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On the discrepancies between theoretical and measured below-cloud particle scavenging coefficients for rain – a numerical study

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Abstract

Existing theoretical formulations for size-resolved scavenging coefficient $\Lambda(r)$ for atmospheric aerosol particles scavenged by rain predict values lower by one to two orders of magnitude than those estimated from field measurements of particle-concentration changes for particles smaller than 3 µm in diameter. Vertical turbulence does not influence the theoretical formulation of $\Lambda(r)$, but contributed to the field-generated $\Lambda(r)$ due to its influence on the overall concentration changes of aerosol particles in the layers undergoing impaction scavenging. A detailed one-dimensional cloud microphysics model is used to simulate rain production and below-cloud particle scavenging, and to quantify the contribution of turbulent diffusion to the overall $\Lambda(r)$ generated from concentration changes. The relative contribution of vertical diffusion to below-cloud scavenging is found to be largest for submicron particles under weak precipitation conditions. The discrepancies between theoretical and field $\Lambda(r)$ values can largely be explained by the contribution of vertical diffusion for all particles larger than 0.01 µm in diameter for which field data were available. The results presented here suggest that the current theoretical framework for $\Lambda(r)$ can provide a reasonable approximation of below-cloud aerosol particle scavenging by rain in size-resolved aerosol transport models if vertical diffusion is also considered.

1 Introduction

Precipitation scavenging is a major removal process for below-cloud aerosol particles. Due to the complex interactions between particles and rain droplets, the scavenging process needs to be parameterized in the mass continuity equations used in atmospheric chemical transport models (CTMs). The parameter known as scavenging coefficient (Λ), which is defined as the time rate of change of the ambient concentration of aerosol particles due to precipitation scavenging, is often used to quantify this process (e.g., Seinfeld and Pandis, 2006). Different formulations of Λ apply to bulk particle

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number, bulk particle mass, and size-resolved particle number or mass concentrations (e.g., Zhang et al., 2004). Theoretical size-resolved formulas for Λ are typically parameterized as a function of collection efficiency, in which some or all of the known particle-droplet collection mechanisms, including Brownian diffusion, interception, impaction, thermo- and diffusiophoresis, and electrostatic forces, are considered (Slinn, 1983; Andronache et al., 2006; Loosmore and Cederwall, 2004; Chate, 2005; Park et al., 2005; Henzing et al., 2006; Tost et al., 2006; Feng, 2007; Croft et al., 2009).

It has long been known, however, that size-resolved values of $\Lambda(r)$ calculated from all existing theoretical parameterizations are one to two orders of magnitude smaller than the majority of field measurements for all particle sizes except for those larger than 3 µm in diameter, for which theoretical and field $\Lambda(r)$ values agree well. Turbulence was suspected to play key roles in these large discrepancies as supported by limited measurements collected under controlled conditions (Sparmacher et al., 1993). In a detailed review paper, Wang et al. (2010) summarized that the enhancement of particle scavenging by turbulence can come from two aspects: (1) the increased droplet/aerosol collision efficiency by turbulence effect (Grover and Pruppacher, 1985; Khain and Pinsky, 1997); and (2) mixing of particles from subcloud layer into cloud layer (Andronache et al., 2006). If the first aspect is dominant, then the current theoretical $\Lambda(r)$ formulations need to be modified to include this effect; and if the second aspect is dominant, then the excising $\Lambda(r)$ formulations can be used in chemical transport models since vertical diffusion is already include in the mass continuity equation.

In the present study we have employed a detailed aerosol-cloud microphysics numerical model to investigate the contribution of vertical turbulent diffusion to overall aerosol particle scavenging (the second aspect mentioned above). If this contribution could be quantified, we would have a better idea of whether or not there is a need to improve existing theoretical $\Lambda(r)$ formulations. CARMA (Community Aerosol and Radiation Model for Atmospheres), the detailed aerosol-cloud microphysics model that has been used in this study, was originally developed at the NASA Ames Research Center (Toon et al., 1988; Ackerman et al., 1995) and was later modified by Zhang et

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al. (2004) to produce light precipitation from low-level warm stratiform clouds in order to study precipitation scavenging. A similar approach was used here to produce rain with intensities from 0.1 to 5 mm h^{-1} for studying below-cloud aerosol particle scavenging (i.e., "wash-out") under conditions with and without vertical turbulent diffusion. The simulation results provide some guidance on the source of the differences between theoretical formulations and field measurements of $\Lambda(r)$ and on future research needs.

2 Methodology

The theory and algorithms used in CARMA have been described in detail by Toon et al. (1988) and Ackerman et al. (1995), while the model set up for producing rain in order to study aerosol scavenging was described in detail in Zhang et al. (2004). Only a brief description of model characteristics and the design of the sensitivity tests is given below.

In the simulations conducted in the present study, the size distributions of liquid (cloud and rain) droplets and aerosol particles were both represented by 50-size-bin sectional distributions. The diameter range spanned 0.001 to 100 µm for particles and 1 µm to 10 mm for droplets with constant volume ratios of 2.12 and 1.78, respectively, between successive bins. The time rates of change of the number concentrations of both particles and droplets were solved using the following continuity equation in a one-dimensional model framework:

$$\frac{\partial C}{\partial t} = \frac{C}{\rho}\frac{\partial \rho w}{\partial z} - \frac{\partial}{\partial z}[(w - V_f)C] + \frac{\partial}{\partial z}[K_z \rho \frac{\partial (C/\rho)}{\partial z}] + P - L \quad , \tag{1}$$

where $C(r,z,t)$ is the number concentration for a size bin, which depends on particle or droplet radius r, time t, and height z, and $C(r)dr$ represents the mean number concentration (m^{-3}) of particles or droplets having radii between r and $r+dr$. $\rho(z,t)$ is the density of dry air (kg m^{-3}), $w(z,t)$ is the vertical air velocity (m s^{-1}), $V_f(r,z)$ is the particle or droplet fall velocity (m s^{-1}), and $K_z(z)$ is the vertical diffusion coefficient (m^2 s^{-1}).

The first term on the right side of Eq. (1) represents the horizontal divergence that compensates for any changes in air density due to vertical convergence. The second term represents the divergence of the vertical flux due to vertical advection and sedimentation. The third term represents the divergence of the vertical flux due to turbulent diffusion. The last two terms represent production (P) and loss (L), respectively, of particles and droplets due to microphysical processes, which include activation of particles (i.e., cloud nuclei) into cloud droplets, condensation/evaporation of droplets, and collision-coalescence (collection) of particle pairs, droplet pairs, and particle-droplet pairs. Apparently, the theoretical formulation of the scavenging coefficient is hidden in the P and L terms mentioned above.

The vertical domain of the model goes from the surface to 6 km with 141 unevenly spaced vertical levels. The vertical grid spacing was chosen to be 10 m for the lowest layer, gradually increasing to 40 m at 400 m, and then remaining at 40 m up to 6 km. A prescribed steady vertical advection function with peak value at cloud mid-height decreasing to zero at both initial cloud base (1000 m) and cloud top (3500 m) was used to form the cloud layer and the subsequent precipitation. Two different peak wind vertical wind speeds were considered: 0.15 and 0.45 m s^{-1}. The vertical diffusion coefficient K_z was parameterized according to Brost and Wyngaard (1978):

$$K_z = \kappa u_* z \left(1 - \frac{z}{h}\right)^{1.5} \left(1 - B\frac{d\theta}{dz}\right) \quad , \tag{2}$$

where κ is the von Karman constant ($= 0.4$), u_* is the friction velocity, h is the boundary-layer height, B is an empirical constant, taken to be 40 m K^{-1}, and θ is potential temperature.

The initial three-mode, rural-type size distribution of the aerosol particles, their chemical composition and vertical distribution, and the initial conditions of vertical profiles of temperature and relative humidity followed Zhang et al. (2004). CARMA was integrated forward in time with a time step of 10 s. Once the precipitation intensity at near-surface levels reached one of the three specific values designed for our sensitivity tests (i.e., 0.1, 1, and 5 mm h^{-1}), the below-cloud particle number concentration profile was

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restored to its initial profile for the purpose of tracking concentration changes in order to calculate the size-resolved scavenging coefficient $\Lambda(r)$ using the formula (e.g., Seinfeld and Pandis, 2006)

$$\Lambda(r) = -\frac{1}{\Delta t}\frac{C(r,t_2) - C(r,t_1)}{C(r,t_1)} \quad , \tag{3}$$

where $C(r,t_1)$ and $C(r,t_2)$ are particle number concentrations at the beginning and end, respectively, of a time period Δt (chosen as 6 min for strong precipitation and 20 min for weak precipitation).

Note that for modelling studies (the theoretical formulations) the scavenging coefficient is generally determined only from the process of the scavenging and not by the concentration changes predicted by the full Eq. (1). Since the vertical diffusion is included in the derivation of the change of concentration in the model, $\Lambda(r)$ derived from Eq. (3) includes contributions from both 'traditionally defined' scavenging and the vertical turbulent diffusion.

3 Results

By modifying the magnitude of the vertical advection within the pre-chosen cloud layer, three different precipitation intensities were obtained at the surface, representing weak (0.1 mm h^{-1}), moderate (1 mm h^{-1}), and strong (5 mm h^{-1}) precipitation, respectively. For each precipitation intensity, four sensitivity tests were conducted. Test 1 represents the situation with no vertical diffusion ($K_z = 0$); Tests 2 to 4 represent extremely weak, relatively weak, and strong vertical diffusion conditions, respectively (see Fig. 1 for the K_z distributions used for Tests 2–4). Size-resolved scavenging coefficients were calculated based on changes in modeled number concentrations from the second lowest model layer (16 m mid-layer height as marked in Fig. 1). The modeled $\Lambda(r)$ distributions for the three different precipitation intensities are shown in Fig. 2 (see four colored solid

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lines in each subset figure). Also shown in Fig. 2 are $\Lambda(r)$ distributions from two theoretical formulations, Andronache et al. (2006) (black solid line) and Mircea et al. (2000) (black dashed line), which represent the range of existing theoretical formulations (see Wang et al., 2010).

With the vertical diffusion coefficient set to 0 in Test 1, $\Lambda(r)$ calculated from modeled concentrations was mainly a result of particle-droplet collection (collision and coalescence) processes. Although gravitational setting of aerosol particles also contributes to the modeled $\Lambda(r)$, its contribution was only noticeable for particles larger than 5 µm in diameter under weak precipitation and for particles larger than 10 µm under moderate to strong precipitation. Thus, this process was not a concern in the discussion presented below. As expected, $\Lambda(r)$ calculated for Test 1 fell in between the two black lines representing the range of existing theoretical $\Lambda(r)$ formulations for all precipitation intensities (Fig. 2). The differences between Test 1 and the theoretical $\Lambda(r)$ formulations were simply caused by differences in their treatments of collection efficiency, droplet spectrum, and to a less extent, terminal fall velocity, and these differences in $\Lambda(r)$ were generally within one order of magnitude (Wang et al., 2010). As shown in Fig. 2, these theoretical $\Lambda(r)$ distributions (including Test 1 here) were also of the same order of magnitude as those calculated for one controlled experiment of Sparmacher et al. (1993), for which turbulence effects were minimized. However, they were almost two orders of magnitudes smaller than $\Lambda(r)$ values from the majority of other field experiments.

When the effect of vertical diffusion is considered in the model, $\Lambda(r)$ calculated using Eq. (3) increased as can be seen from Tests 2–4. For very small K_z (0.013 $m^2 s^{-1}$ at 16 m height), $\Lambda(r)$ was only increased significantly for submicron particles and under weak precipitation conditions (Fig. 2a). When K_z was increased to 0.15 $m^2 s^{-1}$ at 16 m (Test 3), $\Lambda(r)$ increased markedly relative to Test 2, i.e., by a factor of 5 to 10 for submicron particles under moderate to strong precipitation conditions (Fig. 2b, c) and by more than a factor of 10 under weak precipitation conditions (Fig. 2a). However, the contribution of turbulent diffusion was negligible for very small (< 0.01 µm diameter)

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and very large (> 3 µm) particles under moderate to strong precipitation conditions due to their already very high $\Lambda(r)$ values (associated with Brownian diffusion and inertial impaction, respectively). These $\Lambda(r)$ values were still smaller, however, than those from the majority of field measurements. Finally, when K_z was increased to a value representative of fully unstable turbulent conditions ($1.5\,m^2\,s^{-1}$ at 16 m), predicted $\Lambda(r)$ values increased to a level comparable to the field measurements. It should be pointed out that under rain conditions the turbulence intensity is generally strong, even in the nighttime. Thus, the K_z values in Test 4 are more realistic than in Tests 1–3. These results suggest that most of the discrepancy between theoretical and field $\Lambda(r)$ distributions can be explained by the contribution of vertical turbulent diffusion, which is not considered in current theoretical $\Lambda(r)$ formulations but which likely contributed in the field experiments.

Note that vertical diffusion contributes little for particles larger than 3 µm in diameter due to the very high $\Lambda(r)$ for this particle size range due to inertial impaction. However, existing theoretical $\Lambda(r)$ formulations that only consider particle-droplet collection processes can predict $\Lambda(r)$ in the same order of magnitude as available measurements for these larger particles. Note also that vertical diffusion has the potential to increase $\Lambda(r)$, although by a small amount, for particles smaller than 0.01 µm in diameter. Unfortunately, field data for this particle size range are very rare. Finally, the model results from the sensitivity tests presented above suggest that the current theoretical $\Lambda(r)$ formulations can provide a reasonable approximation for below-cloud particle scavenging by rain in size-resolved aerosol transport models provided that vertical diffusion is also accounted for, although further validation of this assumption is needed for aerosols smaller than 0.01 µm in diameter when more field data become available.

4 Summary and conclusions

Numerical sensitivity tests were designed for a detailed aerosol-cloud microphysics model to quantify the contribution of vertical turbulent diffusion to below-cloud particle

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scavenging by rain. When turbulent diffusion was excluded, the model-predicted size-resolved scavenging coefficient $\Lambda(r)$ agreed well with the results of one controlled field experiment in which turbulent effects were believed to be minimized. For weakly turbulent conditions, vertical diffusion was predicted to enhance $\Lambda(r)$ significantly for submicron particles but had very limited impact on very small particles ($< 0.01\,\mu$m in diameter). Under strongly turbulent conditions, vertical diffusion increased $\Lambda(r)$ by two orders of magnitude for submicron particles and by up to one order of magnitude for very small particles. For particles larger than $3\,\mu$m in diameter, on the other hand, turbulent diffusion had little effect on precipitation scavenging. The contribution of vertical diffusion to $\Lambda(r)$ was predicted to vary inversely with precipitation intensity. For example, the influence of vertical diffusion was noticeable for particles smaller than $0.005\,\mu$m in diameter under weak precipitation but became negligible when precipitation intensity increased to $5\,\mathrm{mm\,h^{-1}}$.

The results presented here suggest that vertical diffusion alone can explain almost all of the discrepancies between theoretical and field-derived $\Lambda(r)$ for particles in the 0.01 to $3\,\mu$m diameter range for which size-specific field data were available. This suggests in turn that existing theoretical formulations for $\Lambda(r)$ can be applied without modification in those aerosol transport models in which a vertical diffusion term is already included in the mass continuity equation, although the recommendation in Wang et al. (2010) that the theoretical $\Lambda(r)$ parameterization that predicts the highest scavenging coefficient values should be used still holds. It is recommended that future field studies of precipitation scavenging should be designed if possible to include aerosol particles smaller than $0.01\,\mu$m in diameter as well as other particle size ranges. It is also recommended that field measurements should be made simultaneously under both natural and controlled environmental conditions so the effect of vertical turbulent diffusion can be separated from that of the particle-droplet collection process. For example, the scavenging coefficients can be extracted through the measurements of aerosol concentrations, in natural field environments and in a controlled box where surface turbulence is limited, during rain events.

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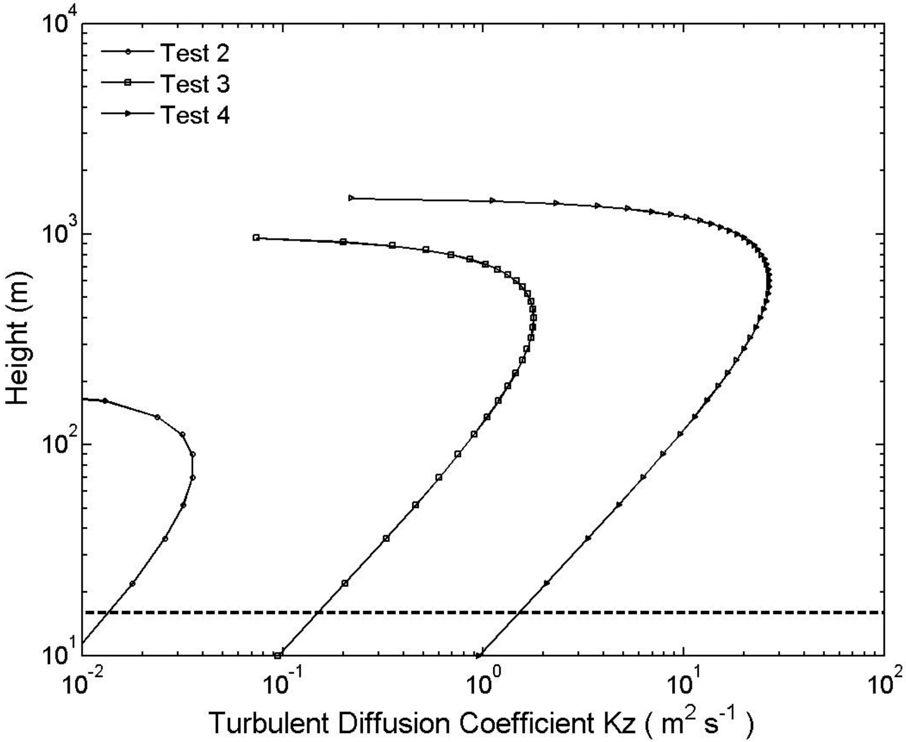

Fig. 1. Vertical profiles of the vertical diffusion coefficient K_z used for the sensitivity tests in which the boundary-layer height was chosen to be 200, 1000, and 1500 m, respectively.

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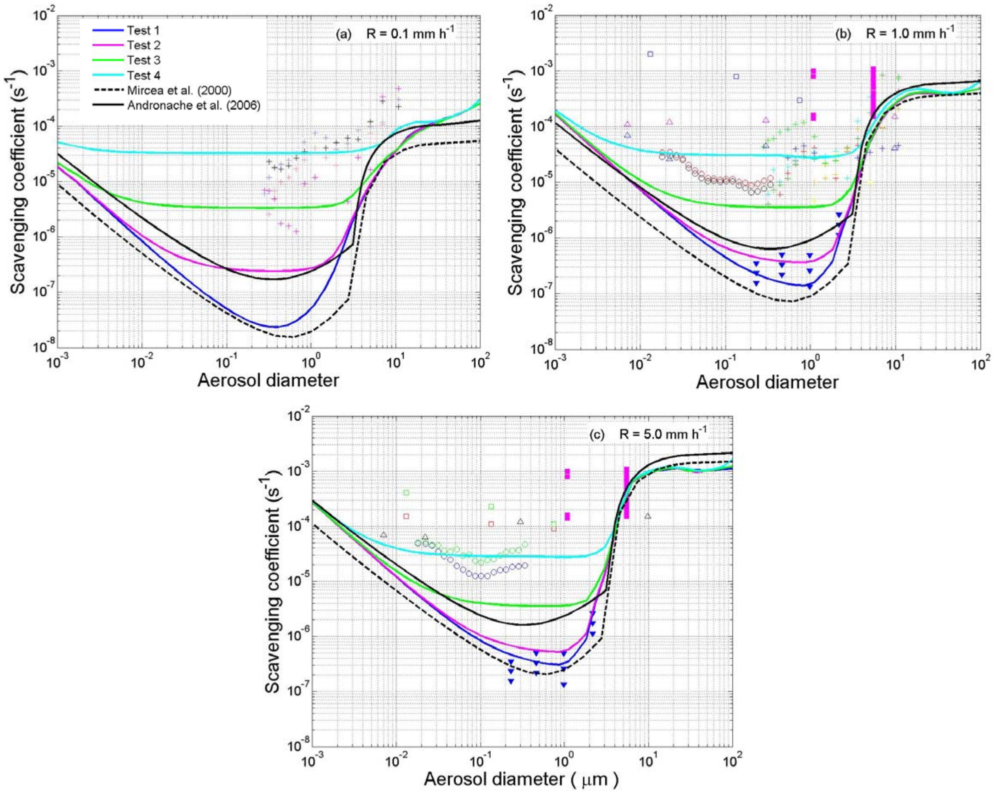

Fig. 2. Size-resolved scavenging coefficient $\Lambda(r)$ for three precipitation intensities calculated from model-predicted concentration changes at 16 m height for tests with and without vertical turbulent diffusion (four solid colored lines). Also shown are scavenging coefficients from two theoretical parameterizations (two black lines) representing the range of existing theoretical formulations. Available measurements of $\Lambda(r)$ were copied from Wang et al. (2010) and are shown by various symbols.

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