The rate of loss of cloud condensation nuclei by coalescence in warm clouds

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Abstract

An approximate analytical expression for the rate of loss of cloud droplets by coalescence in warm clouds is derived from the stochastic collection equation (SCE). The expression depends only upon precipitation rate and cloud droplet concentration and compares well with estimated loss rates derived using observed cloud drop size distributions and the complete collection kernel. Loss rates are found to be surprizingly high even for the modest precipitation rates found in drizzling boundary layer clouds, and can be used to infer the loss rate of cloud condensation nuclei (CCN) through coalescence. The expression can be used to better represent the interdependence of aerosol and cloud properties in the boundary layer.

1. Introduction

The interdependence of clouds and aerosols is currently the subject of considerable debate (Lohmann and Feichter 2005). An understanding of how aerosols impact cloud radiative properties cannot be considered to be complete without understanding how clouds themselves influence the aerosol characteristics. Models that attempt to identify the key processes controlling the aerosol size distribution in the marine boundary layer (MBL) (Raes 1995; Capaldo et al. 1999; Katoshevski et al. 1999) have demonstrated the importance of precipitation scavenging. However, the treatment of this process in these models is somewhat arbitrary and there is little or no attempt to couple the aerosol removal to the meteorology and cloud properties in the MBL. In this study, an expression is derived that can be used, in conjunction with recent expressions for the dependence of precipitation rate on cloud thickness and cloud droplet concentration, to provide important links between the properties of the clouds in the MBL and the rate of removal of aerosols.

2. Analytic expression

We consider an expression for the rate of loss of droplets through coalescence. The stochastic collection equation (SCE) gives an expression for the evolution of the drop size distribution $n(x)$ due to collision-coalescence of drops of volume x with those of volume x' (e.g. Berry 1967)

$$\frac{\partial n(x)}{\partial t} = \frac{1}{2}\int_0^x n(x-x')K(x-x',x')n(x')dx' - \int_0^\infty n(x)K(x,x')n(x')dx', \qquad (1)$$

where $K(x, x')$ is the collection kernel for coalescing drops. The total number concentration of drops N is given by

$$N = \int_0^\infty n(x)dx, \qquad (2)$$

so that the rate of increase of drops through collision-coalescence $\dot{N}$ is given by

$$\dot{N} = \frac{\partial N}{\partial t} = \int_0^\infty \frac{\partial n(x)}{\partial t} dx. \qquad (3)$$

Using (1) we obtain

$$\dot{N} = \frac{1}{2}\int_0^\infty \left\{ \int_0^x n(x-x')K(x-x',x')n(x')dx' \right\} dx - \int_0^\infty \left\{ \int_0^\infty n(x)K(x,x')n(x')dx' \right\} dx. \qquad (4)$$

Exchanging the order of integration in both terms and making the substitutions $y = x - x'$ in the inner integral of the first term and $y = x$ in the inner integral of the second, we find

$$\dot{N} = \frac{1}{2}\int_0^\infty \left\{ \int_0^\infty n(y)K(y,x')dy \right\} n(x')dx' - \int_0^\infty \left\{ \int_0^\infty n(y)K(y,x')dy \right\} n(x')dx' \qquad (5)$$

which can be simplified to

$$\dot{N} = -\frac{1}{2}\int_0^\infty \int_0^\infty n(x)K(x,x')n(x')dxdx'. \qquad (6)$$

Here (6) simply states that for each droplet that coalesces, half a droplet is lost (i.e. one drop is created from two coalescing drops), and is an exact result. To derive a useful relationship that can be expressed as a function of bulk parameters, some approximations for $K(x, x')$ must be made. We can follow the methodology of Liu and Daum (2004) who use the mean value theorem for integrals, namely that if $f(x)$ and $g(x)$ are continuous on the interval $[a, b]$ and $g(x)$ does not change sign in this interval, then there is some point $x_\zeta \in [a, b]$ such that

$$\int_a^b f(x)g(x)dx = f(x_\zeta)\int_a^b g(x)dx. \tag{7}$$

Application of (7) to (6) gives

$$\dot{N} = -\frac{1}{2}\int_0^\infty n(x)K(x, x_\zeta)dx \int_0^\infty n(x')dx' = -N\int_0^\infty n(x)K(x, x_\zeta)dx. \tag{8}$$

Following Long (1974) for small drops ($r < 50\mu$m), i.e. parameterizing $K(x, x_\zeta) = \kappa x^2$ (i.e. the kernel depends only upon the mass of the collector drop), with constant κ=1.1$\times 10^{10}$ m^{-3} s^{-1}, we obtain

$$\dot{N} = -\frac{1}{2}\kappa N\int_0^\infty x^2 n(x)dx. \tag{9}$$

Reverting to radius units, such that $x = 4\pi r^3/3$, we find

$$\dot{N} = -\kappa' N\int_0^\infty r^6 n(r)dr = -\kappa' N^2 R_6^6. \tag{10}$$

with $\kappa' = \frac{1}{2}(4\pi/3)^2\kappa$, and R_6 is the sixth moment weighted radius. For the ith moment in general, $R_i = (\int_0^\infty r^i n(r)dr/N)^{1/i}$. Equation (10) indicates that the rate of loss of cloud drop concentration in this approximation is proportional to the product of the cloud drop concentration and the sixth moment of the cloud droplet size distribution. The validity of this approximation is tested by evaluating (6) using the commonly accepted best estimates for $K(x, x')$ for cloud and drizzle drops (Hall 1980), and the observations of cloud/drizzle drop size distributions in a range of stratiform boundary layer clouds taken from Wood (2005b). Figure 1 shows $-\dot{N} = -dN/dt$ estimated using (6) against the parameterization using the Long kernel (10) indicating some correlation, but with the parameterization tending to overestimate the loss rate. Figure 2 shows the result if we only include collections of one cloud drop (here somewhat arbitrarily defined as $r < 20$ μm) by another, which indicates that the Long small drop kernel approximation is indeed excellent at estimating the collection kernel for the small drops. However, it is a poor representation of the kernel for large drops. When a significant fraction of cloud drop removal is through accretion onto drizzle drops ($r > 20\mu$m), the Long small drop parameterization is insufficient.

For large drops ($r > 50\mu$m) Long (1974) introduces a linear dependence of the kernel upon drop mass $K(x, x_\zeta) = \kappa_2 x$ with constant κ_2=6.33$\times$10^3 s^{-1}. With this kernel in (8), we obtain

$$\dot{N} = -\kappa_2' N \int_0^\infty r^3 n(r) dr = -\kappa_2' N^2 R_3^3 \tag{11}$$

where $\kappa_2' = \frac{1}{2}(4\pi/3)\kappa_2$. Figure 3 shows the comparison of (11) with the observationally-derived values, again indicating significant overprediction and considerable scatter. This is because the Long large drop parameterization severely overpredicts the influence of the small droplets. The general behavior of the Long kernel approximations in overpredicting the coalescence drop loss rate is a result of the concave nature of the combined function, which arises because drizzle drops straddle the gradual transition from the Stokes regime (appropriate for $r < 40\mu$m, where the terminal velocity increases as r^2) to the regime where the drag coefficient is independent of Reynolds number (appropriate for $r > 600\mu$m, where the terminal velocity increases as $r^{1/2}$). We therefore need to reconsider how to best evaluate (6) analytically.

To do so, we begin with the coalescence kernel $K(x, y)$, defined as

$$K(x, y) = \pi[r(x) + r(x')]^2 E(x, x')[w_T(x) - w_T(x')] \tag{12}$$

where $E(x, x')$ is the collection efficiency of the two drops of volume x and x', and w_T is the terminal velocity. Making the same assumptions as Long (1974), that $K(x, y) \approx \pi r^2 \tilde{E} w_T(r)$ where r is the radius of the collector drop and $\tilde{E}$ is a mean collection efficiency, (8) becomes

$$\dot{N} = -\pi N \int_0^\infty \tilde{E} r^2 w_T n(r) dr. \tag{13}$$

Now, $\tilde{E}$ represents the mean collection efficiency for drops that have the greatest impact upon $\dot{N}$. Implications from the evaluation of the SCE using observed drop size distributions (Wood 2005b) are that $\dot{N}$ is largely dominated by cloud droplets being captured by larger drizzle drops, i.e by accretion. For these collections, there is a weak, but approximately linear increase in $E(r, r')$ with collector drop radius r for collected drops in the range $5 < r' < 20$ μm and collector drops in the range $50 < r' < 200$ μm. Thus, to a reasonable approximation, we can assume that $\tilde{E} = E_0 r$, so that (13) becomes

$$\dot{N} \approx -\pi E_0 N \int_0^\infty r^3 w_T n(r) dr = -\frac{3}{4\rho_w} E_0 N P, \tag{14}$$

where P is the precipitation rate. In deriving the latter expression in (14) it was assumed that the precipitation is falling within still air. Figure 4 indicates that (14), with $E_0 = 4 \times 10^3$ m^{-1} (chosen to provide the best fit), is a much better bulk parameterization than either of the Long formulations taken separately, and is simply couched as the product of the cloud drop number concentration and the precipitation rate, both of which are parameters routinely estimated in large scale numerical models. We propose that (14) is a physically based and useful parameterization for CCN loss rates due to coalescence scavenging. An assessment of the importance of the loss rates is presented in the following section.

3. Application and discussion

We have derived a formulation for the rate of loss of cloud droplets by coalescence scavenging. This rate is equal to the loss rate of cloud condensation nuclei (CCN). Equation (14) is a local formulation, and so it is desirable and useful to construct a formula for the rate of CCN loss through coalescence for the cloud-containing layer as a whole. This could represent, for example, the marine boundary layer MBL, in which drizzling stratocumulus are confined. To do this, we integrate (14) over the layer depth. Assuming that the layer extends from the surface to the MBL inversion at height z_i, we obtain

$$\langle \dot{N} \rangle_{MBL} = -\frac{3E_0}{4\rho_w z_i} \int_0^{z_i} N(z) P(z) dz \tag{15}$$

where $\langle \ldots \rangle_{MBL}$ represents a layer average over the MBL. We next assume that $N(z) = 0$ for $z < z_{CB}$ where z_{CB} is the cloud base height, and that $N(z) = N_d$ is a constant in the cloud layer. This is a reasonable assumption supported by observations (e.g. Wood 2005a), and leads to

$$\langle \dot{N} \rangle_{MBL} = -\frac{3E_0 N_d h}{4\rho_w z_i} \langle P \rangle_{CLD} \tag{16}$$

with $\langle P \rangle_{CLD}$ being the mean precipitation rate in the cloud layer as a whole. In marine stratocumulus, for which the proposed parameterization will be most pertinent, recent observational work (Wood 2005a) has demonstrated that the precipitation rate tends to be roughly constant in the lowest two thirds of the cloud layer before decreasing rapidly above this. A reasonable expression for the precipitation profile in marine stratocumulus is $P(z) = P_{CB}(1 - z_*^3)$, where $z_* = (z - z_{CB})/h$, with $h = (z_i - z_{CB})$ being the cloud thickness. Substituting into (16) we obtain

$$\langle \dot{N} \rangle_{MBL} = -\frac{9E_0 h}{16\rho_w z_i} N_d P_{CB}. \tag{17}$$

Figure 5 shows the parameterized $\dot{N}$ from (17) as a function of the precipitation rate P and the cloud droplet concentration N, together with observational values of P and N from 12 aircraft flights. The aircraft data were taken in both drizzling and nondrizzling stratiform boundary layer clouds. A similar dependence of $\dot{N}$ upon P and N was found using bin-resolved microphysical large eddy simulations (Mechem et al. 2006).

It is important to note that even for relatively modest precipitation rates of 1 mm day^{-1}, values of $\dot{N}$ are approximately -100 cm^{-3} day^{-1} indicating that scavenging is likely to be a major term in the cloud condensation nucleus (CCN) budget in the boundary layer. Note that the timescale for complete removal of the CCN population assuming a constant precipitation rate, is $\tau = N/\dot{N} = 4\rho_w/(3E_0P)$, and so is inversely proportional to P.

Recent observations in subtropical marine stratocumulus (Pawlowska and Brenguier 2003; Comstock et al. 2004; Van Zanten et al. 2005) have found consistent relationships between the cloud base preciptiation rate P_{CB}, and cloud properties, such that P_{CB} increases sharply with cloud thickness (or liquid water path) and decreases with increasing cloud droplet concentration. Wood (2005b) presents additional observational evidence that the autoconversion rate is close to being inversely proportional to N_d. Adopting the formulation of (Van Zanten et al. 2005), which gives $P_{CB} = K_{VZ}h^3/N_d$, where $K_{vz} = 1.9\times$ 10^{-5} kg m^{-8} s^{-1}, we find that $\langle\dot{N}\rangle_{MBL}$ is independent of cloud droplet concentration and depends very strongly upon the cloud thickness h, such that

$$\langle\dot{N}\rangle_{MBL} = -\frac{9E_0K_{vz}}{16\rho_w z_i}h^4. \tag{18}$$

This is an interesting result and one that may have important implications for aerosol-cloud-drizzle feedbacks in the MBL. We should note that this expression compares favorably with box model calculations (not shown) driven by realistic trajectories in stratocumulus (see Fig. 9 of Feingold et al. 1996).

It is suggested that expressions of the form of (17) and/or (18) may be used to better couple aerosol process rates to meteorological processes in the boundary layer. This could be achieved using a simple mixed layer model framework which predicts the cloud thickness as a function of the large scale radiative and surface flux forcings on the system. Given the strong dependence of $\langle \dot{N} \rangle_{MBL}$ upon the cloud thickness h, in (18), it seems reasonable to hypothesize that it would be difficult to maintain a steady-state CCN population in boundary layers with very thick stratocumulus clouds. We leave this hypothesis for future investigation.

4. Conclusions

We have derived an analytical expression for the rate of loss of cloud droplets, and hence CCN, through coalescence scavenging. The expression, which depends only upon precipitation rate and cloud droplet concentration, compares well against our best attempt to constrain these loss rates using observational data. The expression can be used as a simple parameterization to express the rate at which CCN are removed by coalescence.

To demonstrate the potential application, we integrated the expression over the depth of the boundary layer (containing clear air below a cloud layer) to provide a formulation for the loss of CCN averaged over the depth of the boundary layer. Combining a recent expression relating the precipitation rate to the cloud thickness and droplet concentration with the integrated expression leads to the conclusion that the loss rate is almost independent of cloud droplet concentration and is determined primarily by cloud thickness, upon which a very strong dependence is found. The expressions presented here may be useful for incorporation into models of the aerosol budget under cloudy conditions.

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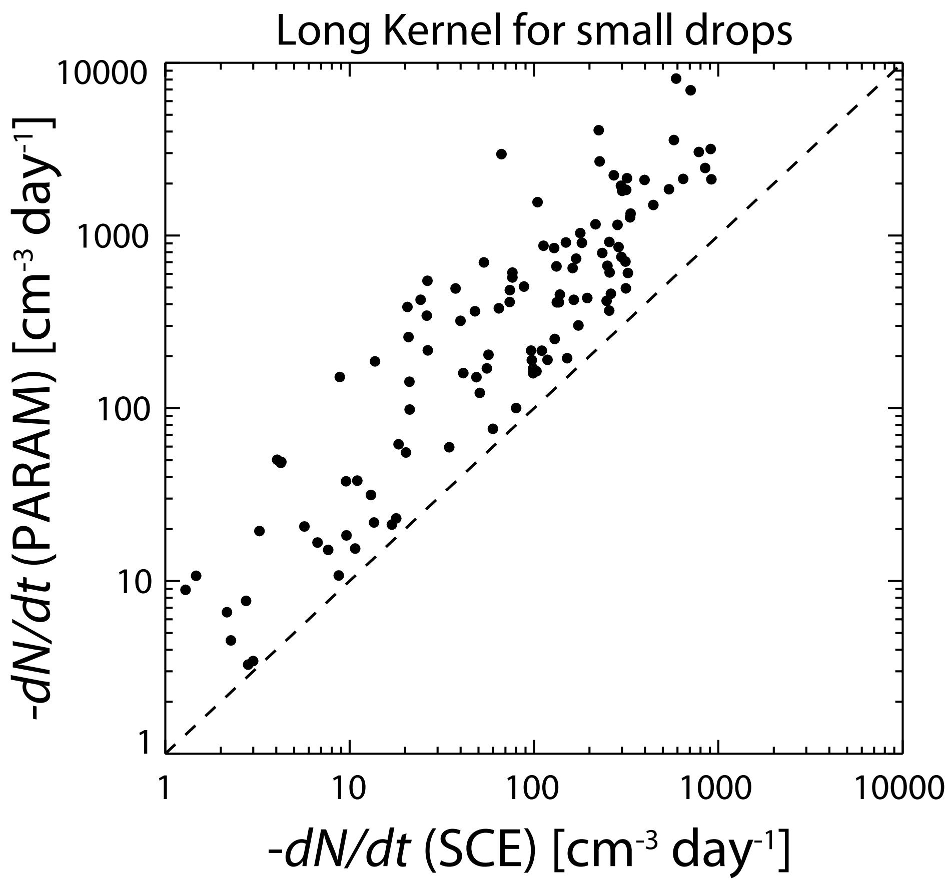
Figure 1: Drop coalescence scavenging rates estimated by integration of the SCE in the form (6) using observed size distributions in stratiform boundary layer clouds Wood (2005b) against the parameterization based upon the Long (1974) analytic kernel approximation for small drops, i.e. Eqn. (10). The SCE integration uses the collection kernel of Hall (1980).

Figure 2: As Fig 1 except that only the collection of cloud droplets ($r < 20\ \mu$m) by other cloud droplets is considered. Inset shows comparison of the SCE derived rates for $r < 20\ \mu$m only (ordinate) against those including both cloud and drizzle drops (abscissa), indicating that for many cases in stratocumulus, the collection of cloud droplets by drizzle droplets represents a major contribution to the loss rates.

Figure 3: As Fig 1 except for the Long (1974) kernel for large drops, i.e. Eqn. (11).

Figure 4: As Fig 1 except using the new parameterization of Eqn. (14).

Figure 5: Parameterized MBL mean drop coalescence scavenging rates (in cm^{-3} day^{-1}) plotted as a function of the cloud base precipitation rate P_{CB} and the mean cloud droplet concentration N_d, estimated using (17) assuming $h/z_i = 0.4$ (dashed contours). Also plotted are values of P_{CB} and N_d from aircraft flights and other observations in stratiform boundary layer clouds around the globe (see Wood (2005a) for details).



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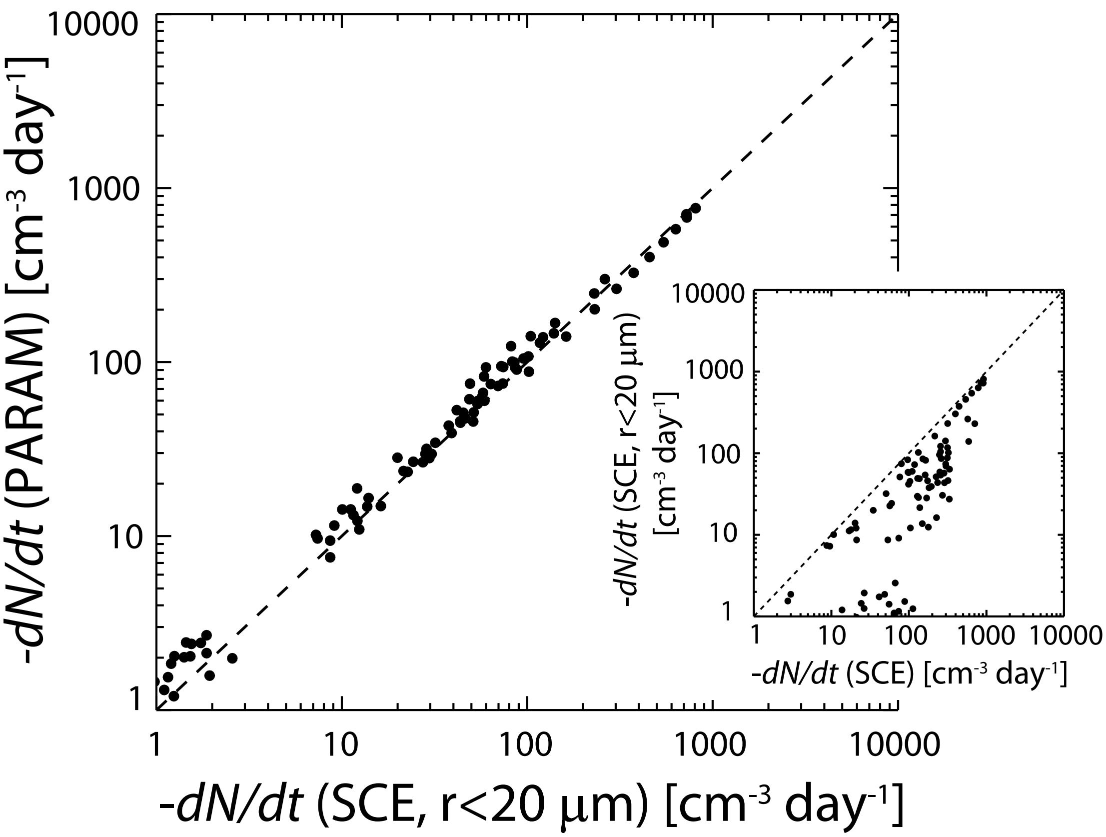

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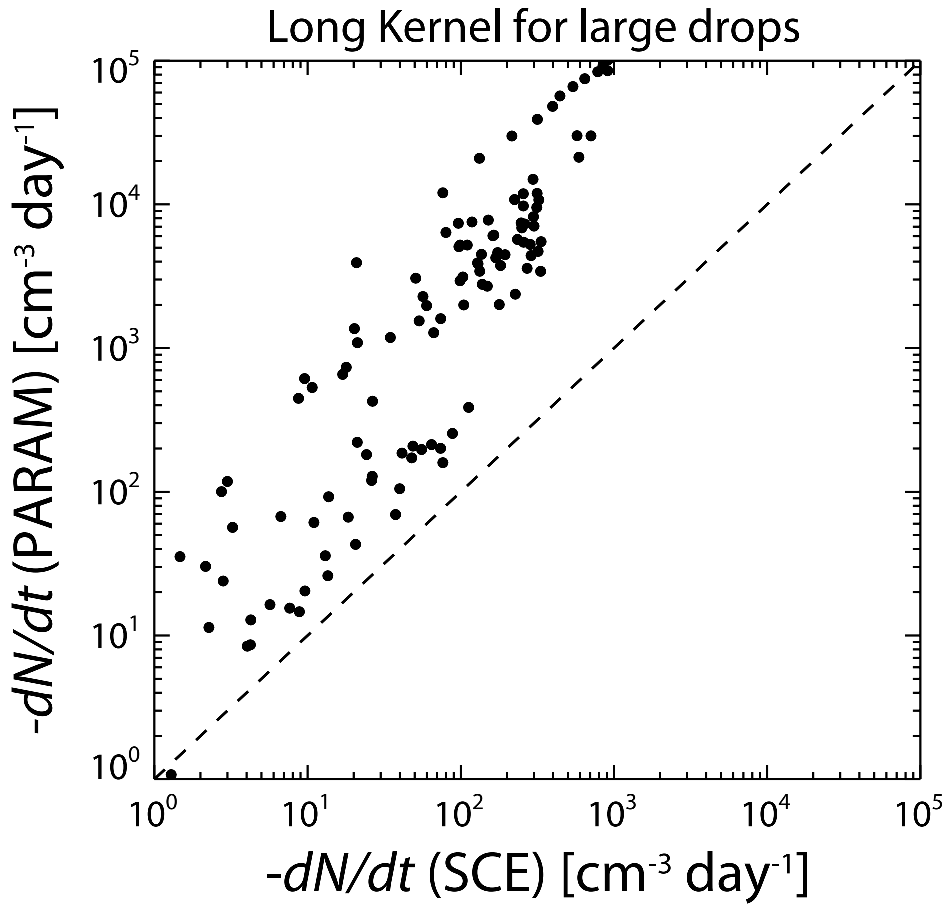

Figure 3: As Fig 1 except for the Long (1974) kernel for large drops, i.e. Eqn. (11).

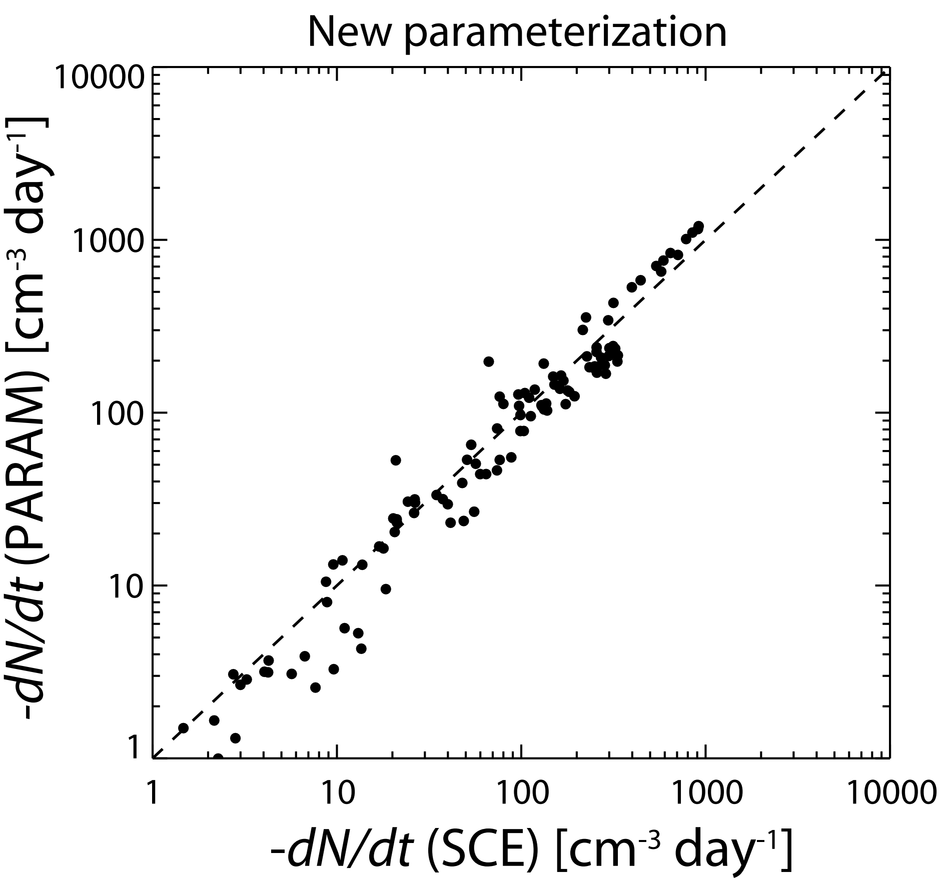

Figure 4: As Fig 1 except using the new parameterization of Equation 14.

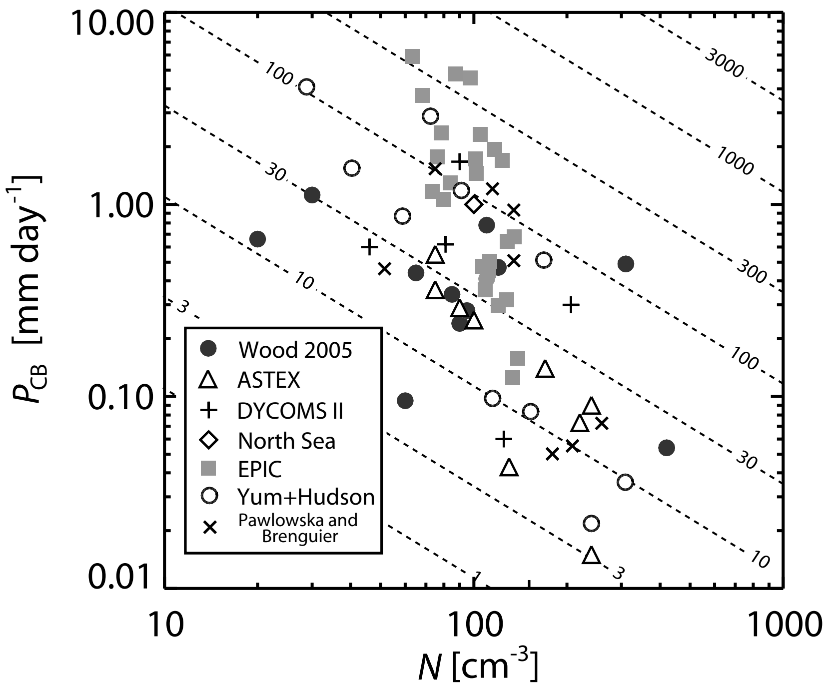

Figure 5: Parameterized MBL mean drop coalescence scavenging rates (in cm^{-3} day^{-1}) plotted as a function of the cloud base precipitation rate P_{CB} and the mean cloud droplet concentration N_d, estimated using (17) assuming $h/z_i = 0.4$ (dashed contours). Also plotted are values of P_{CB} and N from aircraft flights and other observations in stratiform boundary layer clouds around the globe (see Wood (2005a) for details).