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Rate of loss of cloud droplets by coalescence in warm clouds

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[1] 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 surprisingly 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.

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1. Introduction

[2] 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 the marine boundary layer, as we shall see, the main loss mechanism for cloud condensation nuclei (CCN) is through the process of cloud and drizzle drops coalescing with each other, a process we term coalescence scavenging. Each coalescence event effectively removes a single CCN from the MBL as the resulting drop, once evaporated, will result in only one aqueous haze particle. Collection of interstitial or subcloud aerosols by precipitating drops represents a much weaker sink of aerosols and can essentially be neglected.

[3] In this study, an analytic expression for the coalescence scavenging of cloud droplets 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. We also compare our formulation with existing treatments of coalescence scavenging droplet loss rates.

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2. Analytic Expression

[4] We consider an expression for the rate of loss of droplets through coalescence. Given that each coalescence event reduces the number of CCN by one, this can be interpreted as being a loss rate for the CCN. 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]

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

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

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

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

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

Using (1) we obtain

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

[5] Exchanging the order of integration in both terms and making the substitutions $y = x - x'$ in the inner integral of

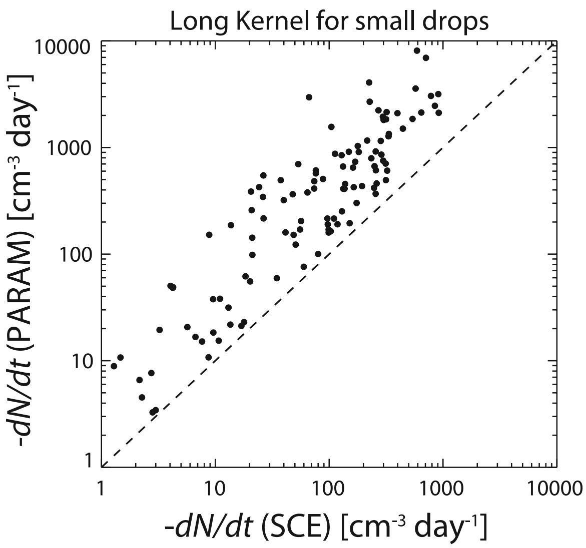

Figure 1. Drop coalescence scavenging rates estimated by integration of the SCE in the form (6) using observed size distributions in stratiform boundary layer clouds [*Wood*, 2005b] against the parameterization based upon the *Long* [1974] analytic kernel approximation for small drops, i.e., equation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

the first term and $y = x$ in the inner integral of the second, we find

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

which can be simplified to

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

[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

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

[7] Application of (7) to (6) gives

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

Following *Long* [1974] for small drops ($r < 50$ μm), 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 $\kappa = 1.1 \times 10^{10}$ m^{-3} s^{-1}, we obtain

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

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

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

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 ($r > 20$ μm), the Long small drop parameterization is insufficient.

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

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

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 $r < 40$ μm, where the terminal velocity increases as r^2) to the regime where the drag coefficient is independent of Reynolds number (appropriate for $r > 600$ μm, where the terminal velocity increases as $r^{1/2}$). We therefore need to reconsider how to best evaluate (6) analytically.

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

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

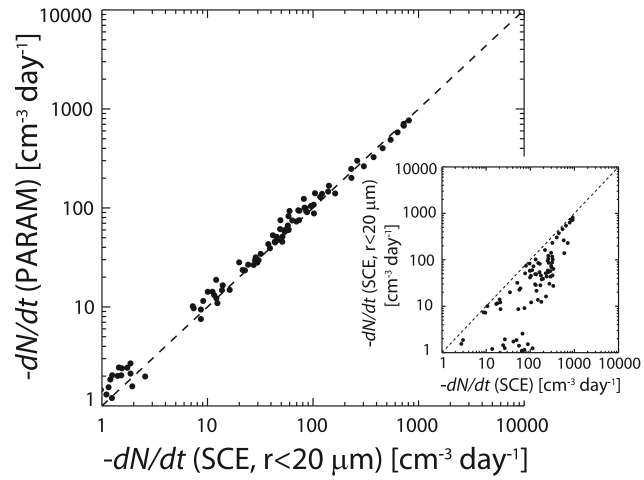

Figure 2. As Figure 1 except that only the collection of cloud droplets (r < 20 μm) by other cloud droplets is considered. Inset shows comparison of the SCE derived rates for r < 20 μm only (ordinate) against those including both cloud and drizzle drops (abscissa), indicating that for many cases in stratocumulus,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

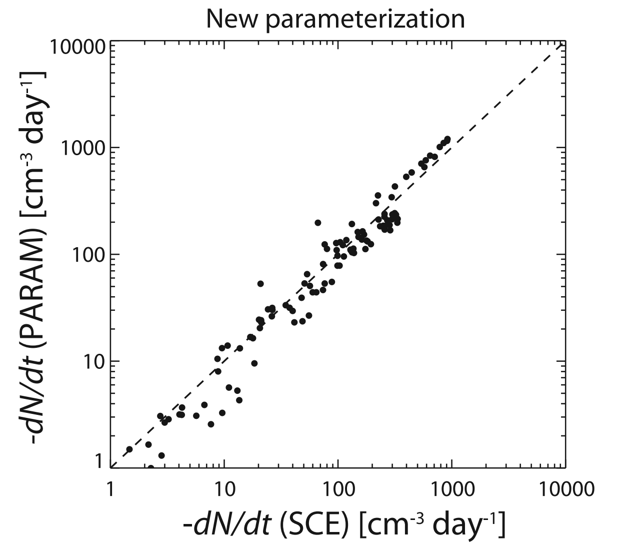

Figure 4. As Figure 1 except using the new parameterization of equation (14).

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.

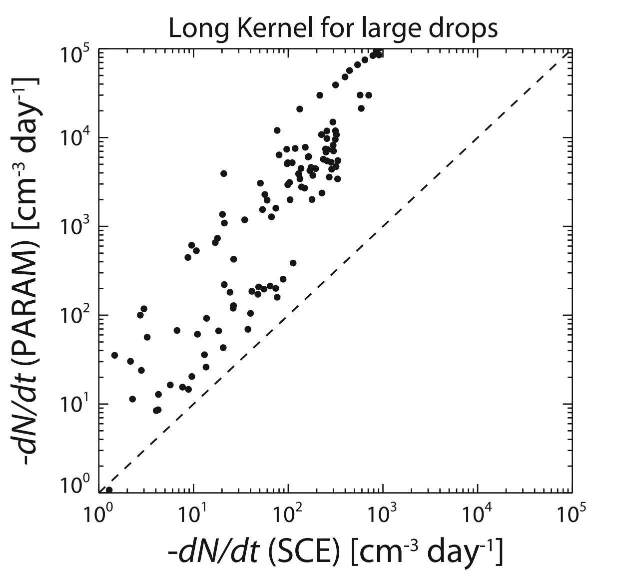

Figure 3. As Figure 1 except for the *Long* [1974] kernel for large drops, i.e., equation (11).

3. Application and Discussion

[10] 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 we discuss it in context of the existing literature on scavenging in the following section. First, we apply the formulation and construct a novel 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

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

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

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

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

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

[11] 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].

[12] 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.

[13] 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 precipitation 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

$$\langle \dot{N} \rangle_{MBL} = -\frac{9E_0 K_{vz}}{16\rho_w z_i} h^4. \quad (18)$$

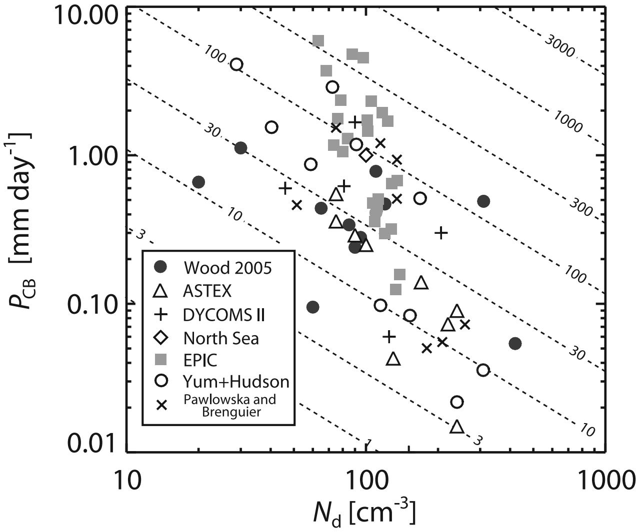

Figure 5. Parameterized MBL mean drop coalescence scavenging rates (in cm^{-3} day^{-1}) plotted as a function of the cloud base precipitation rate P_{CB} and the mean cloud droplet concentration N_d, estimated using (17) assuming $h/z_i = 0.4$ (dashed contours). Also plotted are values of P_{CB} and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

This is a novel and interesting result and one that is not readily apparent until the dependence of precipitation upon the cloud thickness and droplet concentration is introduced. It may have important implications for aerosol-cloud-drizzle feedbacks in the MBL, and it is suggested that expressions of the form of (17) and/or (18) may be used to more succinctly couple aerosol process rates to meteorological processes in heuristic models of the boundary layer such as those of *Baker* [1993]. 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 could be difficult to maintain a steady state CCN population in boundary layers with very thick stratocumulus clouds. Indeed, the high CCN loss rates imply a strongly negative feedback on aerosol (and cloud droplet) concentrations in the planetary boundary layer. We leave this hypothesis for future investigation.

4. Comparison With Existing Studies and Formulations

[14] Both *Beheng* [1994] and *Khairoutdinov and Kogan* [2000] provide expressions for the rate of loss of cloud droplets through coalescence scavenging. Detailed comparisons of these rates with rates estimated from aircraft observations of the drop size distributions were presented by *Wood* [2005b] and will not be repeated here. Because the analytic formulation derived above proceeds directly and naturally to an expression depending upon precipitation rate, i.e., equation (14), it is not directly comparable with the rates presented by *Beheng* [1994] and *Khairoutdinov and Kogan* [2000] which are expressed as functions of the liquid water content in the cloud and drizzle modes, in

addition to the cloud droplet concentration. In general, the Beheng rates tend to be roughly a factor of two larger than those from Khairoutdinov and Kogan, with the latter being in better agreement with the aircraft-estimated rates. The Khairoutdinov and Kogan formulation can be reduced to $\dot{N}/N = \dot{q}_l/q_l$, where q_l is the cloud liquid water mixing ratio; that is, the fractional loss of cloud droplets is equal to the fractional loss of cloud liquid water content. Aircraft measurements indicate that as with the drizzle mass production rates, cloud droplet loss rates tend to be dominated by the accretion process, i.e., drizzle drops ($r > 20$ μm) collecting the smaller cloud drops *Wood* [2005a]. With this assumption, and with the mass accretion rates in all the parameterizations being more or less proportional to the product of the liquid water contents in the cloud and the drizzle mode, this leads to a form for Khairoutdinov and Kogan (and also Beheng) whereby $\dot{N}/N$ depends linearly upon only the drizzle mass mixing ratio. Because the precipitation rate is likely to scale with the drizzle mass, this indicates that the *Beheng* [1994] and *Khairoutdinov and Kogan* [2000] coalescence scavenging expressions are to first-order functionally similar to equation (14) in their dependence upon the characteristics of the precipitation.

[15] It is important to draw the distinction between coalescence scavenging and the precipitation scavenging that has received more attention in the literature [e.g., *Seinfeld and Pandis*, 1996]. The latter involves the scavenging of either interstitial or subcloud unactivated aerosol by falling precipitation, and might be expected to have a different dependency upon the bulk properties of the aerosol and precipitation because of the huge disparity in the collector and collectee size. Much of the literature on precipitation scavenging [see *Pruppacher and Klett*, 1997] has focused upon mass scavenging rates of particles of a micrometer and smaller, where Brownian motion of the collected particle starts to become important. Precipitation scavenging is likely to be the dominant mechanism for aerosol removal in strong precipitation events. However, some studies have specifically examined coalescence scavenging in the lighter precipitation that falls from warm boundary layer clouds [*Feingold et al.*, 1996; *Zhang et al.*, 2004; *Mechem et al.*, 2006] and concluded that this even light precipitation can have a marked impact upon the aerosol concentration. If we make the assumption that the clouds are adiabatic, then it is possible to transform (18) into an expression that depends upon the cloud-mean liquid water content that can be qualitatively compared with the box model calculations of *Feingold et al.* [1996]. These calculations permit a more realistic assessment of the scavenging rates that allows for the limited in-cloud residence time of parcels. This was achieved using in-cloud residence time probability distributions from stratocumulus large eddy model trajectories and a lognormal cloud droplet size distribution with an assumed geometric standard deviation. The scavenging rates thus derived are then a function of the liquid water content, the initial droplet concentration, and the residence time distribution. Similarities between (18) and the results of *Feingold et al.* [1996] are that the depletion rate $\langle \dot{N} \rangle_{MBL}$ is a strongly convex function of liquid water content and is almost independent of the cloud droplet concentration (note that in Figure 9 of *Feingold et al.* [1996] the scavenging is given as fractional rates). These similarities lend additional credence to the functional dependence of the parameterization presented here.

5. Conclusions

[16] 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.

[17] 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.

[18] **Acknowledgments.** I thank Dave Leon, Jefferson Snider, Peter Blossey, Dave Mechem, and Graham Feingold for helpful discussions and two anonymous reviewers for suggestions that have improved the manuscript. I wish to thank the staff of the Meteorological Research Flight and the C-130 aircrew and groundcrew for their dedication to collecting the data presented in this study. This work was supported by NSF grant ATM 0433712.

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