterizations are 40–50% higher than those from the Laakso et al. (2003) empirical Λ scheme after just 2–5 mm of rain, while the mass concentrations are 15–25% higher.

The above analyses also indicate that the impacts of using different Λ parameterizations are qualitatively different for the bulk number and mass concentrations. This is because the bulk mass concentration is generally associated with large particles, whereas the bulk number concentration is associated with small particles, as can be seen from the initial particle size distributions shown in Fig. 10. Clearly, for both marine and urban aerosol distributions, particles smaller than 0.1 µm dominate the bulk number concentration while particles larger than 1 µm dominate the bulk mass concentration. Thus, uncertainties in the Λ parameterizations for very small particles contribute to the uncertainties in the predicted bulk number concentration, whereas uncertainties in Λ for coarse particles contribute to the uncertainties in the predicted bulk mass concentration, as discussed above for Fig. 9.

Figure 10 also shows the predicted size-resolved aerosol number and mass concentrations after two hours of precipitations (using the same conditions as in Fig. 9). The largest differences were found for ultrafine particles (d_p<0.01 µm) and for coarse particles. The differences for the particles in the size range of 0.01 µm to 3 µm are generally small, except for the Baklanov and Sorensen (2001) parameterization, which had much higher Λ values than the rest of the parameterizations for this particle size range. This is because the Λ values in this size range are extremely low (10^{-7}–10^{-5} s^{-1}, Fig. 8) so that below-cloud scavenging processes will generally play an insignificant role in the predicted concentrations. However, in the cases of long-lasting rainfalls or very heavy rainfalls, scavenging of these particles might be important.

It is also evident in Fig. 9 that the differences in the predicted bulk concentrations from most Λ parameterizations (except the one of Baklanov and Sorensen, 2001) increase with time during the first hour and then gradually decrease during the next several hours. This is because the majority of very small particles (which dominate number concentration) and large particles (which dominate mass concentration) have been scavenged quickly during the first hour (cf. Fig. 10). This conclusion is also supported by the value of the 0.5-folding time, defined as $t = \ln(2)/\Lambda(d)$, which is about 1 h for coarse particles for R of 1 mm h^{-1} (Andronache, 2003).

6 Conclusions

To identify the sources of uncertainties in the current theoretical Λ parameterizations for below-cloud scavenging of particles by rain in atmospheric chemical transport models, a detailed literary review and set of numerical sensitivity tests in a common framework have been conducted in the present study. The largest uncertainties are associated with the specification of raindrop-particle collection efficiency. Inclusion of the additional collection mechanisms of thermophoresis, diffusiophoresis and electrostatic forces in the droplet-particle collection efficiency calculations can enhance predicted Λ values by nearly one order of magnitude for particle sizes 0.1–2.0 µm. Another large source of uncertainty is associated with the choice of raindrop number size distribution. Various raindrop size distributions can yield Λ values that differ by a factor of 3 to 5 depending on rainfall intensity and particle size. The uncertainty in Λ caused by choosing different raindrop terminal velocity formulations is not negligible but is generally small than a factor 2.

Most current theoretical size-resolved Λ parameterizations predict Λ values that agree well with the available measurements for particles larger than 3 µm. However, for particles smaller than 3 µm, the theoretical Λ parameterizations underpredict Λ values by up to two orders of magnitude in comparison with the majority of field measurements. The combined uncertainty due to the use of different formulations of raindrop-particle collection efficiency, raindrop size spectra, and raindrop terminal velocity in the current theoretical framework is not sufficient to explain the large discrepancies between the theoretical and measured Λ values. This suggests that new collection mechanisms need to be identified and/or that current collection mechanisms need to be modified. Enhanced particle collection due to small-scale turbulence is one candidate. Numerical studies using comprehensive cloud model with explicit aerosol horizontal and vertical transport, turbulent diffusion and detailed size-resolved microphysics are also needed to evaluate the importance of various terms in contributing to the overall below-cloud scavenging.

The two empirical size-resolved Λ parameterizations, that were identified, fit well to the data from which the empirical formulas were generated but not necessarily to other datasets. In addition, they are only valid for a specific range of particle sizes and they may only apply to certain types of precipitation. More scavenging data are needed to develop these empirical parameterizations and to evaluate them under different rainfall conditions. More comprehensive and detailed information during rainfall scavenging events (e.g., aerosol size distribution, raindrop size distribution, electric charge, turbulence intensity) besides what is commonly observed is also needed in order to better understand the scavenging process and to identify new collection mechanisms.

The predicted bulk aerosol number and mass concentrations calculated using different theoretical and empirical Λ parameterizations can differ by 40%–50% and by 15–25%, respectively, after just 2–5 mm of rain. The largest differences in the predicted size-resolved concentration are for ultrafine particles and for coarse particles due to their larger Λ values (i.e., faster removal). However, the impact of different Λ parameterization on predicted concentrations for particles in the 0.01 to 2 µm size range

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could also be important if enough rain occurs.

Another uncertainty (not discussed in the previous sections) related to below-cloud particle scavenging in large-scale aerosol transport models is the choice of the time step in the integration of the number or mass continuity equation (e.g., Eq. 1). If the concentration changes substantially over one model time step, then large errors may then be introduced when solving Eq. (1). This could be the case for very large particles (which control the bulk mass concentration) and very small particles (which control the bulk number concentration) due to their higher Λ values. Sensitivity tests using Λ values presented in Fig. 8 show that an error of >5% in mass concentration will be caused in just one time step if the time step is larger than 20 min.

Appendix A

A1 Henzing et al. (2006) formula fitted from comprehensive numerical simulation

Henzing et al. (2006) developed a simple parameterization that represents below-cloud scavenging coefficients as a function of aerosol particle size and rainfall intensity. The parameterization is a simple three-parameter fit through below-cloud scavenging coefficients calculated at high particle size resolution. The calculations were based on the concept of collection efficiency between polydisperse aerosol particles and raindrop distributions. Specifically, Slinn's semi-empirical formula was used for the raindrop-particle collection efficiency. The gamma function fit of de Wolf (2001) and the empirical formula of Atlas et al. (1973) were applied to represent the raindrop size distribution and the terminal fall velocity, respectively. The parameterization has been applied in a global chemical transport model. The final fitting function has the form

$$\Lambda\left(d_{\mathrm{p}}\right) = A_0\left(e^{A_1 \mathrm{R}^{A_2}} - 1\right), \qquad \text{(A1-1)}$$

where the parameters A_0, A_1 and A_2 are provided in a table that is available at http://www.knmi.nl/~velthove/wet_deposition/coefficients.txt.

A2 Baklanov and Sorensen (2001) empirical parameterization

Baklanov and Sorensen (2001) suggested a simple parameterization representing scavenging rate as a function of rainfall intensity and aerosol particle size based on earlier experiment data from several different groups:

$$\Lambda(r_p) = \begin{cases} a_0 R^{0.79} & r_p \prec 1.4\,\mu\text{m} \\ (b_0 + b_1 r_p + b_2 r_p^2 + b_3 r_p^3) f(R) & 1.4\,\mu\text{m} \prec r_p \prec 10\,\mu\text{m}, \\ f(R) & r_p \succ 10\,\mu\text{m} \end{cases} \tag{A2-1}$$

and

$$f(R) = a_1 R + a_2 R^2, \tag{A2-2}$$

where r_p is particle radius (in µm), a_0=8.4×10^{-5}, a_1=2.7×10^{-4}, a_2=−3.618×10^{-6}, b_0=−0.1483, b_1=0.3220133, b_2=−3.0062×10^{-2}, b_3=9.34458×10^{-4}, and R is rainfall intensity (in mm h^{-1}). The parameterization has been incorporated into the Danish Emergency Response Model of the Atmosphere (DERMA).

A3 Laakso et al. (2003) empirical parameterization

Laakso et al. (2003) suggested a parameterization for $\Lambda(d_p)$ based on their analysis of six years of field measurements over forests in Southern Finland:

$$\begin{aligned} \log_{10}\Lambda(d_p) = {} & a_1 + a_2[\log_{10} d_p]^{-4} + a_3[\log_{10} d_p]^{-3} + a_4[\log_{10} d_p]^{-2} \\ & + a_5[\log_{10} d_p]^{-1} + a_6 R^{1/2}, \end{aligned} \tag{A3-1}$$

where d_p is particle diameter (in µm), a_1=274.35758, a_2=332 839.59273, a_3=226 656.57259, a_4=58 005.91340, a_5=6588.38582, a_6=0.244984, R is rainfall intensity (in mm h^{-1}). The formula is valid only for limited ranges of particle diameters 0.01–0.5 µm and for rain intensities 0–20 mm h^{-1}.

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Table 1. Some parameterizations for raindrop terminal velocity (in $\mathrm{cm\,s^{-1}}$).

Source	Approximate formula
Kessler (1969)	$V(D_p)=1300D_p^{0.5}$
Atlas and Ulbrich (1977)	$V(D_p)=1767D_p^{0.67}$
Willis (1984)	$V(D_p)=4854D_p\exp(-1.95D_p)$
Best (1950)	$V(D_p)=958\left[1-\exp\left(-\left(\frac{D_p}{0.171}\right)^{1.147}\right)\right]$
Atlas et al. (1973)	$V(D_p)=965-1030\exp(-6D_p)$
Brandes et al. (2002)	$V(D_p)=-10.21+4932D_p-9551D_p^2+7934D_p^3-2362D_p^4$

Here D_p in centimeters.

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Table 2. The total number concentration N_{total} yielded by three typical raindrop size distributions at different rainfall intensities.

R	MP (Exponential)			DE (Gamma)			FL (Lognormal)		
($mm\,h^{-1}$)	N_{total} (m^{-3})	f_1^a (%)	f_2^b (%)	N_{total} (m^{-3})	f_1 (%)	f_2 (%)	N_{total} (m^{-3})	f_1 (%)	f_2(%)
0.01	732.0	65.8	34.2	29.5	4.4	95.6	62.3	0.5	99.5
0.1	1191.9	48.4	51.6	65.6	1.2	98.8	103.3	0.0	100.0
1	1937.8	33.5	66.5	145.8	0.3	99.7	171.5	0.0	100.0
5	2720.0	25.3	74.7	254.8	0.1	99.9	244.3	0.0	100.0
10	3147.4	22.2	77.8	324.1	0.1	99.9	284.6	0.0	100.0
20	3641.8	19.6	80.4	412.2	0.0	100.0	331.5	0.0	100.0
50	4416.1	16.4	83.6	566.5	0.0	100.0	405.5	0.0	100.0
70	4740.0	15.4	84.6	636.6	0.0	100.0	436.7	0.0	100.0
100	5109.3	14.4	85.6	720.5	0.0	99.9	472.3	0.0	99.9

[a] f_1 is the percentage of the number concentration with the raindrop diameter less 0.1 mm;
[b] f_2 is the percentage of the number concentration with the raindrop diameter between 0.1 and 6 mm.

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Table 3. Some commonly used parameterizations for below-cloud particle scavenging by rain.

Source	Raindrop size distribution	Terminal velocity	Collection efficiency	Types
Feng (2007)	Marshall-Palmer (Exponential)	Theoretical calculation	Slinn (1983)	Type I
Andronache (2003)	Marshall-Palmer (Exponential)	Kessler (1969)	Slinn (1983)	Type I
Calderon et al. (2008)	Massambani and Morales (Gamma)	Theoretical calculation	Slinn (1983)	Type I
Mircea et al. (2000)	Feingold and Levin (1986) (Lognormal)	Theoretical calculation	Slinn (1983)	Type I
Andronache et al. (2006)	Marshall-Palmer (Exponential)	Atlas and Ulbrich (1977)	Slinn (1983) +phoresis +electric forces	Type I
Loosmore and Cederwall (2004)	Monodisperse $D_p = 0.97\mathrm{R}^{0.158}$ mm	Willis (1984)	Slinn (1983)	Type I
AURAMS (Gong et al., 2006)	Monodisperse $D_p = 0.7\mathrm{R}^{0.25}$ mm	Theoretical calculation	Slinn (1983)	Type I
Henzing et al. (2006)	Fitted functions from explicit calculation			Type II
Laakso et al. (2003)	Empirical formula from observations			Type III
Baklanov and Sorensen (2001)	Empirical formula from observations			Type III

Table 4. Nomenclature.

C_c	Cunningham correction factor	q_p	mean charge of a particle (C)
c_p	heat capacity of air ($m^2\,s^{-2}\,K^{-1}$)	R	rainfall intensity ($mm\,h^{-1}$)
d_p	particle diameter (m)	Re	raindrop Reynolds number
D_p	raindrop diameter (m)	RH	relative humidity (%)
$\bar{D}_p$	mean raindrop diameter of lognormal spectra (m)	Sc	particle Schmidt number
D_r	representative diameter of monodisperse raindrop spectra (m)	Sc_w	Schmidt number for water in air
D_{diff}	particle Brownian diffusivity coefficient ($m^2\,s^{-1}$)	St	particle Stokes number
$D_{diffwater}$	water vapour diffusivity in air ($m^2\,s^{-1}$)	St^*	critical Stokes number of particle
$E(d_p,D_p)$	overall collection efficiency	T_a	air temperature (K)
$E_{th}(d_p,D_p)$	collection efficiency due to thermophoresis	T_s	temperature of the raindrop surface (K)
$E_{dph}(d_p,D_p)$	collection efficiency due to diffusiophoresis	$v(d_p)$	particle terminal velocity ($m\,s^{-1}$)
$E_{es}(d_p,D_p)$	collection efficiency due to charge effect	$V(D_p)$	raindrop terminal velocity ($m\,s^{-1}$)
g	Acceleration of gravity ($m\,s^{-2}$)	β_g	slope parameter for gamma raindrop size distribution
k_a	thermal conductivity of air ($J\,m^{-1}\,s^{-1}\,K^{-1}$)	β_e	slope parameter for exponential raindrop size distribution
k_p	thermal conductivity of particle ($J\,m^{-1}\,s^{-1}\,K^{-1}$)	γ	shape parameter for gamma raindrop size distribution
k_b	Boltzmann constant ($J\,K^{-1}$)	λ	mean free path of air molecules (m)
M_a	air molecular weight	$\Lambda(d_p)$	a size-resolved scavenging coefficient of particle (s^{-1})
M_w	water vapour molecular weight	μ_a	air viscosity ($kg\,m^{-1}\,s^{-1}$)
$N(D_p)$	raindrop number size distribution (m^{-4})	μ_w	water viscosity ($kg\,m^{-1}\,s^{-1}$)
N_{0e}	parameter for exponential raindrop size distribution (m^{-4})	ρ_a	air density ($kg\,m^{-3}$)
N_{0g}	parameter for gamma raindrop size distribution ($m^{-\gamma-1}\,m^{-3}$)	ρ_p	particle density ($kg\,m^{-3}$)
N_{total}	total number concentration of raindrops (m^{-3})	ρ_w	water density ($kg\,m^{-3}$)
P	atmospheric pressure (Pa)	σ_D	standard deviation of lognormal raindrop size distribution
Pr	Prandtl number for air	τ	characteristic relaxation time of particle (s)
P_a^0	vapour pressure of water at temperature T_a (Pa)		
P_s^0	vapour pressure of water at temperature T_s (Pa)		
Q_r	mean charge of a raindrop (C)		

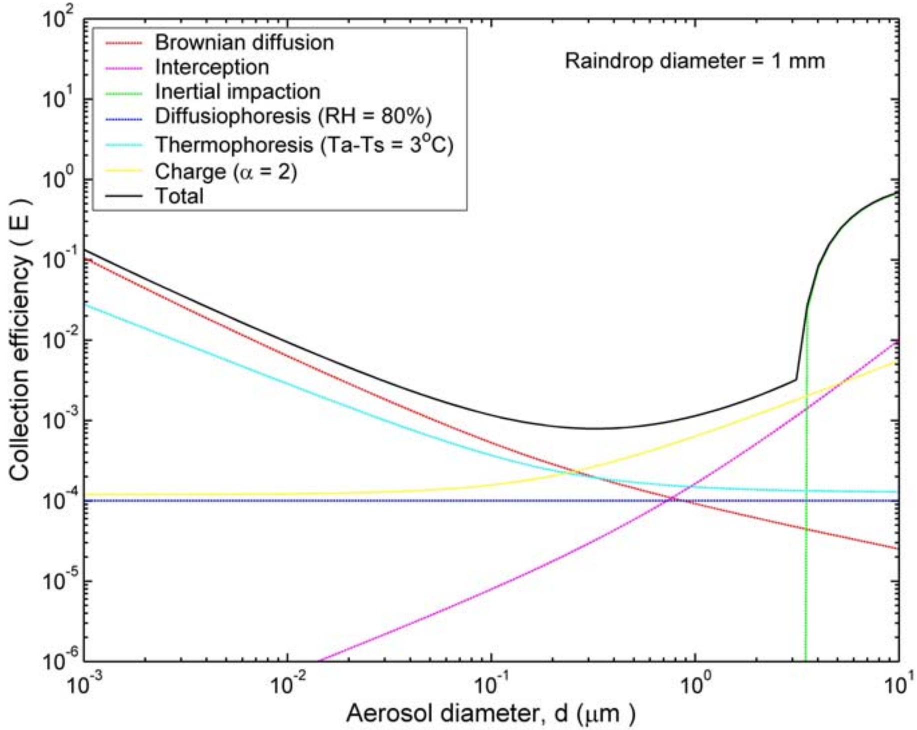

Fig. 1. Contributions of various collection processes to the collection efficiency $E(d_p, D_p)$ as a function of the aerosol particle size for a raindrop with diameter of 1 mm.

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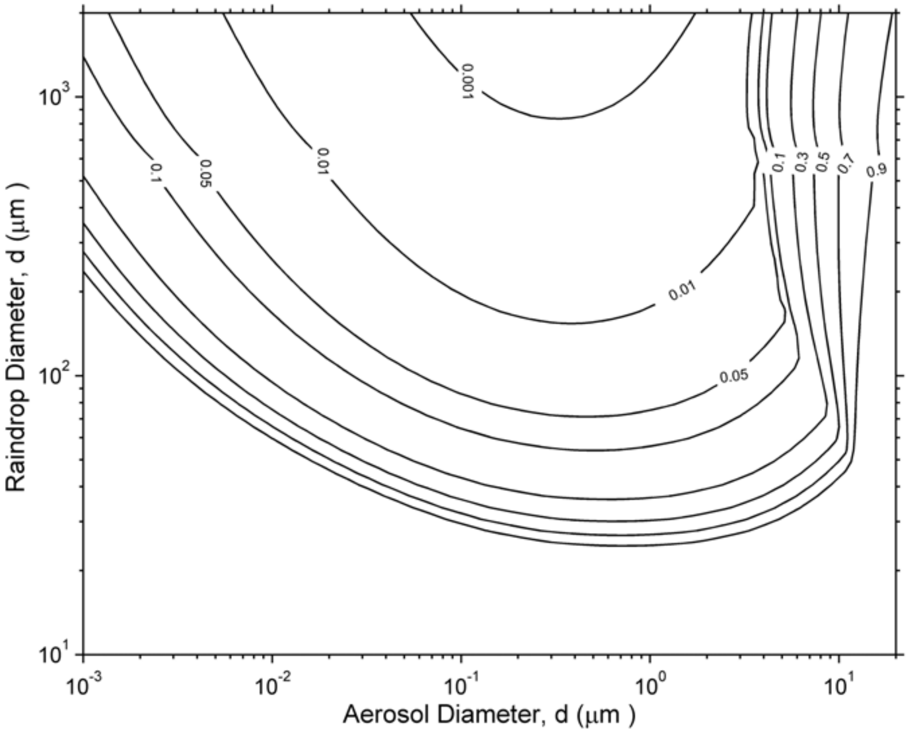

Fig. 2. Total collection efficiency $E(d_p,D_p)$ (contoured) taking into account the contributions of various processes shown in Fig. 1 as a function of both particle diameter and raindrop diameter.

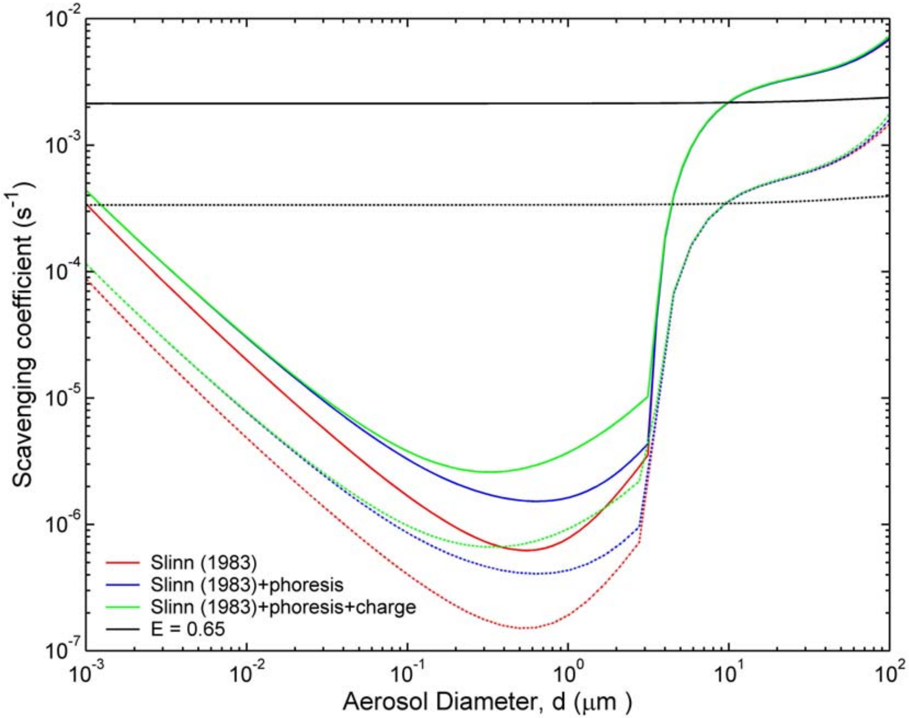

Fig. 3. Scavenging coefficients determined with different collection efficiency formulations as a function of particle size for rainfall intensities of 1 mm h^{-1} (dotted line) and 10 mm h^{-1} (solid line).

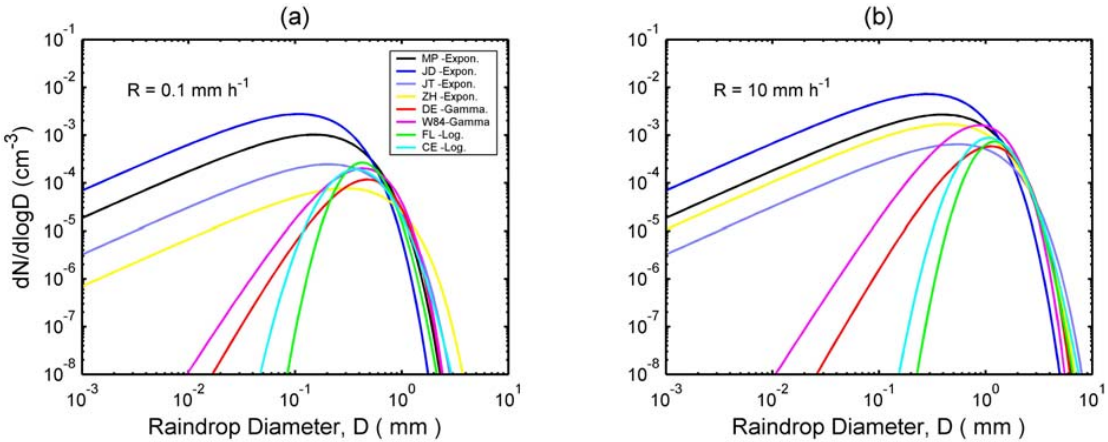

Fig. 4. Several typical raindrop number size distributions for rainfall intensities of **(a)** R=0.1 mm h^{-1}, and **(b)** R=10 mm h^{-1}. Here, MP is for Marshall and Palmer (1948), JD for Joss Drizzle (Joss et al., 1968), JT for Joss thunderstorms (Joss et al., 1968), ZH for Zhang et al. (2008), DE for de Wolf (2001), W84 for Willis (1984), FL for Feingold and Levin (1986) and CE for Cerro et al. (1997). MP and DE represent stratiform rain, JD represents drizzle rain, ZH, FL and CE represent convective rain, and JH and W84 represent thunderstorms.

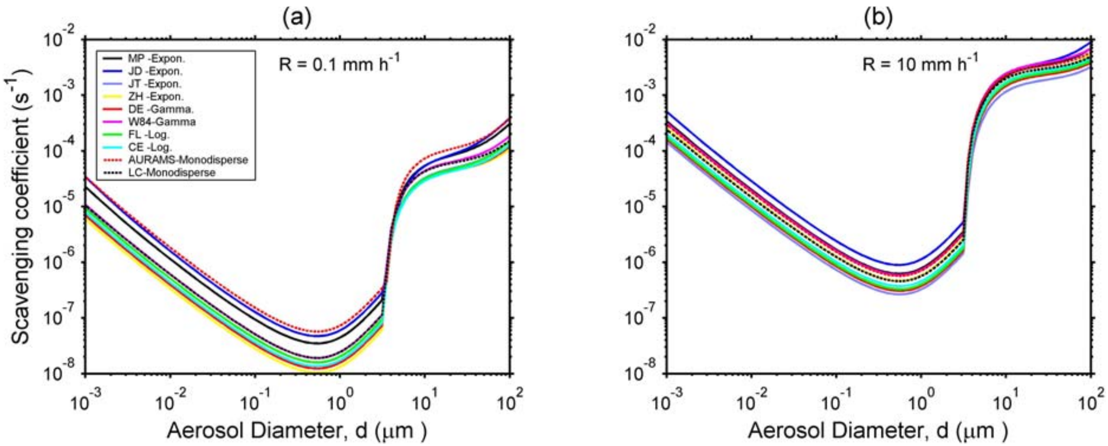

Fig. 5. Scavenging coefficients as a function of particle size derived from several raindrop size distributions for rainfall intensities of **(a)** R=0.1 mm h^{-1} and **(b)** R=10 mm h^{-1}.

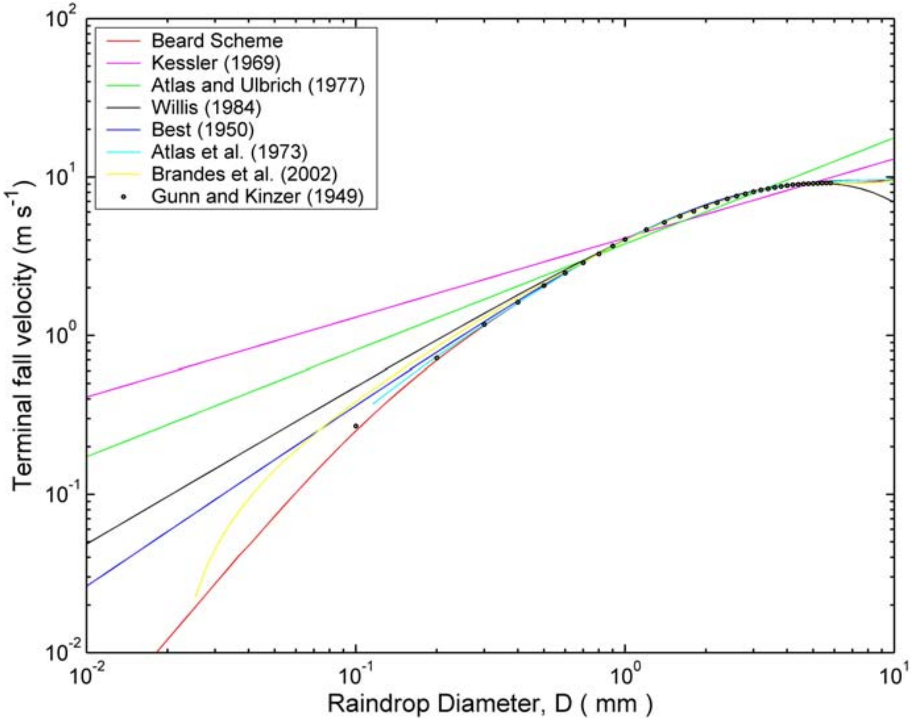

Fig. 6. Terminal fall velocity versus drop diameter.

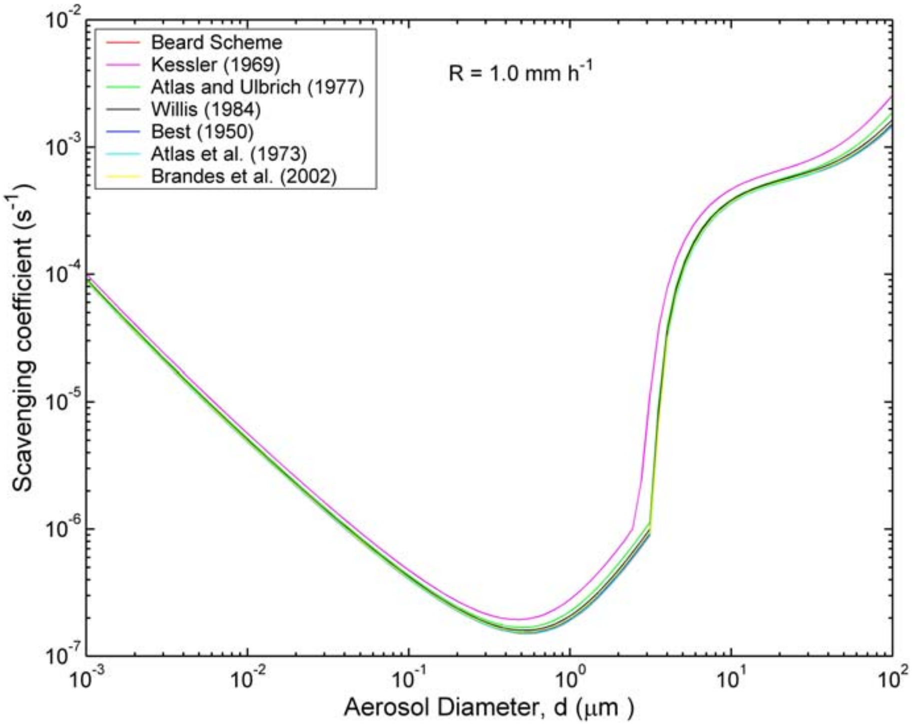

Fig. 7. Scavenging coefficients as a function of particle size for different terminal velocity parameterizations assuming a rainfall intensity of 1.0 mm h^{-1}.

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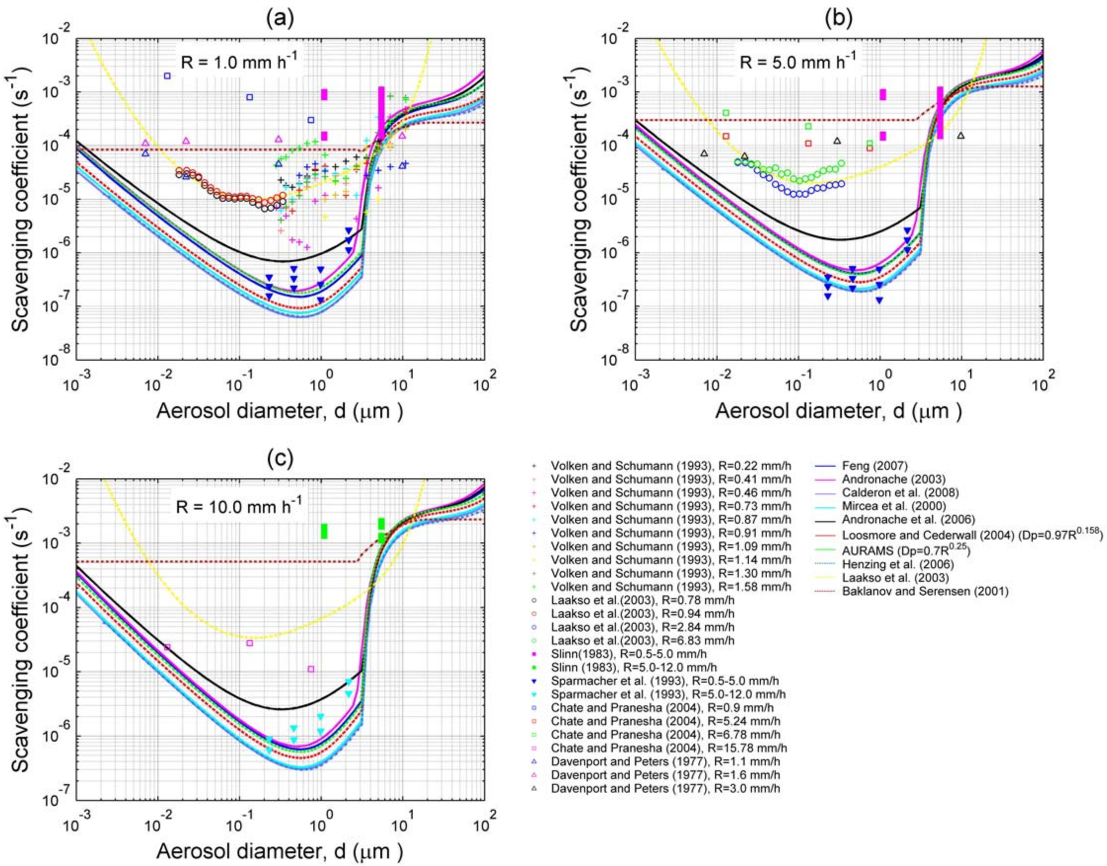

Fig. 8. Scavenging coefficients as a function of aerosol diameter for various parameterizations listed in Table 3 and the measurements for different rainfall intensities of **(a)** R=1 mm h^{-1}, **(b)** R=5 mm h^{-1}, and **(c)** R=10 mm h^{-1}.

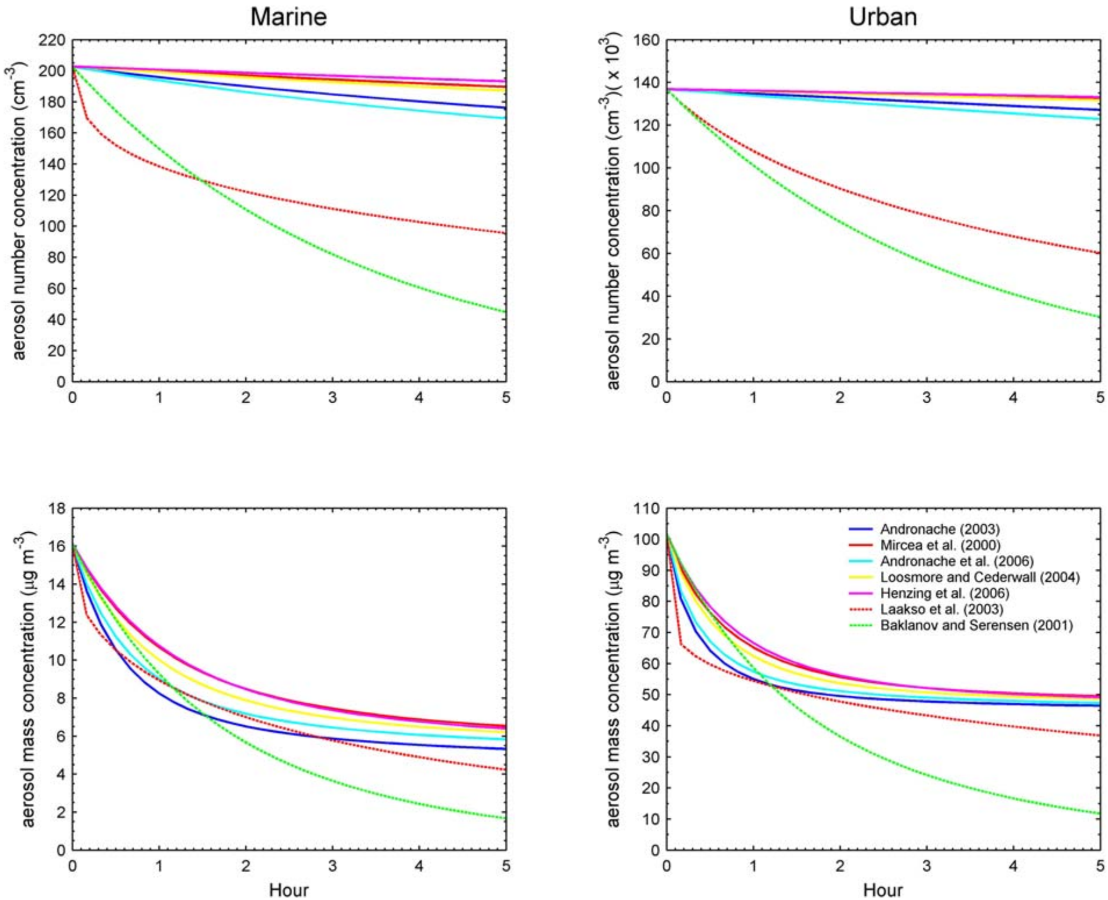

Fig. 9. Impact of different Λ parameterizations on the total number (upper row) and mass (lower row) concentrations for two aerosol types of marine and urban as a function of time with a rainfall intensity of 1 mm h^{-1}.

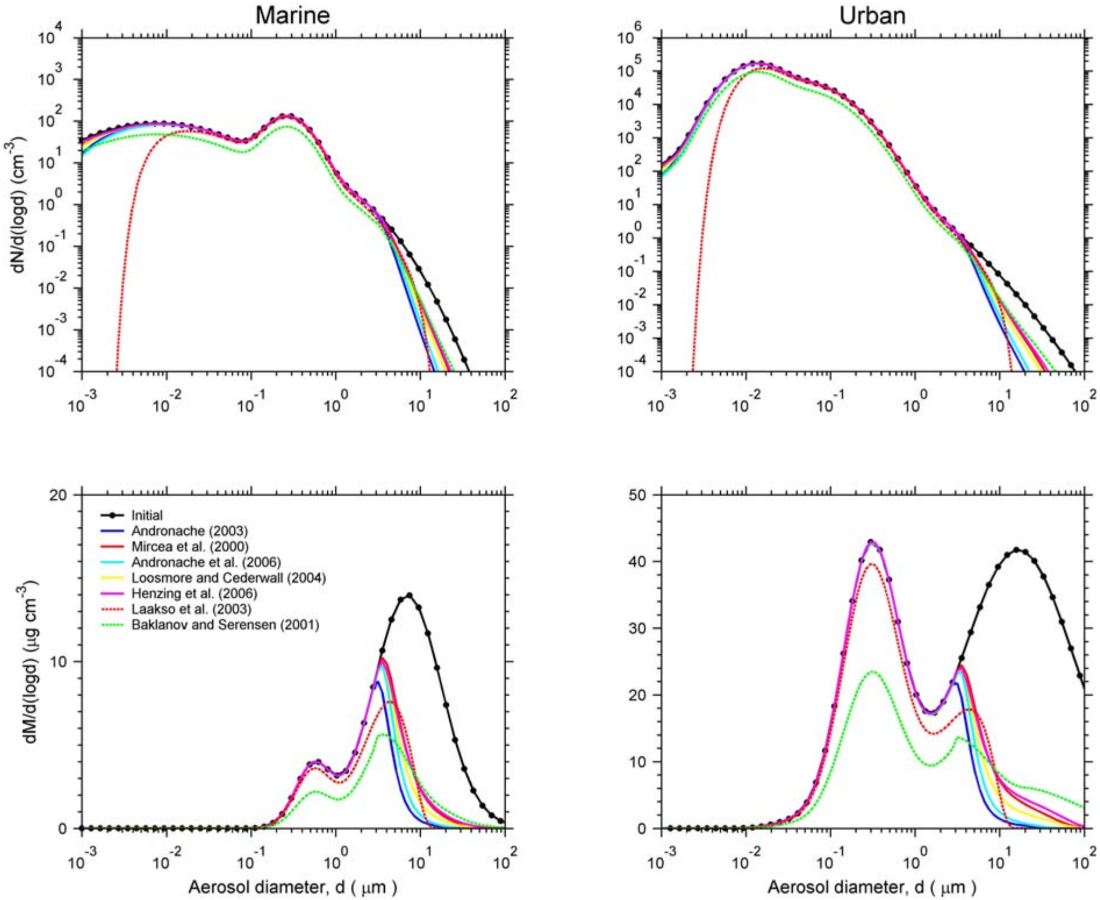

Fig. 10. Impact of different Λ parameterizations on the size-resolved number (upper row) and mass (lower row) distributions for two aerosol types (marine and urban) after two hours of rain with a rainfall intensity of 1 mm h^{-1}. The initial size distributions of each aerosol type is plotted as well. Note that different y-axis scales have been used.